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(54) WASTE TREATMENT CHEMICAL AND WASTE DISPOSAL METHOD

(57) A waste-treating material wherein an effect for preventing elution of harmful metals by the action of a water glass aqueous solution is improved by adding additives of additive A (acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters or dialdehydes), additive B (phosphates, carbonates, sulfates, carboxylates or hydroxides of potassium, sodium or ammonium, phosphonates of potassium or sodium, or organic chelating agents having carboxyl group (-COOX, wherein X represents hydrogen, potassium or sodium) or dithiocarbamic acid group (>NCSSY, wherein Y represents hydrogen, potassium or sodium)), additive C (coagulating sedimentators), additive D (monovalent metal salts or ammonium salts, which do not produce compounds which are insoluble or sparingly soluble in water by reacting with calcium ions), and the like. In particular, the waste-treating material can prevent re-elution of Pb in fly ash discharged from refuse incinerating facility. Further, one part stability of the treating liquid is improved by adding additive E (gluconates, tartarates, benzoates, ligninsulfonates, polysaccharides, or bases comprising monovalent cations and hydroxide ions) and

the like, resulting in excellent convenience.

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DescriptionBACKGROUND OF THE INVENTION

1. Field of the Invention

The present invention relates to a waste-treating material and a method for treating waste, which are effective for stabilizing harmful metals contained in the waste which contains a calcium compound such as calcium hydroxide, calcium oxide or calcium chloride. More particularly, the invention relates to a waste-treating material and a method for treating waste, which are effective for stabilization treating harmful metals such as Pb in waste incineration fly ash which contains a calcium compound such as calcium hydroxide, calcium oxide or calcium chloride, and is difficult to suppress elution of lead (Pb).

2. Description of the Related Art.

At present in treating an industrial waste containing harmful metals and the like, a method for stabilization is used wherein a cement is used as a treating material, the cement and the waste are mixed, water is added to the mixture and kneaded, and the resulting kneaded mixture is aged and solidified, thereby preventing elution of harmful metals. However, the conventional treatment method of industrial waste of simply solidifying the same with the cement involves various problems, and unless the purpose of use is limited, secondary pollution may possibly induce. In particular, in an incinerator or the like of municipal refuse, slaked lime is blown for the purpose of suppressing an amount of hydrogen chloride gas generated during operation, resulting in forming an alkali atmosphere. However, it is generally known that Pb is easily eluted under an alkali atmosphere. For this reason, despite that in incinerating refuse, harmful metals such as Pb are contained in fly ash captured by an electric collector or a bag filter in a high concentration, elution of Pb cannot sufficiently be prevented by the cement treatment which is the conventional technique. At present the fly ash is discarded with insufficient stabilization of harmful metals and the like, and there arises a lot of problems on secondary pollution after treatment.

In this time of day, it becomes apparent in the country and abroad that it is difficult to stabilize the industrial waste containing harmful metals in the state such that the harmful metals do not elute, by simply solidifying the same with the cement. Therefore, there has been demanded a waste-treating material and a treatment method, wherein the harmful metals are surely sealed at time of landfill reclamation or ocean dumping, so that harmful metals do not elute again and secondary pollution does not occur.

To such a subject, JP-A-53-6270 discloses a method of treating waste using a water glass and hydrogen carbonate in treatment of lime cake discharged from sugar refining or sugar refining factory. However, since the hydrogen carbonate acts to water glass as an acid, if a mixture of those is prepared before use, water glass gels, and it cannot be used as one part treating material. Thus, there is a big problem on convenience.

In view of the present stage of such a waste treatment, an object of the present invention is to provide a waste-treating material and a method for treating waste, which can stabilize various harmful metals contained in the waste containing calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride so as not to re-elute those by surely solidifying those.

SUMMARY OF THE INVENTION

The present inventors have made extensive investigations to overcome the problems involved in the prior art and have developed a method for treating waste using a water glass. This method is a method for preventing elution of harmful metals in waste by mixing the waste containing calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride with a water glass. As a result of further investigations on the above method for treating waste, the present invention has reached to obtain a new method for treating waste which can achieve the above object more surely.

A first waste-treating material according to the present invention in order to achieve the above object comprises a water glass aqueous solution and at least one selected from the group consisting of the following additive A and additive B, as main structural components. The additive A herein is at least one selected from the group consisting of acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters and dialdehydes. The additive B herein is at least one selected from the group consisting of materials which form calcium compounds which are insoluble or sparingly soluble in water by reacting with calcium ions, and ion sealing agents which seal calcium ions.

A second waste-treating material according to the present invention comprises a treating material comprising a water glass aqueous solution as the main structural component, and an additive C comprising a coagulating sedimentator, added to the treating material.

A third waste-treating material according to the present invention comprises a treating material comprising a water glass aqueous solution as the main structural component, and an additive D comprising at least one selected from the group consisting of monovalent metal salts and ammonium salts, which do not produce calcium compounds which are insoluble or sparingly soluble in water by reacting with calcium ions, added to the treating material.

The method for treating waste according to the present invention comprises kneading the waste and the above-described waste-treating material, and aging the resulting mixture.

The water glass aqueous solution used in the present invention may be a glass water aqueous solution for general purpose. Examples of an alkali component of the water glass include Na, K and ammonia. Of those, Na, i.e., sodium silicate ($\text{Na}_2\text{O} \cdot n\text{SiO}_2$) is preferred from the viewpoints of stabilization performance of harmful heavy metals and the like, industrial availability, cost, and the like. The water glass aqueous solution is represented by $\text{M}_2\text{O} \cdot n\text{SiO}_2$, and n ($\text{SiO}_2/\text{M}_2\text{O}$ compositional ratio) is in the range of about 0.5 to 4.2 in the commercially available product. When n is 2.0 or more, the performance for preventing elution of harmful heavy metals is excellent. On the other hand, if n is less than 2.0, stability in an aqueous solution is poor, and solid component in an water glass aqueous solution may precipitate depending on the storage condition at a low temperature in the season of winter. From this, it is generally preferred to use a water glass aqueous solution defined by JIS (Japanese Industrial Standard). Further, there is no problem even if the glass water aqueous solution used in the present invention contains unavoidable impurities such as iron.

As described above, it is generally known that Pb in the waste incineration ash is liable to elute under an alkali atmosphere. However, in the incinerator or the like of municipal refuse, since slaked lime is blown for the purpose of suppressing the amount of hydrogen chloride gas generated during the operation, an electric collector trapped fly ash or a bag filter trapped fly ash produced under such an operation state particularly has an increased elution amount of Pb, and the elution cannot sufficiently be prevented by the prior art technique. Contrary to this, the waste-treating material and the method for treating waste according to the present invention show high elution preventing effect against harmful metals. This action mechanism is not always clear, but it is assumed as follows. It is considered that the performance for stabilizing harmful metals in the water glass aqueous solution is developed by sealing of harmful metals, adsorption of harmful metals on a gel produced, and the like by a gelation reaction of calcium ions and the like eluted from the waste with polyvalent metal ions, but depending on the amount of polyvalent metal ions such as calcium ions, the effect may sufficiently develop or may not sufficiently develop. This is considered to be that the gel produced by the reaction of the water glass aqueous solution with calcium ions takes a different form depending on the amount of calcium ions, and the difference in the form affects sealing and adsorption actions of harmful metals. In particular, if the amount of calcium ions is too large as compared with the amount of the water glass aqueous solution, a gel in the form which decreases sealing and adsorption abilities is produced, and the performance for stabilizing harmful metals does not sufficiently develop. In other words, clear numerical values are not known, but in the relationship in the amount between the water glass aqueous solution and the calcium ions, it is considered that the optimum value exists in the performance for stabilizing harmful metals.

The additive A added to the water glass aqueous solution in the treating material is that acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters, dialdehydes, and the like are reacted with the water glass aqueous solution, and solid component monomers in the water glass aqueous solution are partially polymerized, so that the water glass solid component is polymerized. The polymerized water glass gels by merely reacting with a slight amount of calcium compounds, and further solidifies by aging. As a result, reaction rate between water glass and calcium compounds in the waste is increased, and the action for sealing and solidifying harmful metals such as Pb is accelerated.

The additive B in the treating material reacts with calcium ions to form the calcium compounds which are insoluble or sparingly soluble in water, or to seal the calcium ions, thereby the relationship in the amount between the water glass aqueous solution and the calcium ions can be made optimum. Although described above, in the incinerator or the like of municipal refuse, since slaked lime is blown for the purpose of suppressing the amount of hydrogen chloride generated during the operation, a considerably large amount of calcium compounds are contained in the fly ash collected by an electric collector or a bag filter, and this becomes a source of a large amount of calcium ions. Therefore, the additive B is added to the waste-treating material to appropriately control so as to decrease the amount of calcium ions, and this enables the performance for stabilizing harmful metals by the water glass aqueous solution to markedly improve.

It is considered in the second treating material that by adding the additive C comprising a coagulating sedimentator to the treating material comprising the water glass aqueous solution as the main structural component, fine particles which constitute the waste are coagulated to decrease a contact area with water, making it difficult to elute harmful heavy metals, and also since a gel of the water glass, produced by the reaction with polyvalent metal ions such as calcium ions, includes and seals the coagulated particles, the effect for preventing elution of heavy metals is further improved. It is further considered that even if heavy metals are eluted, the produced water glass gel adsorbs eluted ions, thereby preventing reelution.

Furthermore, as described before, it is considered that the performance for stabilizing harmful metals of the water glass aqueous solution is developed by sealing of harmful metals involved in gelation reaction of the water glass aqueous solution with polyvalent metal ions such as calcium ions eluted from the waste. In order to sufficiently exhibit the Pb

stabilizing performance of the water glass aqueous solution by this action, it is necessary that the gelation reaction of the water glass aqueous solution proceeds faster than the elution of harmful metals, thereby sealing the harmful metals.

The third treating material of the present invention comprises a treating material comprising a water glass aqueous solution as the main structural component, and the additive D comprising at least one of monovalent metal salts and ammonium salts, added to the treating material. Therefore, ion concentration in the aqueous solution of the treating material increases by monovalent metal ions or ammonium ions released from the additive D, and by the salting out effect, the solid component of the water glass aqueous solution is liable to precipitate. As a result, it is considered that a gel production rate when the water glass in the treating material aqueous solution reacts with polyvalent metal ions such as calcium ions in fly ash becomes fast, and the performance for stabilizing Pb is improved. However, if the amount of the additive D added is too large, ion concentration in the treating material approaches its saturation, and solid component in the water glass aqueous solution precipitates by the salting out effect of co-existent ions in the treating material aqueous solution. As a result, it is impossible to seal harmful metals in the waste. For the reasons above, specific attention should be paid to storage conditions (storage form, storage place, storage time, and the like) of the treating material aqueous solution, and kind and amount of the additive D used, and it is necessary to determine optimum values on materials appropriately selected.

DETAILED DESCRIPTION OF THE INVENTION

The waste-treating material of the present invention is described in more detail below.

In the first waste-treating material of the present invention, if the acid is added in an amount such that hydrogen ion (H^+) released from the acid exceeds 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution (where water glass is sodium silicate, cation is replaced by sodium, and hereinafter the same), its pH reaches a neutral region. As a result, the treating material may gel instantaneously or after several minutes. The treating material gelled can be expected to be a certain degree of the performance for stabilizing Pb, but such a performance is not sufficient. Further, the gelled treating material is in the form which cannot be used as a liquid treating material, and this may possibly induce undesirable problems such as clogging of pipings of an injection device for the treating material, a kneader of the waste and the treating material, and the like. For this reason, the amount of the acid added is preferably such that the hydrogen ion (H^+) in the acid is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

Any acid can be used as the acid so long as it ionizes in water or the water glass aqueous solution to release hydrogen ion (H^+). Examples of the acid which can be used include hydrochloric acid (HCl), nitric acid (HNO_3), phosphinic acid (HPH_2O_2), phosphonic acid (H_2PHO_2), phosphoric acid (H_3PO_4), diphosphoric acid ($H_4P_2O_7$), tripolyphosphoric acid ($H_5P_3O_{10}$), sulfurous acid (H_2SO_3), sulfuric acid (H_2SO_4), carbonic acid (H_2CO_3), monochloroacetic acid ($CH_2ClCOOH$), dichloroacetic acid ($CHCl_2COOH$), trichloroacetic acid (CCl_3COOH), acetic acid (CH_3COOH), citric acid ($(OH)C_3H_4(COOH)_3 \cdot H_2O$), adipic acid ($(CH_2)_4(COOH)_2$), salicylic acid ($C_6H_4(OH)(COOH)$), oxalic acid ($(COOH)_2$) and phenol (C_6H_5OH). Of those, hydrochloric acid, nitric acid, phosphoric acid, sulfuric acid, carbonic acid, acetic acid and oxalic acid are preferred, considering industrial availability, the cost, the performance for stabilizing Pb, and the like. If necessary, two or more of different kinds of acids can be used together.

In the first treating material, if the polyvalent metal salt is added in an amount such that the product of the part by mol of polyvalent metal ions in the polyvalent metal salts and its number of valency exceeds 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution, it is an amount such that metal ions make the treating material gel instantaneously or after several minutes. The gelled treating material may have the same disadvantages as described before, and is undesirable. From this reason, the amount of the polyvalent metal salts added is preferably such that the product of the part by mol of polyvalent metal ions of the polyvalent metal salts and its number of valency is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

Examples of the polyvalent metal salts include chlorides, nitrates, sulfates, carbonates and phosphates of magnesium, calcium, strontium, barium, iron, aluminum and zinc. Specific examples thereof include magnesium chloride ($MgCl_2$), calcium chloride ($CaCl_2$), strontium chloride ($SrCl_2$), barium chloride ($BaCl_2$), iron (II) chloride ($FeCl_2$), iron (III) chloride ($FeCl_3$), zinc chloride ($ZnCl_2$), aluminum chloride ($AlCl_3$), magnesium nitrate ($Mg(NO_3)_2$), calcium nitrate ($Ca(NO_3)_2$), strontium nitrate ($Sr(NO_3)_2$), barium nitrate ($Ba(NO_3)_2$), iron (II) nitrate ($Fe(NO_3)_2$), iron (III) nitrate ($Fe(NO_3)_3$), aluminum nitrate ($Al(NO_3)_3$), magnesium sulfate ($MgSO_4$), calcium sulfate ($CaSO_4$), strontium sulfate ($SrSO_4$), barium sulfate ($BaSO_4$), iron (II) sulfate ($FeSO_4$), zinc sulfate ($ZnSO_4$), aluminum sulfate ($Al_2(SO_4)_3$), potassium aluminum sulfate ($KAl(SO_4)_2 \cdot 12H_2O$), zinc carbonate ($ZnCO_3$), magnesium carbonate ($MgCO_3$), calcium carbonate ($CaCO_3$), magnesium phosphate ($Mg_3(PO_4)_2$), magnesium hydrogen phosphate ($MgHPO_4$), magnesium dihydrogen phosphate ($Mg(H_2PO_4)_2$), calcium phosphate ($Ca_3(PO_4)_2$), calcium hydrogen phosphate ($CaHPO_4$), calcium dihydrogen phosphate ($Ca(H_2PO_4)_2$), and calcium superphosphate ($CaH_4(PO_4)_2 \cdot H_2O$). Further, in order that the treating material comprising the water glass and the polyvalent metal salts, as the main structural components exhibits the performance for stabilizing the desired harmful metals, it is necessary that the polyvalent metal salts rapidly dissolve in water or the water glass aqueous solution to react with water glass, and the solid component of the water glass is

polymerized. From this fact, of the above compounds, considering solubility of the polyvalent metal salts in the water glass aqueous solution and industrial availability, the preferred polyvalent metal salts are magnesium chloride, calcium chloride, iron (II) chloride, iron (III) chloride, aluminum chloride, magnesium nitrate, calcium nitrate, iron (II) nitrate, iron (III) nitrate, aluminum nitrate, magnesium sulfate, iron (II) sulfate and aluminum sulfate. If necessary, those polyvalent metal salts can be used as mixtures of two or more thereof.

The alcohols, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters and dialdehydes, contained as the additive A in the treating material according to the present invention can be general compounds. Examples of the alcohols include methanol (CH_3OH) and ethanol ($\text{C}_2\text{H}_5\text{OH}$). Examples of the polyhydric alcohol esters include ethylene glycol diacetate ($\text{C}_2\text{H}_4(\text{OCOCH}_3)_2$) and triacetylene ($\text{C}_3\text{H}_5(\text{OCOCH}_3)_3$). Examples of the carbonates include ethylene carbonate ($(\text{CHO}_2)_2\text{CO}$). Examples of the intramolecular esters include gamma-butyrolactone ($\text{C}_4\text{H}_6\text{O}_2$). Examples of the dibasic acid esters include succinic acid dimethyl ester ($\text{CH}_3\text{OOC}(\text{CH}_2)_2\text{COOCH}_3$) and glutaric acid dimethyl ester ($\text{CH}_3\text{OOC}(\text{CH}_2)_3\text{COOCH}_3$). Examples of dialdehydes include glyoxal ($(\text{CHO})_2$). Besides the above, compounds which dissolve in water or the water glass aqueous solution and react with the water glass aqueous solution, whereby the solid component of the water glass is polymerized, can also be used. The alcohols, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters and dialdehydes each can be used as mixtures of two or more thereof. The amount of the additive A added can be optionally selected, but it is preferred to select the amount that the treating material has less possibility to gel when the water glass aqueous solution and the additive A are mixed. As described before, although the gelled treating material is expected to have a certain degree of the performance for stabilizing Pb, the performance is not sufficient. This may induce undesirably problems of clogging of pipes of an injection device for the treatment, a kneader of the waste and the treating material, and the like.

Further, if necessary, it is possible in the first treating material to add as the additive A at least two kinds selected from the acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters, and dialdehydes to the water glass aqueous solution.

In the first treatment material of the present invention, the amount of the additive B added is 10 to 400 parts by weight per 100 parts by weight of the solid content of water glass (the content of silicic acid salt in water glass aqueous solution; i.e., total weight of M_2O content and SiO_2 content, wherein M is cation in the water glass aqueous solution; hereinafter the same) in the water glass aqueous solution. If the amount of additive B is less than 10 parts by weight, insoluble amount or sealing amount of calcium ions is small, and the desired performance is not exhibited. On the other hand, if the amount of the additive B exceeds 400 parts by weight, the performance for preventing elution of lead may rather be decreased. The reason of this is not clear, but it is considered that the amount of sodium ion or potassium ion, which is not common to lead ion, is increased, and also elution of lead ion is increased due to increase of ion strength. The amount of the additive B added in the treating material of the present invention varies depending on concentration of water glass aqueous solution, compositions of water glass aqueous solution, amount of water glass aqueous solution added to waste, addition method of the additive B, Pb content in the waste, calcium compound content, elution amount of harmful heavy metals from waste when not treated, allowable elution amount of harmful heavy metals to be a target, and the like. Practically, the amount is determined by the blending ratio of the water glass aqueous solution and the additive B so as to have the desired performance for stabilizing harmful heavy metals and to make the cost most inexpensive. In general, the additive B is preferably used in an amount of 15 to 280 parts by weight per 100 parts by weight of water glass solid content in the water glass aqueous solution, so that good performance for preventing elution of harmful heavy metals can be obtained.

Examples of the material which reacts with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water, used as the additive B in the treating material of the present invention include tripotassium phosphate, trisodium phosphate, trilithium phosphate, triammonium phosphate, dipotassium hydrogen phosphate, disodium hydrogen phosphate, diammonium hydrogen phosphate, potassium dihydrogen phosphate, sodium dihydrogen phosphate, ammonium dihydrogen phosphate, tetrapotassium diphosphate (potassium pyrophosphate), tetrasodium diphosphate (sodium pyrophosphate), tenpotassium triphosphate (potassium tripolyphosphate), pentasodium triphosphate (sodium tripolyphosphate), hexapotassium tetraphosphate (potassium tetrapolyphosphate), hexasodium tetraphosphate (sodium tetrapolyphosphate), potassium trimetaphosphate, sodium trimetaphosphate, potassium hexametaphosphate, sodium hexametaphosphate, potassium carbonate, sodium carbonate, sodium potassium carbonate, lithium carbonate, ammonium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate, ammonium hydrogen carbonate, potassium sulfate, sodium sulfate, lithium sulfate, ammonium sulfate, potassium hydrogen sulfate, sodium hydrogen sulfate, ammonium hydrogen sulfate, potassium oxalate, sodium oxalate, lithium oxalate, ammonium oxalate, potassium hydrogen oxalate, sodium hydrogen oxalate, ammonium hydrogen oxalate, sodium stearate, potassium stearate, and lithium stearate. Phosphates, carbonates, sulfates, carboxylates or hydroxides of potassium, sodium or ammonium are preferred, considering industrial availability, cost, Pb stabilizing performance, and the like. Of those, it is preferred to use any of tripotassium phosphate, pentasodium triphosphate (sodium tripolyphosphate), hexasodium tetraphosphate (sodium tetrapolyphosphate), sodium hexametaphosphate, potassium carbonate, sodium carbonate, sodium hydroxide, potassium hydroxide, ammonium sulfate and potassium oxalate, from the standpoint of good Pb stabilizing performance.

It is more preferred to use carbonates, particularly potassium carbonate and sodium carbonate, or hydroxides of monovalent alkali metals, particularly sodium hydroxide, as the materials which react with calcium ions of the additive B to produce calcium compounds which are insoluble or sparingly soluble in water, from the standpoints of Pb elution preventing performance of the treating material and industrial availability. By containing carbonates or hydroxides of monovalent alkali metals in the treating material, the amount of calcium ions in the waste is appropriately be controlled to the direction of decrease, so that the Pb elution preventing performance of the water glass aqueous solution can markedly be improved. This is considered due to that carbonic acid ions or hydroxide ions originating from the dissolved carbonates or hydroxides of monovalent alkali metals react with calcium ions to produce calcium ions which are sparingly soluble in water. Therefore, materials which dissolve to produce carbonic acid ions or hydroxide ions are expected to have the same effect.

However, of the carbonates, hydrogen carbonate releases hydrogen ion (H^+) at the dissolution thereof, and induces gelation reaction with water glass. Therefore, it cannot be used as a one part treating material of a mixing system with water glass. That is, in the case that hydrogen carbonate is used, it is necessary to add the same separately from water glass, and it is very inconvenient practically that, for example, two sets of a tank for waste-treating material, a pump for conveying the treating material, a control system, and the like are required. Contrary to this, by using carbonates, particularly potassium carbonate or sodium carbonate, carbonic acid ions can be supplied without gelling water glass. By this, one part system of the treating material is possible, and the treating material excellent in the convenience compared with the prior art can be provided. Lithium carbonate can be exemplified as the carbonates having the above action. However, lithium carbonate has low solubility, and unless it has sufficiently low concentration when blending with the treating material, precipitates are formed, lacking in practical convenience.

On the other hand, hydroxides of monovalent alkali metals exhibit the same effect if those are compounds which produce hydroxide ions by dissolution. Examples of the hydroxides of monovalent alkali metals having the action as described above include sodium hydroxide, potassium hydroxide and lithium hydroxide. In the case that hydroxides of polyvalent (divalent or more) metal salts are used, it acts as a gelling agent, and the treating material may undesirably gel during the storage thereof.

The blending ratio of the water glass and potassium carbonate is preferably that a ratio of water glass solid content : potassium carbonate is in the range of 90 : 10 to 40 : 60. If the amount of potassium carbonate is less than the above range, supply of calcium ions is not sufficient, and on the other hand, if the amount exceeds the above range, the amount of water glass component is relatively decreased, and it cannot sufficiently exhibit the effect. The reason that the preferred blending ratio is not specified is that the optimum blending ratio varies depending on the kinds of waste to be applied, mainly calcium content. The desired effect can be exhibited to an average waste in the above-described range of the blending ratio.

In the case that a blend of water glass and potassium carbonate is used as the treating material, a large amount of the treating material is required if its solid content concentration is low. Therefore, a storage tank of a large size is necessary, which is disadvantageous on practical use. For this reason, it is preferred that water glass used in preparing the treating material is directly used without diluting a crude liquid of aqueous solution which is industrially available. On the other hand, it is considered to be preferable that an aqueous solution of potassium carbonate having higher concentration is prepared, and water glass and potassium carbonate are mixed for use. The concentration of the potassium carbonate aqueous solution is preferably 40% or more. However, in the case that water glass is sodium silicate, there is the possibility that sodium carbonate is produced in the blended solution when blending potassium carbonate. Sodium carbonate has a solubility far lower than that of potassium carbonate. Therefore, in the case that potassium carbonate aqueous solution having high concentration is added to a crude liquid of sodium silicate, salt of sodium carbonate may precipitate. Condition that this phenomenon develops varies depending on the blending ratio of water glass and potassium carbonate. The concentration of potassium carbonate aqueous solution when preparing a blend is preferably set about 50% or less. It is considered that a method of using a blend in a suspended state that the entire amount of potassium carbonate added to water glass is not dissolved is also effective, when considering volume of the treating material, and the like.

The blending ratio of the water glass and sodium carbonate is preferably such that a ratio of water glass solid content : sodium carbonate is in the range of 90 : 10 to 40 : 60, and more preferably such that the ratio of water glass content and sodium carbonate is in the range of 60 : 40 to 50 : 50 per 100 parts by weight of the water glass solid content. If the blending amount of sodium carbonate is less than the above range, capture of calcium is not sufficient, and on the other hand, if it exceeds the above range, the water glass content is relatively small, and it cannot sufficiently exhibit the effect. The reason that the preferred blending ratio is not specified is that the optimum blending ratio varies depending on the kinds of waste to be applied, mainly calcium content. However, from the fact that the treating material having the above-described range exhibits the desired effect to many average waste, it can be said that the above-described range is most preferably.

In the case that a mixed aqueous solution of water glass and sodium carbonate is used as the treating material, if its solid content concentration is low, a large amount of the treating material is required, so that a storage tank of large size is necessary, which is practically disadvantageous. Therefore, it is preferred that water glass used in preparing the

treating material is directly used without diluting a crude liquid of aqueous solution which is industrially available. On the other hand, it is preferred that an aqueous solution of sodium carbonate of higher concentration is prepared, and such an aqueous solution is mixed with the above water glass. In order to prepare a sodium carbonate aqueous solution of the highest concentration, sodium carbonate is dissolved in heated water of 40°C or more, and in this case, the solid content concentration is about 32% by weight. However, it is industrially disadvantageous that heated water must be provided in dissolution of sodium carbonate. The solubility of sodium carbonate at normal temperature (15°C) is about 15% by weight. Since dissolution of sodium carbonate involves heat generation, when dissolving in water at normal temperature, about 20% by weight of sodium carbonate is substantially dissolved. Thus, concentration and preparation method of the mixed aqueous solution must be determined, considering the disadvantage of providing heated water and the advantage of making the concentration of the mixed solution high. In any case, it is preferred that the concentration of sodium carbonate is set to 20% by weight or more. It is also possible to use a mixed liquid in a suspended state that the entire amount of sodium carbonate added to water glass does not dissolve.

The blending ratio of the water glass aqueous solution and sodium hydroxide is preferably that a ratio of water glass solid content : sodium carbonate is in the range of 90 : 10 to 40 : 60. If the blending amount of sodium hydroxide is less than the above range, replenishment of calcium is not sufficient, and on the other hand, if the blending amount thereof exceeds the above range, the amount of the water glass component is relatively small, and the elution preventing effect of harmful metals is not sufficiently exhibited. Since the optimum blending ratio varies depending on the kinds of waste to be applied, particularly calcium content, the preferred blending ratio is not always specified. However, from the fact that the treating material having the blending ratio of water solid content : sodium hydroxide = 60 : 40 exhibits the desired effect to many average wastes, it can be said that such a ratio is the most preferred ratio.

In the case that a mixed aqueous solution of the water glass aqueous solution and sodium hydroxide is used as the treating material, if the solid content concentration is low, a large amount of the treating material is required. Therefore, a storage tank of a large size is necessary, which is disadvantageous on practical use. For this reason, it is preferred for the water glass aqueous solution used in preparing the treating material to directly use a crude liquid of an aqueous solution industrially available without dilution. On the other hand, it is preferred that an aqueous solution of a hydroxide having the higher concentration is prepared and the water glass aqueous solution and such a hydroxide solution are mixed and used. In order to prepare a hydroxide aqueous solution of the highest concentration, a hydroxide is dissolved in heated water. However, if the concentration of the hydroxide aqueous solution is too high, there is the problem that structural components of the treating material are liable to precipitate during storage by temperature variation of surroundings. Therefore, the concentration and preparation method of a mixed aqueous solution must be determined considering volume and storage stability of the treating material. In any event, it is preferred that the concentration of the hydroxide aqueous solution is set to 20% by weight or more. A method of using a blend in a suspended state that the entire amount of the hydroxide added to the water glass aqueous solution is not dissolved is also within the scope of the present invention.

A mixed aqueous solution of the water glass aqueous solution and carbonates, or hydroxides of monovalent alkali metals of the present invention may precipitate salts of the structural components depending on temperature and solid content concentration. In such a case, there arises practical problems such as clogging of a filter. Therefore, it is necessary that a mixed aqueous solution is heated at a constant temperature from the time of preparation according to temperature of surroundings and concentration of the solution, or when salts are precipitated without maintaining at a constant temperature, the mixed aqueous solution is used after re-dissolving part or the whole of salts. The temperature for maintaining at a constant temperature or heating temperature is preferably 30 to 70°C. If the temperature is less than 30°C, it is difficult to completely prevent precipitation of salts of a blend in high concentration, and also it requires much time to re-dissolve salts precipitated. On the other hand, if the temperature exceeds 70°C, there arises such a problem that a gel which is assumed to be a reaction product of air and the water glass is formed.

In the case that potassium is selected as carbonates, since solubility of potassium hydroxide is high, it is sufficient only to heat to a temperature of about 5 to 30°C.

The ion sealing agent which seals calcium ions, as the additive B means a material which is generally known to seal metal ions by bonding with metal ions. Examples of the ion sealing agent include potassium 1-hydroxyethane-1,1-diphosphonate, sodium 1-hydroxyethane-1,1-diphosphonate, potassium aminotrimethylene phosphonate, ethylene diamine tetraacetic acid, potassium ethylene diamine tetraacetate, sodium ethylene diamine tetraacetate, lithium ethylene diamine tetraacetate, nitrilotriacetic acid, potassium nitrilotriacetate, sodium nitrilotriacetate, chromotropic acid, sodium chromotropate, potassium chromotropate, dimethyldithiocarbamic acid, sodium dimethyldithiocarbamate, potassium dimethyldithiocarbamate, diethyldithiocarbamic acid, sodium diethyldithiocarbamate, potassium diethyldithiocarbamate, sodium dibutyldithiocarbamate and potassium dibutyldithiocarbamate. Further, it is known that tetrapotassium diphosphate (potassium pyrophosphate), tetrasodium diphosphate (sodium pyrophosphate), pentapotassium triphosphate (potassium tripolyphosphate), pentasodium triphosphate (sodium tripolyphosphate), hexapotassium tetraphosphate (potassium tetrapolyphosphate), hexasodium tetraphosphate (sodium tetrapolyphosphate), potassium trimetaphosphate, sodium trimetaphosphate, potassium hexametaphosphate, sodium hexametaphosphate, and the like which are exemplified as the materials which produce calcium compounds which are insoluble or sparingly soluble in

water, by reacting with calcium ions, and bond to calcium ions to form calcium complex ions. Therefore, those compounds can be exemplified as the examples of the ion sealing agent which seals calcium ions. Of those, phosphonates of sodium or organic chelating agents having carboxyl group ($-\text{COOX}$, wherein X represents hydrogen, potassium or sodium) or dithiocarbamic acid group ($>\text{NCSSY}$, wherein Y represents hydrogen, potassium or sodium) are preferably used considering the sealing ability to metal ions, particularly calcium ion, and potassium ethylene diamine tetraacetate, sodium ethylene diamine tetraacetate, potassium 1-hydroxyethane-1,1-diphosphonate, sodium 1-hydroxyethane-1,1-diphosphonate, sodium dimethyldithiocarbamate, potassium diethyldithiocarbamate and sodium dibutyldithiocarbamate are more preferably used considering industrial availability, cost, Pb stabilizing performance and the like.

It is also within the scope of the present invention to use together at least two of materials which produce calcium compounds which react with calcium ions to produce calcium compounds which are insoluble in water, material which react with calcium ions to produce calcium compounds which are sparingly soluble in water, and ion sealing agents which seal calcium ions, as the additive B if necessary.

It should be noted that, in the waste-treating material as described above, when phosphates, sulfonates, carbonates, or materials having carboxyl group are used as acids and polyvalent metal salts as the additive A, the function of the additive B that those are reacted with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water or to seal calcium ions is further imparted to the function of the additive A, so that it is expected that the Pb stabilizing effect of the treating material is improved by the synergistic effect.

In the waste-treating material of the present invention, if the additive A is blended, it is required that solid component of the water glass aqueous solution is polymerized by the action of the additive A. However, if the polymerization reaction proceeds excessively, gel of water glass precipitates, i.e., solidifying with only the treating material, and as a result, it is impossible to seal harmful metals in waste. Therefore, care should sufficiently be made to storage conditions (storage form, storage place, storage time, etc.) of the treating material, and the kind and amount of the additive A added, and it is necessary to set the optimum value to ones appropriately selected. In other words, it is required that solid component in the water glass aqueous solution is polymerized by the action of acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters, dialdehydes and the like added as the additive A. However, if this polymerization reaction proceeds excessively, gel of water glass precipitates, i.e., solidifying with only the treating material, so that it is impossible to seal harmful metals and the like in the waste.

Further, if the polymerization reaction by the water glass aqueous solution and the additive B proceeds excessively, or if the amount of the additive B added is too large, gel of water glass precipitates, i.e., solidifying with only the treating material, or solid component in the water glass aqueous solution precipitates by salting out effect of co-present ions in the treating material. As a result, it is impossible to seal harmful materials in the waste. Therefore, care should sufficiently be made to storage conditions (storage form, storage place, storage time, etc.) of the treating material, and the kind and amount of the additive A and additive B added, and it is necessary to set the optimum value to ones appropriately selected.

In order to avoid this problem, for the purpose of stably storing the treating material comprising the water glass aqueous solution and the additive A or the additive B as the main structural components in the state that an industrial problem does not arise, it is preferred that in addition to the above main structural components, at least one selected from gluconates, tartarates, benzoates, ligninsulphonates, polysaccharides, and bases comprising monovalent cation and hydroxide ion is added as a one part stabilizing agent as an additive E. The mechanism that the additive E stabilizes the treating material comprising the water glass aqueous solution and the additive A or the additive B as the main structural components, as a liquid is not clear, but it is assumed that such is due to an action of chelating the additive A or the additive B present in excess to conceal, or an action of controlling balance of electric charges in the treating material and pH thereof.

The addition method and amount of those stabilizing agents vary depending on properties of the stabilizing agent added, desired stabilization of the treating material, and the like, but basically it is preferred that after the reaction between the water glass aqueous solution and acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters, dialdehydes, and the like sufficiently proceeds and polymerization of solid component in the water glass aqueous solution is completed, the treating material is stored by adding a stabilizing agent thereto. The amount of the stabilizing agent added is practically a minimum amount necessary for attaining the desired one part stability of the treating material. Of those stabilizing agents, if the amount of a stabilizing agent added is excess, some stabilizing agents may act as a gelling agent of the water glass aqueous solution. Therefore, the amount of the stabilizing agent added is about 20 parts by weight or less per 100 parts by weight of the solid component (the sum of amount of M_2O and amount of SiO_2) of the water glass aqueous solution. If the stabilizing agent is added in an amount exceeding the above amount, progress of gelation of the treating material is promoted, and stability of the treating material may be undesirably decreased. Of the above stabilizing agents, it is preferred that the base comprising the monovalent cation and hydroxide ion is sodium hydroxide (NaOH), potassium hydroxide (KOH) and aqueous ammonia (NH_4OH), considering industrial availability, cost, the performance for stabilizing the treating material in one part, and the like.

Method of adding the treating material to the waste in the treating method of kneading the treating material com-

prising the water glass aqueous solution and the additive A or the additive B as the main structural components with the waste according to the present invention, and aging the mixture are, for example:

- (1) a method of mixing at least the water glass aqueous solution and the additive A in the main structural components of the treating material, and after 5 minutes or more, adding the resulting mixture to the waste;
- (2) a method of separately storing the water glass aqueous solution and the additive A or the additive B, which are the main structural components of the treating material, mixing those immediately before adding to the waste, and adding the resulting mixture to the waste;
- (3) a method of separately storing the water glass aqueous solution and the additive A or the additive B, which are the main structural components, adding the water glass aqueous solution to the waste, if necessary followed by kneading, and then adding the additive A or the additive B thereto.

Those methods should appropriately be selected depending on properties of the treating material used, treating facilities, and the like.

When the above method (1) is selected, time for sufficiently proceed a polymerization reaction of the water glass aqueous solution and the additive A is secured, and such is useful. In other words, in order that this treating material exhibits the desired performance, it is required that the solid component in the water glass aqueous solution reacts with the additive A to polymerize. However, if the treating material is mixed and then added to the waste within less than 5 minutes, it is considered that the polymerization reaction does not proceed sufficiently, and the treating material may not show the desired performance for stabilizing harmful metals. Therefore, it is preferred that among the main structural components, the water glass aqueous solution and the additive A are first mixed, and after 5 minutes or more from the mixing, the resulting mixture is added to the waste.

Separately storing the water glass aqueous solution and the additive A or the additive B, which are the main structural components of the treating material, is the preferred embodiment in the method of treating waste in the case that it is impossible on stabilization of the treating material to store this treating material as a one part, or in the case that polyvalent metal salts are contained as the additive A, and the additive B reacts with polyvalent metal ions in the polyvalent metal salts prior to polymerization reaction of the water glass aqueous solution with the polyvalent metal salts to produce polyvalent metal compounds which are insoluble or sparingly soluble in water, or to seal the polyvalent metal ions, whereby the desired performance for stabilizing harmful metals may not be exhibited. Either of the above method (2) of mixing the water glass aqueous solution and the second component or the third component immediately before adding to the waste, and immediately adding the resulting mixture to the waste or the above method (3) of adding the water glass aqueous solution to the waste, if necessary followed by kneading, adding the additive A or the additive B thereto, kneading and aging can be selected as the method for adding the treating material to the waste.

The above method (2) is effective in the case that a gel is not instantaneously formed when the water glass aqueous solution and the second component are reacted, or in the case that polyvalent metal salts of the additive A are instantaneously reacted with the additive B, so that polyvalent metal compounds which are insoluble or sparingly soluble in water are not formed, or polyvalent metal ions are not sealed. In other words, the method (2) is effective to the treating material that although it is impossible to stably store the treating material as one part, polymerization reaction between the water glass aqueous solution and the second component proceeds until adding to the waste after mixing the treating material to form a one part, and the treating material does not gel until adding to the waste.

On the other hand, the above method (3) is effective in the case that a gel is instantaneously formed when the water glass aqueous solution and the additive A are mixed or in the case that polyvalent metal salts of the additive A are instantaneously reacted with the additive B to produce polyvalent metal compounds which are insoluble or sparingly soluble in water or polyvalent metal ions are sealed. In this case, it is conditions for sufficiently exhibiting harmful metal stabilizing performance of the treating material that harmful metal in the waste do not elute until the water glass aqueous solution is added to the waste, if necessary, followed by kneading, and the additive A and the additive B are then added thereto, and all the water glass aqueous solution does not completely react with calcium compounds in the waste. In other words, the water glass aqueous solution added to the waste must be instantaneously reacted with the additive A which are added thereafter to produce a polymer of the water glass solid component, and then must be reacted with calcium compounds and the like in the waste.

In practicing any one of the above methods (1) to (3) for treating waste, it is required that a device for introducing the treating material, treating conditions and the like are appropriately set according to the properties of the treating material. Further, contrary to the above methods (1) to (3), if a method is employed that the water glass aqueous solution, the additive A and the additive B are separately stored and the additive A and the additive B are added to the waste prior to adding the water glass aqueous solution, elution of harmful metals from the waste occurs before the water glass aqueous solution stabilizes harmful metals. Therefore, this method is not preferable.

The second waste-treating material of the present invention comprises the water glass aqueous solution having added thereto a coagulating sedimentator in an amount of 1 to 40 parts by weight per 100 parts by weight of the water glass solid component in the water glass aqueous solution. If the amount of the coagulating sedimentator added is less

than 1 part by weight, the synergistic effect by co-use with the water glass is not almost recognized, and on the other hand if the amount of the coagulating sedimentator added exceeds 40 parts by weight, water glass may gel, whereby it will be difficult to use the treating material. In this treating material, although the coagulating sedimentator may not always be completely dissolved in the water glass aqueous solution. However, in the case of treating by adding the treating material to the waste, it is preferred that the treating material is uniformly added, and from this point, it is advantageous that the coagulating sedimentator is completely dissolved in the water glass aqueous solution. To achieve this, water may additionally be added to the treating material or the coagulating sedimentator may previously be dissolved in water and the resulting solution is then mixed with water glass to prepare a treating material.

Examples of the coagulating sedimentator used in the treating material include polyaluminum chloride, sulfuric acid band, polyacrylamide, sodium polyacrylate, carbamate, polymethacrylic acid ester, polyvinyl alcohol, methyl cellulose, hydroxypropylmethyl cellulose, hydroxyethylmethyl cellulose and carboxymethyl cellulose. Polyaluminum chloride, carbamate, polyacrylamide, sodium polyacrylate, polyvinyl alcohol, methyl cellulose and carboxymethyl cellulose are preferred considering Pb stabilizing performance and the like. Those compounds may be used alone or as mixtures of two or more thereof as the coagulating sedimentator.

In the third treating material of the present invention, where monovalent metal salts as the additive D are added such that the amount of monovalent metal ions released from monovalent metal salts exceeds 100 parts by mol per 100 parts by mol of cations (sodium where the water glass aqueous solution is sodium silicate, and hereinafter the same) in the water glass aqueous solution, ion concentration in the treating material aqueous solution approaches its saturation, so that solid component in the water glass aqueous solution is liable to precipitate. The treating material aqueous solution wherein the solid component is precipitated becomes a form unusable as a liquid, and disadvantageous problems induce that pipings of a device for introducing the treating material aqueous solution, a kneader for the waste and the treating material aqueous solution, and the like may possibly clog, which is undesirable. Further, the treating material aqueous solution in which solid component is precipitated can be expected to have the Pb stabilizing performance in a certain degree, but it is not sufficient. For this reason, the amount of the monovalent metal salts added is preferably such that the amount of monovalent metal ions of the monovalent metal salts is 100 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution. If the amount the monovalent metal salts added is too small, its salting out effect cannot be expected, and there is the possibility that the Pb stabilizing performance of the treating material may sufficiently be improved. From this, it is preferred that the amount of the monovalent metal salts added is such that the amount of monovalent metal ions of the monovalent metal salts is 5 parts by mol or more per 100 parts by mol of cations in the water glass aqueous solution.

From the same reason as above, the amount of ammonium salts as the additive D is preferably such that the amount of ammonium ions of the ammonium salts is more than 5 to 100 parts by mol per 100 parts by mol of cations in the water glass aqueous solution.

The monovalent metal salts may be any salts so long as it ionizes in water or the water glass aqueous solution to release monovalent metal ions, and it does not produce calcium compounds which are insoluble or sparingly soluble in water by reacting with calcium ions. Examples of the monovalent metal salts include chlorides or nitrates of sodium or potassium. Of those, sodium chloride and potassium chloride are preferred considering industrial availability, Pb stabilizing performance, and the like. It is also within the scope of the present invention that at least two of different kinds of monovalent metal salts are used together if necessary.

The ammonium salts may be any salt so long as it ionizes in water of the water glass aqueous solution to release ammonium ion, and it does not produce calcium compounds which are insoluble or sparingly soluble in water by reacting with calcium ions. Of those, ammonium chloride is preferably used considering industrial availability, cost, Pb stabilizing performance, and the like. If necessary, at least two different kinds of ammonium salts can be used together.

It is also possible to use at least one selected from the monovalent metal salts and at least one selected from the ammonium salts together as the additive D.

The amounts of those additives A, B, C and D vary depending on concentration of the water glass aqueous solution, compositions of the water glass aqueous solution, amount of the water glass aqueous solution added to the waste, method for adding the additive A, Pb content in the waste, calcium compound content, elution amount of harmful metals from the waste in the case of non-treatment, and intended elution allowable amount of harmful metals, such as a rule-regulated value in a filled-in land and the like. Practically, those amounts are determined by the blending ratio of the water glass aqueous solution and the additives A, B, C and D such that the treating material has a predetermined performance for stabilizing harmful metals, and is most inexpensive. If desired and necessary, the water glass aqueous solution may be combined with the additives A, B, C and D in any optional blending ratio.

It is effective means in the method for treating waste according to the present invention for sufficiently exhibiting the effect of the treating material to mix the treating material and the waste, knead the resulting mixture, and then age the resulting kneaded mixture, and this is also within the scope of the present invention. This suggests that although reaction is instantaneously initiated by contacting the treating material of the present invention with the waste to exhibit the effect, the reaction gradually proceeds thereafter.

The aging temperature in this case is preferably about 40 to 100°C. The function of this aging is not entirely under-

stood, but it is considered that by aging a blend of the treating material and the waste by heating to the above temperature range, reaction which proceeds during aging is accelerated. It is also assumed that reaction product produced in aging becomes more stable state, and the effect for more strongly preventing elution of harmful metals contained is improved. The aging temperature varies depending on Pb content in the waste, calcium compound content, elution amount of harmful metals from the waste in the case of non-treatment, and intended elution allowable amount, such as rule-regulated value applied to a place where treated materials are disposed, and the like. In general, where the intended elution allowable amount is severely restricted, the ability for stabilizing harmful metals and the like are improved by setting the aging temperature to higher temperature. However, if a blend is aged at a temperature exceeding 100°C, water in the blend rapidly evaporates, so that the harmful metal stabilizing performance of the treating material may markedly be decreased. Further, the blend becomes a dried state, so that the blend containing harmful metals scatters in conveyance working, and there may be the possibility to contaminate surrounding environment. Therefore, it is not preferable to age at a temperature exceeding 100°C. For the reasons above, the temperature in aging a blend of the waste and the treating material is preferably 40 to 100°C. Further, considering the fact that working conditions deteriorate if heating to a temperature more than necessity, the aging temperature is more preferably 80° or less. Aging under heating up to such a temperature is effective in the case that a site for aging the treated material after treating the treating material is not sufficiently secured. In other words, it is possible to shorten the aging time by aging a blend obtained by kneading the waste and the treating material at a predetermined temperature, and this enables the treated material to rapidly discard.

It is preferred in the method for treating waste according to the present invention to age a blend of the waste and the treating material for 6 hours or more. In general, a solidification strength of a blend of the waste and the treating material increase as increasing the aging time. Therefore, where aging is not conducted for sufficient time, the blend lacks in solidification strength. As a result, a treated material disintegrates during transportation of the same from aging pit to a cart, filled-in land, and the like, or in the filled-in land, dusts containing harmful metals scatter and there is the possibility to contaminate surrounding environment. Further, if the aging time is less than 6 hours, it is insufficient to proceed polymerization reaction during the period of aging, and the treating material may not exhibit the desired performance for stabilizing harmful metals. From the reasons above, the blend of the waste and the treating material is preferably aged for 6 hours or more.

In many cases, a site for treating waste, particularly intermediate treating site of waste incineration fly ash, is located near waste incineration facilities. In general, in those incineration sites, heat generated in waste incineration is recovered as steam. Additionally, there is the case that generation of electric power is conducted utilizing the steam. It is industrially useful in the method for treating waste according to the present invention to utilize steam recovered from a heat exchanger of the incineration site as heat source in aging a blend obtained by kneading the waste and the treating material, and to utilize electric power obtained by utilizing the steam, leading to an effective use of energy from an industrial standpoint.

The amount of the water glass aqueous solution added to the waste is not generally determined because the Pb stabilizing performance of the treating material varies depending on the amounts of the additives A, B, C and D added. However, practically it is a factor for determining the amount of the water glass aqueous solution added from the standpoint of cost that the intended elution amount or less is achieved by addition of the water glass aqueous solution in the smallest amount, although varying depending on Pb content in the waste, calcium compound content, elution amount of harmful metals from the waste in the case of non-treatment, and intended elution allowable amount, such as rule-regulated value. Actually, in substantially all of wastes, it is possible to suppress the elution amount to 0.3 ppm which is the rule-regulated value in Japan by adding the water glass aqueous solution in an amount of about 30 parts by weight or less, in terms of the solid component therein, per 100 parts by weight of the waste. If the water glass aqueous solution in terms of the solid component is added exceeding the above range, such is an excessive amount, except that large strength is required in re-utilization of treated material and the like, volume of solidified product increases. This may induce the problems such that filled-in land for solidified product is not secured, and cost increases. For this reason, the amount of the water glass aqueous solution is preferably 20 parts by weight or less in terms of the solid component therein per 100 parts by weight of the waste.

In mixing and kneading the treating material of the present invention with the waste, it is important for exhibition of the effect to sufficiently contact the treating material with the waste. To achieve this, the treating material may previously be diluted with water, or the treating material and the waste may be mixed or kneaded and water may then be added to the resulting mixture, followed by kneading. Although it is not generally defined because water content, wettability with the treating material, and the like vary depending on the kinds of waste, the sum of water in the treating material and water added is preferably 25 parts by weight per 100 parts by weight of the waste where the waste is relatively dried material. If the amount of water is too large, problems may induce such that handling property of the waste after kneading is poor, and the treating material does not exhibit the desired effect. Therefore, care should be taken in this regard.

The waste-treating material of the present invention can also stabilize harmful metals including Pb contained in industrial waste such as collected dust generated in electric furnace or zinc plating step, which does not contain calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride, which may be capable of forming a gelling

agent of the water glass aqueous solution, or which does not contain other polyvalent metal salts. More specifically, calcium hydroxide, calcium oxide, calcium chloride and the like which are capable of forming a gelling agent of the water glass aqueous solution, or other polyvalent metal salts are previously blended with the waste, so that it is possible to exhibit the harmful metal stabilizing performance of the treating material and the method for treating waste, according to the present invention by the same action mechanism as in the treatment of the waste containing calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride.

Therefore, use of the treating material and the method for treating waste, according to the present invention makes it possible to conduct stabilization treatment of harmful metals in the waste containing calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride, in particular Pb in waste incineration fly ash.

DESCRIPTION OF THE PREFERRED EMBODIMENT

The present invention is described in more detail by reference to the following examples and comparative examples, but it should be understood that the invention is not construed as being limited thereto.

COMPARATIVE EXAMPLE 1

Analytical result of components of bag filter collected fly ash (I) formed in municipal refuse incineration facility is shown in Table 1 below.

TABLE 1

Analytical result of components of bag filter collected fly ash (I) formed in municipal refuse incineration facility (% by weight)	
Component	Analytical result
$\text{Ca}(\text{OH})_2$	12.5
$\text{CaCl}_2 \cdot \text{Ca}(\text{OH})_2 \cdot \text{H}_2\text{O}$	24.6
Pb	0.65

This fly ash (I) containing calcium compound was subjected to an elution test defined by Notification No. 13 of the Environment Agency (Japan) (hereinafter referred to as "elution test" for simplicity) in a non-treated state.

COMPARATIVE EXAMPLE 2

26.0 g of water was added to 6.5 g (solid content: 2.5 g) of a sodium silicate solution (I) (a sodium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd., trade name: J SODIUM SILICATE No. 3, solid content: 38.5%, $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio = 3.15) to prepare a water glass aqueous solution as a treating material. This treating material was added to 50 g of the fly ash (I) obtained in Comparative Example 1 above, and kneaded. The resulting kneaded mixture (5 parts by weight of the solid content of the water glass aqueous solution per 100 parts by weight of the ash) was aged and solidified at 20°C for 24 hours. After completion of the solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from particles which were passed through the sieve and was subjected to the elution test.

COMPARATIVE EXAMPLE 3

1.58 g of hydrochloric acid (36% HCl) was added to a water glass aqueous solution prepared by adding 26.0 g of water to the same sodium silicate solution (I) as used in Comparative Example 2 above. The resulting solution (hydrogen ion of hydrochloric acid was 75 parts by mol per 100 parts by mol of alkali metal of the water glass aqueous solution) formed gel instantaneously. Thus, it was impossible to add such a solution as a treating material to 50 g of the fly ash (I) and to knead those.

COMPARATIVE EXAMPLE 4

A calcium chloride aqueous solution (0.86 g of CaCl_2 was dissolved in 26.0 g of water) was added to 6.5 g (solid

content: 2.5 g) of the sodium silicate solution (I) as used in Comparative Example 2 above (hereinafter referred to a "sodium silicate solution (I)" for simplicity). The resulting solution (the product of the part by mol of calcium ion and the number of valency was 75 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) formed gel instantaneously. Thus, it was impossible to add such a solution as a treating material to 50 g of the fly ash (I) and to knead those.

COMPARATIVE EXAMPLE 5

0.52 g of sulfuric acid (97% H_2SO_4) was added to an water glass aqueous solution prepared by adding 26.0 g of water to the sodium silicate solution (I). 0.75 g of sodium gluconate (30 parts by weight per 100 parts by weight of water glass solid content) was further added to the solution obtained above (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution). The resulting solution formed gel instantaneously. Thus, it was impossible to add such a solution as a treating material to 50 g of the fly ash (I) and to knead those.

COMPARATIVE EXAMPLE 6

6.5 g of the sodium silicate solution (I) (solid content: 2.5 g) was instantaneously added to a material prepared by adding hydrochloric acid aqueous solution (4.20 g of 36% hydrochloric acid was dissolved in 26.0 g of water) to 50 g of the fly ash (I) and kneading those. The kneaded product (solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged, solidified, pulverized, classified with a sieve, and then subjected to the elution test in the same manner as in Comparative Example 2.

COMPARATIVE EXAMPLE 7

0.52 g of sulfuric acid (97% H_2SO_4) was added to an water glass aqueous solution prepared by adding 26.0 g of water to sodium silicate solution (I). The resulting solution (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as a treating material was added to the fly ash (I) and kneaded. The kneaded product (solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged and solidified at 120°C for 6 hours. As a result, the solidified product was in a dry state, that is, in the state that it was liable to scatter. The solidified product was classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from particles which passed through the sieve, and then subjected to the elution test.

The results of the elution test in Comparative Examples 1, 2, 6 and 7 are shown in Table 2 below.

TABLE 2

Elution amount of Pb (ppm)	
Comparative Example 1	240
Comparative Example 2	50
Comparative Example 6	150
Comparative Example 7	80

EXAMPLE 1

1.05 g of hydrochloric acid (36% HCl) as the additive A was added to a water glass aqueous solution prepared by adding 26.0 g of water to the sodium silicate aqueous solution (I). The resulting solution (hydrogen ion of hydrochloric acid was 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution) was allowed to stand for 5 minutes, and then added as a treating material to 50 g of the fly ash (I) above, and kneaded. The resulting kneaded product (an amount of the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged and solidified at 20°C for 24 hours. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from particles which passed through the sieve, and then subjected to the elution test.

EXAMPLE 2

Elution test was conducted in the same manner as in Example 1 except that 0.48 g of ethanol (C_2H_5OH) was added in place of hydrochloric acid as the additive A.

EXAMPLE 3

A calcium chloride aqueous solution (0.57 g of $CaCl_2$ was dissolved in 26.0 g of water) as the additive A was added to 6.5 g of the sodium silicate aqueous solution (I) (the solid content: 2.5g). The resulting solution (the product of the part by mol of calcium ions and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) was allowed to stand for 5 minutes, and then added as a treating material to 50 g of the fly ash (I) above. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged, solidified, pulverized, and classified with a sieve in the same manner as in Example 1, and then subjected the elution test.

EXAMPLES 4 TO 8

The elution test was conducted in the same manner as in Example 3 except that the following solution was used in place of the calcium chloride aqueous solution as the additive A.

Example 4: Ethylene glycol diacetate aqueous solution (0.57 g of $C_2H_4(OCOCH_3)_2$ was dissolved in 26.0 g of water)

Example 5: Ethylene carbonate aqueous solution (1.00 g of $(CH_2O)_2CO$ was dissolved in 26.0 g of water)

Example 6: γ -butyrolactone aqueous solution (1.00 g of $C_4H_6O_2$ was dissolved in 26.0 g of water)

Example 7: Succinic acid dimethyl ester aqueous solution (1.00 g of $CH_3OOC(CH_2)COOCH_3$ was added to 26.0 g of water)

Example 8: Glyoxal aqueous solution (1.00 g of $(CHO)_2$ was dissolved in 26.0 g of water)

EXAMPLES 9 TO 14

The elution test was conducted in the same manner as in Example 1 except that the following compound was used in place of hydrochloric acid as the additive A.

Example 9: 1.07 g of nitric acid (61% HNO_3) (hydrogen ion of nitric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 10: 0.40 g of phosphoric acid (85% H_3PO_4) (hydrogen ion of phosphoric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 11: 1.00 g of dry ice (CO_2) as carbonic acid source

Example 12: 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 13: 0.62 g of acetic acid (CH_3COOH) (hydrogen ion of acetic acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 14: 0.47 g of oxalic acid (98% $(COOH)_2$) (hydrogen ion of oxalic acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

EXAMPLES 15 TO 29

The elution test was conducted in the same manner as in Example 3 except that the following solution was used in place of the calcium chloride aqueous solution as the additive A.

Example 15: Strontium carbonate aqueous solution (0.03 g of $SrCO_3$ was dissolved in 26.0 g of water)

Example 16: Barium phosphate aqueous solution (0.03 g of $Ba_3(PO_4)_2$ was dissolved in 26.0 g of water)

Example 17: Zinc sulfate aqueous solution (0.83 g of $ZnSO_4$ was dissolved in 26.0 g of water) (the product of the part by mol of zinc ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 18: Magnesium chloride aqueous solution (0.49 g of $MgCl_2$ was dissolved in 26.0 g of water) (the product of the part by mol of magnesium ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 19: Iron (II) chloride aqueous solution (0.65 g of $FeCl_2$ was dissolved in 26.0 g of water) (the product of

the part by mol of iron (II) ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 20: Iron (III) chloride aqueous solution (0.56 g of FeCl_3 was dissolved in 26.0 g of water) (the product of the part by mol of iron (III) ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 21: Aluminum chloride aqueous solution (0.46 g of AlCl_3 was dissolved in 26.0 g of water) (the product of the part by mol of aluminum ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 22: Magnesium nitrate aqueous solution (0.76 g of $\text{Mg}(\text{NO}_3)_2$ was dissolved in 26.0 g of water) (the product of the part by mol of magnesium ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 23: Calcium nitrate aqueous solution (0.84 g of $\text{Ca}(\text{NO}_3)_2$ was dissolved in 26.0 g of water) (the product of the part by mol of calcium ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 24: Iron (II) nitrate aqueous solution (0.92 g of $\text{Fe}(\text{NO}_3)_2$ was dissolved in 26.0 g of water) (the product of the part by mol of iron (II) ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 25: Iron (III) nitrate aqueous solution (0.83 g of $\text{Fe}(\text{NO}_3)_3$ was dissolved in 26.0 g of water) (the product of the part by mol of iron (III) ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 26: Aluminum nitrate aqueous solution (0.73 g of $\text{Al}(\text{NO}_3)_3$ was dissolved in 26.0 g of water) (the product of the part by mol of aluminum ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 27: Magnesium sulfate aqueous solution (0.62 g of MgSO_4 was dissolved in 26.0 g of water) (the product of the part by mol of magnesium ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 28: Iron (II) sulfate aqueous solution (0.78 g of FeSO_4 was dissolved in 26.0 g of water) (the product of the part by mol of iron (II) ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 29: Aluminum sulfate aqueous solution (0.59 g of $\text{Al}_2(\text{SO}_4)_3$ was dissolved in 26.0 g of water) (the product of the part by mol of aluminum ion and the number of valency was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

EXAMPLE 30

0.52 g of sulfuric acid (97% H_2SO_4) as the additive A was added to a water glass aqueous solution prepared by adding 26.0 g of water to the sodium silicate aqueous solution (I). 0.03 g of sodium gluconate (1 part by weight per 100 parts by weight of the water glass solid content) as an additive E was added to the resulting mixture (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) to prepare a treating material. The treating material was added to 50 g of the fly ash (I) above, and kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged, solidified, pulverized and classified with a sieve in the same manner as in Example 1, and then subjected to the elution test. Further, gelation of the above treating material was visually observed, and a one part stability was examined.

EXAMPLES 31 TO 38

The elution test was conducted in the same manner as in Example 30 except that the following compound was used in place of sodium gluconate as the additive E. Further, gelation of each treating material was visually observed, and the one part stability was examined.

Example 31: 0.03 g of potassium tartrate

Example 32: 0.03 g of ammonium benzoate

Example 33: 0.03 g of sodium ligninsulfonate

Example 34: 0.03 g of starch

Example 35: 0.50 g of lithium hydroxide (20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 36: 0.50 g of sodium hydroxide (20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 37: 0.50 g of potassium hydroxide (20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 38: 0.50 g of 25% aqueous ammonia (20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

EXAMPLE 39

Hydrochloric acid (2.10 g of 36% HCl was dissolved in 26.0 g of water) as the additive A was added to 6.5 g of the sodium silicate aqueous solution (I) (solid content: 2.5 g) to prepare a treating material. The treating material was instantaneously added to 50 g of the fly ash (I) above, and kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was subjected to the elution test in the same manner as in Example 1.

EXAMPLES 40 TO 46

The elution test was conducted in the same manner as in Example 39 except that the following solution was used in place of hydrochloric acid as the additive A.

Example 40: Ethanol (0.96 g of C_2H_5OH was dissolved in 26.0 g of water)

Example 41: Calcium chloride aqueous solution (1.04 g of $CaCl_2$ was dissolved in 26.0 g of water)

Example 42: Ethylene glycol diacetate aqueous solution (1.04 g of $C_2H_4(OCOCH_3)_2$ was dissolved in 26.0 g of water)

Example 43: Ethylene carbonate aqueous solution (2.00 g of $(CH_2O)_2CO$ was added to 26.0 g of water)

Example 44: γ -Butyrolactone aqueous solution (2.00 g of $C_4H_6O_2$ was added to 26.0 g of water)

Example 45: Succinic acid dimethyl ester aqueous solution (2.00 g of $CH_3OOC(CH_2)_2COOCH_3$ was added to 26.0 g of water)

Example 46: Glyoxal aqueous solution (2.00 g of $(CHO)_2$ was added to 26.0 g of water)

EXAMPLE 47

6.5 g of the sodium silicate aqueous solution (I) (solid content: 2.5 g) was added to 50 g of the fly ash (I) above and kneaded. Hydrochloric acid (4.20 g of 36% HCl was dissolved in 26.0 g of water) as the additive A was instantaneously added to the resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash), and then further kneaded. The resulting kneaded product was aged, solidified, pulverized, classified with a sieve, and then subjected to the elution test in the same manner as in Example 1.

EXAMPLES 48 TO 54

The elution test was conducted in the same manner as in Example 47 except that the following solution was used in place of hydrochloric acid as the additive A.

Example 48: Ethanol (1.92 g of C_2H_5OH was dissolved in 26.0 g of water)

Example 49: Calcium chloride aqueous solution (2.08 g of $CaCl_2$ was dissolved in 26.0 g of water)

Example 50: Ethylene glycol diacetate aqueous solution (2.08 g of $C_2H_4(OCOCH_3)_2$ was dissolved in 26.0 g of water)

Example 51: Ethylene carbonate aqueous solution (4.00 g of $(CH_2O)_2CO$ was added to 26.0 g of water)

Example 52: γ -Butyrolactone aqueous solution (4.00 g of $C_4H_6O_2$ was added to 26.0 g of water)

Example 53: Succinic acid dimethyl ester aqueous solution (4.00 g of $CH_3OOC(CH_2)_2COOCH_3$ was added to 26.0 g of water)

Example 54: Glyoxal aqueous solution (4.00 g of $(CHO)_2$ was added to 26.0 g of water)

EXAMPLE 55

0.52 g of sulfuric acid (97% H_2SO_4) was added to a water glass aqueous solution prepared by adding 26.0 g of water to the sodium silicate aqueous solution (I) to prepare a treating material. The treating material (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution) was added to 50 g of the fly ash (I) above, and kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged and solidified at 40°C for 24 hours. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5

mm. 30 g of particles was dispensed from particles which passed through the sieve, and then subjected to the elution test.

EXAMPLE 56

The elution test was conducted in the same manner as in Example 55 except that the aging temperature was 60°C and the aging time was 6 hours.

EXAMPLE 57

The elution test was conducted in the same manner as in Example 55 except that the aging temperature was 60°C and the aging time was 2 hours.

EXAMPLE 58

The elution test was conducted in the same manner as in Example 1 except that a treating material obtained by adding 0.97 g of phenol (C_6H_5OH) in place of hydrochloric acid was used (hydrogen ion of phenol was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution).

EXAMPLE 59

The elution test was conducted in the same manner as in Example 12 except that a treating material obtained by adding 0.52 g of sulfuric acid (97% H_2SO_4) to an aqueous solution obtained by adding 26.0 g of water to 6.3 g (solid content: 2.5 g) of a potassium silicate aqueous solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: A POTASSIUM SILICATE, solid content: 40.0%) was used.

EXAMPLE 60

The elution test was conducted in the same manner as in Example 12 except that sulfuric acid (97% H_2SO_4) was added to a water glass aqueous solution prepared by adding 26.0 g of water to 7.9 g (solid content: 2.5 g) of a sodium silicate aqueous solution manufactured by Nippon Chemical Industrial Co, Ltd. (trade name: 30% LIQUID ORTHO, solid content: 31.5%, SiO_2/Na_2O compositional ratio = 0.50) to prepare a treating material (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cation of the water glass aqueous solution).

The elution test results obtained in Examples 1 to 60 above are shown in Table 3 below. Further, the one part stability of each treating material is shown in Table 4 below.

TABLE 3

Elution Amount of Pb (ppm)					
Example 1	5.8	Example 2	41	Example 3	11
Example 4	38	Example 5	35	Example 6	42
Example 7	44	Example 8	39	Example 9	4.7
Example 10	16	Example 11	30	Example 12	7.3
Example 13	3.0	Example 14	5.8	Example 15	43
Example 16	45	Example 17	36	Example 18	13
Example 19	6.3	Example 20	4.1	Example 21	2.1
Example 22	23	Example 23	21	Example 24	15
Example 25	12	Example 26	8.1	Example 27	24
Example 28	19	Example 29	9.5	Example 30	4.1
Example 31	7.6	Example 32	7.4	Example 33	7.1
Example 34	7.7	Example 35	7.3	Example 36	5.9
Example 37	6.0	Example 38	6.3	Example 39	5.4
Example 40	36	Example 41	8.0	Example 42	29
Example 43	26	Example 44	35	Example 45	39
Example 46	31	Example 47	6.0	Example 48	39
Example 49	13	Example 50	40	Example 51	34
Example 52	39	Example 53	44	Example 54	37
Example 55	2.8	Example 56	1.8	Example 57	30
Example 58	48	Example 59	11	Example 60	26

TABLE 4

One Part Stability of Treating Material (Gelation Time of Treating Material/min)			
Example 12	335	Example 30	700
Example 31	710	Example 32	690
Example 33	750	Example 34	735
Example 35	720	Example 36	1,045
Example 37	980	Example 38	875

From the comparison of Comparative Examples 1 and 2, it was proved that by adding the water glass aqueous solution, the elution amount of Pb from the fly ash was decreased as compared with the non-treatment, and the water glass aqueous solution had the performance for stabilizing harmful metals (Pb). However, only such Pb stabilizing performance is not sufficient.

From the comparison between Examples 1 to 8 and Comparative Example 2, it is apparent that the treating material of the present invention has excellent Pb stabilizing performance as compared with the case of treating with only the water glass aqueous solution.

From the comparison between Example 1 and Comparative Example 3, it is apparent that the amount of the acid

added in the treating material of the present invention is preferably that hydrogen ion of the acid is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

From the comparison between Examples 1, 9 to 14 and 58 and Comparative Example 2, it is apparent that by using any of hydrochloric acid, nitric acid, phosphoric acid, sulfuric acid, carbonic acid, acetic acid and oxalic acid, the treating material having excellent Pb stabilizing performance can be obtained as compared with the case of using other acids.

From the comparison between Example 3 and Comparative Example 4, it is apparent that the amount of the polyvalent metal salts added as the additive A in the treating material of the present invention is preferably that the product of the part by mol of polyvalent metal ions of the polyvalent metal salts and the number of valency is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

From the comparison between Examples 3 and 15 to 29 and Comparative Example 2, it is apparent that where the waste-treating material comprises the water glass aqueous solution and the polyvalent metal salts as the additive A as the main structural components, when the polyvalent metal salts used are any of chlorides, nitrates, sulfates, carbonates or phosphates of magnesium, calcium, strontium, barium, iron, aluminum or zinc, and preferably any of magnesium chloride, calcium chloride, iron (II) chloride, iron (III) chloride, aluminum chloride, magnesium nitrate, calcium nitrate, iron (II) nitrate, iron (III) nitrate, aluminum nitrate, magnesium sulfate, iron (II) sulfate, and aluminum sulfate, the treating material has excellent Pb stabilizing performance as compared with the use of only the water glass aqueous solution.

From the comparison between Example 12 and Example 59, it is apparent that the water glass aqueous solution comprising sodium silicate ($\text{Na}_2\text{O} \cdot n\text{SiO}_2$) as the main structural component has excellent Pb stabilizing performance as compared with the case of using potassium silicate ($\text{K}_2\text{O} \cdot n\text{SiO}_2$) as the main structural component.

From the comparison between Example 12 and Example 60, it is apparent that when $\text{SiO}_2/\text{Na}_2\text{O} = 0.50$ and 3.15 are compared as the compositional ratio of sodium silicate which is the structural component of the water glass aqueous solution, the treating material wherein the compositional ratio is 3.15 shows excellent Pb stabilizing performance, and that it is preferred as the treating material that the treating material comprises the water glass aqueous solution ($\text{M}_2\text{O} \cdot n\text{SiO}_2$) wherein the main structural component of the water glass aqueous solution is $\text{SiO}_2/\text{M}_2\text{O} = 2.00$ or more (wherein M represents a cation in the water glass aqueous solution).

From the comparison between Example 12 and Examples 30 to 38, it is apparent that the treating material comprising the water glass aqueous solution and the additive A as the main structural components, and the treating material comprising the above treating material and having added thereto any of gluconates, tartarates, benzoates, ligninsulfonates, polysaccharides, and bases comprising monovalent cations and hydroxide ions, as the additive E do not have substantially clear difference when comparing the Pb stabilizing performance, but the one part stability of the treating material is apparently improved; and from Examples 36 to 38, if sodium hydroxide, potassium hydroxide or aqueous ammonia is preferably used as the additive E, the one part stability is further improved as compared with other one part stabilizing agents, and such a treating material has excellent Pb stabilizing performance as compared with the treating material having other stabilizing agents added thereto.

From the comparison between Examples 12 and 30 and Comparative Example 5, it is apparent that where the amount of the additive E added as the one part stabilizing agent exceeds 20 parts by weight per 100 parts by weight of the solid content (the sum of the amount of M_2O and SiO_2) of the water glass aqueous solution, gel is instantaneously formed, the one part stability of such a treating material is markedly decreased as compared with the treating material to which the stabilizing agent is not added; on the other hand, where the amount of the additive E added is 20 parts by weight or less per 100 parts by weight of the solid content of the water glass aqueous solution, the one part stability is improved without decreasing the Pb stabilizing performance of the treating material.

From the comparison between Examples 39 to 46 and Comparative Example 2, it is apparent that the Pb stabilizing performance by the treatment method comprising separately storing the water glass aqueous solution, and acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic esters, dialdehydes, and the like as the additive A, mixing those immediately before adding to the waste, and adding the resulting mixture is higher than the performance of the treatment method using only the water glass aqueous solution.

From the comparison between Examples 47 to 54 and Comparative Example 6, it is apparent that the Pb stabilizing performance by the treatment method comprising separately storing the water glass aqueous solution, and acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic esters, dialdehydes, and the like as the additive A, adding the water glass aqueous solution to the waste, and then adding the additive A to the waste shows excellent Pb stabilizing performance as compared with the method of adding the additive A to the waste, and then adding the water glass aqueous solution to the waste.

From the comparison between Example 55 and Example 12, it is apparent that the Pb stabilizing performance is improved by conducting the aging at 40°C or more.

From the comparison between Examples 12 and 55 to 57 and Comparative Example 7, it is apparent that the Pb stabilizing performance is markedly improved by conducting the aging for 6 hours or more, and further, it is possible to maintain the excellent Pb stabilizing performance by setting the aging temperature to higher temperature regardless of short aging time; however, if aging is conducted at a temperature exceeding 100°C, the solidified product becomes dry,

resulting in remarkable decrease in the Pb stabilizing performance; therefore, it is preferred to conduct the aging at a temperature of 100°C or less.

Thus, by adding the additive A to the water glass aqueous solution, the elution amount of Pb can be decreased as compared with the case of using only the water glass aqueous solution. Further, the present inventors recognize that by appropriately setting the amount of the treating material added, the aging time after kneading the waste and the treating material, the temperature during aging, and the like, the treating material and the treatment method as described above are capable of decreasing the elution amount of Pb to 0.3 ppm or less which is the standard value for filling-up.

COMPARATIVE EXAMPLE 8

The analytical results of the components of a fly ash (II) generated in municipal refuse incineration facility are shown in Table 5 below.

TABLE 5

Analytical Results of Components of Fly Ash (II) Generated in Municipal Refuse Incineration Facility (% by weight)	
	Analytical result
Ca(OH) ₂	11.0
CaCl ₂	9.5
CaO	5.7
Pb	0.33

The fly ash (II) containing the calcium compound was subjected to the elution test as described before without treatment.

COMPARATIVE EXAMPLE 9

26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I) to prepare a water glass aqueous solution which was used as a treating material. The treating material was added to 50 g of the above fly ash (II), and kneaded. The kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

COMPARATIVE EXAMPLE 10

The elution test was conducted in the same manner as in Comparative Example 9 except that the amount of the sodium silicate aqueous solution (I) used was changed to 9.1 g (solid content: 3.5 g) (the solid content of the water glass aqueous solution was 7 parts by weight per 100 parts by weight of the ash).

COMPARATIVE EXAMPLE 11

The elution test was conducted in the same manner as in Comparative Example 9 except that the amount of the sodium silicate aqueous solution (I) used was changed to 13 g (solid content: 5.0 g) (the solid content of the water glass aqueous solution was 10 parts by weight per 100 parts by weight of the ash).

COMPARATIVE EXAMPLE 12

An aqueous solution prepared by adding 0.73 g of sulfuric acid to 26.0 g of water (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution) was added to 9.1 g (solid content: 3.5 g) of the sodium silicate solution (I) to prepare a treating material. After 2 minutes from the preparation of the treating material, the treating material was tried to add to the fly ash (II). However, the treating material was solidified, and

it was impossible to add the treating material to the fly ash and knead those.

COMPARATIVE EXAMPLE 13

5 A treating material was prepared in the same manner as in Comparative Example 12 except for using 13 g (solid content: 5.0 g) of the sodium silicate solution (I) and 1.0 of sulfuric acid. Immediately after adding the sulfuric acid aqueous solution to the water glass aqueous solution, the treating material was solidified, and it was impossible to add the treating material to the fly ash (II) and knead those.

10 EXAMPLE 61

An aqueous solution prepared by adding 0.52 g of sulfuric acid to 26.0 g of water (hydrogen ion of sulfuric acid was 50 parts by mol per 100 parts by mol of cations in the water glass aqueous solution) was added to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I) to prepare a treating material. After preparation of the treating material, 15 the treating material was instantaneously added to 50 g of the fly ash (II), and kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

20 EXAMPLE 62

The elution test was conducted in the same manner as in Example 61 except the after 5 minutes from the preparation of the treating material, the treating material was added to the fly ash (II).

25 Example 63

The elution test was conducted in the same manner as in Example 61 except for using 9.1 g (solid content: 3.5 g) of the sodium silicate aqueous solution (I) and 0.73 g of sulfuric acid.

30 EXAMPLE 64

An aqueous solution prepared by adding 26.0 g of water to 13.0 g (solid content: 5.0 g) of the sodium silicate aqueous solution (I) was used as a treating material. The treating material was added to 50 g of the fly ash (II), and kneaded. 35 1.0 g of sulfuric acid was further added to the kneaded mixture, and the kneaded. The resulting kneaded product was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

The elution test results obtained in Comparative Examples 8 to 11 and Examples 61 to 64 above are shown in Table 6 below.

TABLE 6

Elution Test Result	
	Pb elution amount (ppm)
Comparative Example 8	50
Comparative Example 9	10
Comparative Example 10	5.3
Comparative Example 11	3.5
Comparative Example 61	1.3
Comparative Example 62	0.77
Comparative Example 63	0.41
Comparative Example 64	0.26

From the comparison between Comparative Example 9 and Examples 61 and 62, it is shown that the method for treating waste according to the present invention is effective. Further, it is apparent that where the product after 5 minutes or more from the addition of sulfuric acid is used as the treating material, the performance is improved.

From the comparison between Comparative Examples 10 and 12 and Example 63, it is apparent that the treatment method of the present invention is effective to the product wherein after addition of sulfuric acid, the treating material does not instantaneously solidify, but the treating material solidifies as a whole after several minutes after the addition.

From the comparison between Comparative Examples 11 and 13 and Example 64, it is apparent that the method for treating waste of the present invention is effective to the product wherein after addition of sulfuric acid, the treating material as a whole solidifies instantaneously.

COMPARATIVE EXAMPLE 14

The analytical results of components of a bag filter collected fly ash (III) (hereinafter referred to as "fly ash (III)) generated in municipal refuse incineration facility are shown in Table 7 below. The elution test was conducted using the fly ash (III) containing calcium compound without treatment.

TABLE 7

Analysis of Components of Fly Ash (III) generated in Municipal Refuse Incineration Facility (% by weight)	
Component	Analytical result
Ca(OH)_2	26.7
$\text{CaCl}_2 \cdot \text{Ca(OH)}_2 \cdot \text{H}_2\text{O}$	32.6
Pb	0.16

COMPARATIVE EXAMPLE 15

26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I) to prepare a water glass aqueous solution which was used as a treating material. The treating material was added to 50 g of the above fly ash (III), and kneaded. The kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

COMPARATIVE EXAMPLE 16

The elution test was conducted in the same manner as in Comparative Example 2 except that the amount of the sodium silicate solution (I) used was changed to 9.1 g (solid content: 3.5 g) (the solid content of the water glass aqueous solution was 7 parts by weight per 100 parts by weight of the ash).

COMPARATIVE EXAMPLE 17

The elution test was conducted in the same manner as in Comparative Example 15 except the the aging time was changed to 7 days (168 hours).

The elution test results obtained in Comparative Examples 14 to 17 above are shown in Table 8 below.

TABLE 8

Pb Elution Amount (ppm)	
Comparative Example 14	16
Comparative Example 15	2.9
Comparative Example 16	2.2
Comparative Example 17	0.30

EXAMPLE 65

1.0 g of tripotassium phosphate as the additive B was added to an aqueous solution prepared by adding 26.0 g of water to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I) to prepare a treating material (the amount of tripotassium phosphate was 40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution). The treating material was added to 50 g of the above fly ash (III), and kneaded. The kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

EXAMPLES 66 TO 96

The elution test was conducted in the same manner as in Example 65 except the the following compound was used in place of tripotassium phosphate as the additive B.

Example 66: 0.15 g of tripotassium phosphate (6 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 67: 0.25 g of tripotassium phosphate (10 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 68: 10.0 g of tripotassium phosphate (400 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 69: 12.5 g of tripotassium phosphate (500 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 70: 1.0 g of pentasodium triphosphate (sodium tripolyphosphate) (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 71: 1.0 g of hexasodium tetrphosphate (sodium tripolyphosphate) (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 72: 1.0 g of sodium hexametaphosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 73: 1.0 g of potassium carbonate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 74: 1.0 g of ammonium sulfate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 75: 1.0 g of potassium oxalate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 76: 1.0 g of potassium ethylene diamine tetraacetate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 77: 1.0 g of sodium ethylene diamine tetraacetate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 78: 1.0 g of sodium dimethyldithiocarbamate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 79: 1.0 g of potassium 1-hydroxyethane-1,1-diphosphonate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 80: 1.0 g of sodium 1-hydroxyethane-1,1-diphosphonate (40 parts by weight per 100 parts by weight of

the solid content of the water glass aqueous solution)

Example 81: 1.0 g of trisodium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 82: 1.0 g of tetrapotassium diphosphate (potassium pyrophosphate) (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 83: 1.0 g of sodium carbonate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 84: 1.0 g of potassium sulfate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 85: 1.0 g of sodium oxalate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 86: 1.0 g of potassium aminotrimethylenesulfonate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 87: 3.5 g of potassium carbonate (140 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 88: 3.5 g of potassium oxalate (140 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 89: 7.0 g of potassium oxalate (280 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 90: 3.5 g of sodium ethylene diamine tetraacetate (140 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

EXAMPLE 91

The elution test was conducted in the same manner as in Example 61 except that 6.3 g (solid content: 2.5 g) of a potassium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: A POTASSIUM SILICATE, solid content: 40.0%, $\text{SiO}_2/\text{K}_2\text{O}$ compositional ratio = 2.0) was used in place of the sodium silicate aqueous solution (I).

EXAMPLE 92

The elution test was conducted in the same manner as in Example 65 except that 8.2 g (solid content: 2.5 g) of a sodium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: SODIUM SILICATE No.4, solid content: 30.5%, $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio = 3.8) was used as the sodium silicate aqueous solution (I).

EXAMPLE 93

The elution test was conducted in the same manner as in Example 65 except that 7.9 g (solid content: 2.5 g) of a sodium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: 30% LIQUID ORTHO, solid content: 31.5%, $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio = 0.50) was used as the sodium silicate aqueous solution.

EXAMPLE 94

The elution test was conducted in the same manner as in Example 65 except that 4.6 g (solid content: 2.5 g) of a sodium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: J SODIUM SILICATE No. 1, solid content: 54.5%, $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio = 2.0) was used as the sodium silicate aqueous solution.

EXAMPLE 95

7.0 g of potassium oxalate (280 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B was added to an aqueous solution prepared by adding 26.0 g of water to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I), and 0.03 g of potassium tartrate (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive E was further added thereto, to prepare a treating material. The treating material was added to 50 g of the above fly ash (III), and kneaded. The kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test. Further, gelation of the above treating material was visually observed to examine the one part stability.

EXAMPLES 96 TO 102

The elution test was conducted in the same manner as in Example 88 except that the following compound was used in place of potassium tartrate as the additive E. Further, gelation of the treating material was visually observed to examine the one part stability.

Example 96: 0.03 g of potassium gluconate (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 97: 0.03 g of potassium benzoate (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 98: 0.03 g of potassium ligninsulfonate (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 99: 0.03 g of starch (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 100: 0.03 g of sodium hydroxide (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 101: 0.03 g of potassium hydroxide (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 102: 0.12 g of aqueous ammonia (25% aqueous solution) (4.8 parts by weight (1.2 parts by weight in terms of ammonia) per 100 parts by weight of the solid content of the water glass aqueous solution)

EXAMPLES 103 TO 109

The elution test was conducted in the same manner as in Example 75 except that the aging conditions were changed as follows.

Example 103: The aging temperature was 60°C

Example 104: The aging temperature was 90°C

Example 105: The aging time was 4 hours

Example 106: The aging time was 6 hours

Example 107: The aging time was 8 hours

Example 108: The aging time was 48 hours

Example 109: The aging temperature was 90°C, and the aging time was 48 hours.

EXAMPLE 110

The elution test was conducted in the same manner as in Example 75 except that the aging temperature was 120°C and the aging time was 48 hours. However, in this case, the solidified product was in a dry state, that is, in the state that it was liable to scatter.

EXAMPLE 111

When 7.0 g of ammonium sulfate (280 parts by weight per 100 parts by weight of the glass aqueous solution) as the additive B was added to an aqueous solution prepared by adding 26.0 g of water to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I), precipitates were instantaneously formed. This mixture containing the precipitates was used as a treating material. The treating material was added to 50 g of the above fly ash (III), and kneaded. The kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and was subjected to the elution test.

EXAMPLE 112

An aqueous solution prepared by adding 26.0 g of water to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I) was added to 50 g of the above fly ash (III), and then kneaded (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash), and 7.0 g of ammonium sulfate (280 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was additionally added thereto, and then kneaded. The kneaded product was aged at 20°C for 24 hours to solidify. After completion of the solidification, the solidified product was pulverized, and classified with a sieve having an opening of 5 mm. 30 g of particles was dis-

pensed from the particles which passed through the sieve, and was subjected to the elution test.

EXAMPLE 113

5 The elution test was conducted in the same manner as in Example 94 except that the additive E was changed to 0.50 g of potassium benzoate (20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution). Further, gelation of the treating material was visually observed to examine the one part stability.

EXAMPLE 114

10

The elution test was conducted in the same manner as in Example 94 except that the additive E was changed to 0.65 g of potassium benzoate (26 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution). Further, gelation of the treating material was visually observed to examine the one part stability.

15 The elution test results obtained in Examples 65 to 114 are shown in Table 9 below. Further, the one part stability is shown in Table 10 below.

TABLE 9

Elution Amount of Pb (ppm)					
Example 65	0.62	Example 66	1.5	Example 67	0.87
Example 68	0.28	Example 69	1.3	Example 70	0.69
Example 71	0.57	Example 72	0.55	Example 73	0.60
Example 74	0.77	Example 75	0.69	Example 76	0.75
Example 77	0.78	Example 78	0.70	Example 79	0.70
Example 80	0.72	Example 81	0.78	Example 82	0.80
Example 83	0.75	Example 84	0.82	Example 85	0.75
Example 86	0.85	Example 87	0.41	Example 88	0.57
Example 89	0.51	Example 90	0.72	Example 91	0.75
Example 92	0.60	Example 93	0.97	Example 94	0.65
Example 95	0.50	Example 96	0.53	Example 97	0.55
Example 98	0.53	Example 99	0.55	Example 100	0.55
Example 101	0.55	Example 102	0.55	Example 103	0.44
Example 104	0.40	Example 105	0.87	Example 106	0.75
Example 107	0.73	Example 108	0.47	Example 109	0.35
Example 110	1.0	Example 111	0.71	Example 112	0.52
Example 113	0.58	Example 114	0.72		

TABLE 10

One Part Stability of Treating Material (gelation time of treating material)			
Comparative Example 14	0 hour	Example 89	85 hours
Example 95	180 hours	Example 96	190 hours
Example 97	200 hours	Example 98	200 hours
Example 99	120 hours	Example 100	240 hours or more
Example 101	240 hours or more	Example 102	240 hours or more
Example 113	150 hours	Example 114	40 second

From the comparison of Comparative Examples 14 to 16, it was proved that by adding the water glass aqueous solution to the fly ash, the Pb elution amount is decreased as compared with the non-treatment, and therefore, the water glass aqueous solution has the performance to stabilize harmful metals (Pb). However, the Pb stabilizing performance is not still insufficient by this method.

From the comparison between Comparative Examples 14 and 15 and Examples 73, 75, 77 and 87 to 90, it is apparent that the amount of the additive B is preferably 10 to 400 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution in view of the Pb stabilizing performance.

From the comparison between Examples 65 to 86 and Comparative Examples 15 and 16, it is apparent that by adding as the additive B materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water, ion sealing agents which seal calcium ions, chelating agents, or the like to the water glass aqueous solution, the treating material has excellent Pb stabilizing performance as compared with the case of using only the water glass aqueous solution.

From the comparison between Example 65 and Example 91, it is apparent that as the water glass in the treating material of the present invention, the treating material water glass aqueous solution comprising sodium silicate ($\text{Na}_2\text{O} \cdot n\text{SiO}_2$) as the main structural component has excellent Pb stabilizing performance as compared with potassium silicate ($\text{K}_2\text{O} \cdot n\text{SiO}_2$), and is more preferable embodiment as the treating material.

From the comparison of Examples 65 and 92 to 94, it is apparent that sodium silicate which is the main structural component of the water glass aqueous solution wherein $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio is 2.00 or more has excellent Pb stabilizing performance.

From Examples 89 and 95 to 102, it is apparent that by adding any of gluconates, tartrate, benzoates, ligninsulfonates, polysaccharides or bases comprising monovalent cations and hydroxide ions as the additive E to the treating material comprising the water glass aqueous solution and the additive B as the main structural components, the one part stability of the treating material is improved without decreasing the Pb stabilizing performance. In particular, from Examples 99 to 101, it is apparent that if sodium hydroxide, potassium hydroxide or aqueous ammonia is used as the additive E, the one part stability of the treating material is further improved as compared with other additives.

From Examples 97, 107 and 108, it is apparent that where the amount of the additive E added is 20 parts by weight or less per 100 parts by weight of the solid content of the water glass aqueous solution, the additive acts as the one part stabilizing agent, but if the amount thereof exceeds 20 parts by weight, the additive does not act as the stabilizing agent.

From Examples 75 and 103 to 110, it is apparent that if the aging is conducted at a temperature of 40 to 100°C or if the aging is conducted for 6 hours or more, the Pb stabilizing performance is further improved.

From Examples 111 and 112, it is apparent that where gel is formed at the time of mixing the additive B with the water glass aqueous solution, if the water glass aqueous solution is added to the waste and the additive B is then separately added thereto rather than using the mixed solution as it is as the treating material, the Pb stabilizing performance is excellent.

From Comparative Example 15 and Comparative Example 17, it is possible to decrease the Pb elution amount to 0.30 ppm or less which is the amount defined by the regulation, with only the water glass aqueous solution by sufficiently aging the waste. Therefore, it is apparent that the Pb elution amount is further decreased by sufficiently aging even in Examples 65 to 114. As a result, the waste-treating material comprising the water glass aqueous solution and the additive B and the treatment method using the waste treating material can sufficiently be applied to the rule-regulated value of Pb.

COMPARATIVE EXAMPLE 18

The analytical results of Pb content and Ca content in bag filter collected fly ashes (IV), (V) and (VI) generated in municipal refuse incineration facility are shown in Table 11. The elution test was conducted using those fly ashes in the non-treated state, and the Pb elution amount was measured. The results obtained are also shown in Table 11 below.

TABLE 11

Component Analysis of Fly Ash Generated in Municipal Refuse Incineration Facility (% by weight)			
Kind of fly ash	Pb content (mg/kg)	Ca content (mg/kg)	Pb non-treated elution amount (ppm)
Fly Ash (IV)	6,800	220,000	240
Fly Ash (V)	1,400	250,000	35
Fly Ash (VI)	2,500	260,000	97

COMPARATIVE EXAMPLES 19 AND 20 AND EXAMPLES 115 TO 120

A crude solution of the sodium silicate aqueous solution (I) and a sodium carbonate aqueous solution (solid content: 30.8%) at 70°C were mixed in the blending ratio as shown in Table 10 to prepare a treating material. 20 g of water was added to each of the treating material, and kneaded such that the sum (solid content) of the solid content of the water glass aqueous solution and the solid content of sodium carbonate contained in the treating material aqueous solution was 0.5 g per 50 g of the fly ash (V). The kneaded product was aged at 20°C for 24 hours. After aging, the product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test. The results obtained are shown in Table 12 below.

TABLE 12

Elution Test Result			
Type of treating material	Treating material (parts by weight)		Pb elution amount (ppm)
	Water glass solid content	Na ₂ CO ₃ solid content	
Comparative example 19	0	100	7.0
Example 115	20	80	1.4
Example 116	40	60	0.50
Example 117	50	50	0.29
Example 118	60	40	0.25
Example 119	90	10	0.78
Example 120	95	5	1.3
Comparative example 20	100	0	1.8

EXAMPLE 121

The treating material of Example 117 was evaluated for the storage stability at 6 to 70°C. The evaluation was conducted using two methods of evaluation method 1 and evaluation method 2. The evaluation method 1 was that 150 g of the treating material was allowed to stand at each temperature for 40 days, precipitation of salt, and formation of gel were visually observed, and the results were recorded. The evaluation method 2 was that the treating material was

allowed to stand one day at each temperature, then 0.3 g of sodium carbonate solid powder was added to 150 g of the treating material, the resulting mixture was allowed to stand for 6 hours, formation of gel was visually observed, and the results were recorded. The results obtained are shown in Table 13 below.

TABLE 13

Storage Stability of Treating Material		
Temperature (°C)	Evaluation method 1	Evaluation method 2
6	Precipitation of salt observed	No data
15	No precipitation of salt, and no formation of gel	Precipitation of salt observed
20	No precipitation of salt, and no formation of gel	Precipitation of salt observed
25	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel
30	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel
40	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel
70	Formation of gel observed	No data

From Comparative Examples 18 to 20 and Examples 115 to 120, it is understood that the treating material comprising a mixed solution of the water glass aqueous solution and sodium carbonate is extremely effective for preventing the Pb elution in the waste. Further, it is understood that a blend wherein the weight ratio of the water glass solid content and the sodium carbonate solid content in the mixed solution is 90 : 10 to 40 : 60 shows excellent Pb elution preventing performance. It is also apparent from the above examples that the optimum mixing ratio varies depending on types of the waste, but it is understood that on the average, the mixed solution wherein the weight ratio of the water glass solid content and the sodium carbonate solid content is 60 : 40 to 50 : 50 is most preferred.

On the other hand, it is shown in Example 121 to be necessary to maintain a temperature of at least 25°C, but it is understood that it is necessary to maintain a temperature of 30°C to less than 70°C, considering reliability from the industrial standpoint.

COMPARATIVE EXAMPLES 21 AND 22 AND EXAMPLES 122 TO 127

A crude solution of the sodium silicate aqueous solution (I) and a sodium hydroxide aqueous solution (solid content: 45.5 %) at 70°C were mixed in the blending ratio as shown in Table 14 to prepare a treating material. 23 g of water was added to each of the treating material, and kneaded such that the sum (solid content) of the solid content of the water glass aqueous solution and the solid content of sodium hydroxide contained in the treating material aqueous solution was 5 g per 50 g of the fly ash (IV). The kneaded product (the solid content of the treating material was 10 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours. After aging, the product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test. The results obtained are shown in Table 14 below.

TABLE 14

Elution Test Result			
Type of treating material	Treating material (parts by weight)		Pb elution amount (ppm)
	Water glass solid content	Na ₂ CO ₃ solid content	
Comparative Example 21	0	100	110
Example 122	20	80	14
Example 123	40	60	6.3
Example 124	50	50	6.2
Example 125	60	40	5.9
Example 126	70	30	6.6
Example 127	90	10	8.5
Comparative Example 22	100	0	16

EXAMPLE 128

A treating material having blended therewith potassium hydroxide in an amount of 67 parts by weight per 100 parts by weight of the water glass solid content was prepared in the same manner as in Example 125 except that potassium hydroxide was used in place of sodium hydroxide, and was evaluated in the same manner as in Example 125. As a result, the Pb elution amount was 6.00 ppm.

From Comparative Examples 18 and 21 and Examples 122 and 127, it is understood that the treating material comprising a mixed aqueous solution of the water glass aqueous solution and the hydroxide as the additive B according to the present invention is extremely effective for preventing the Pb elution in the waste. Further, it is understood that a blend wherein the weight ratio of the water glass solid content and the hydroxide solid content is 90 : 10 to 40 : 60 shows excellent Pb preventing performance. Furthermore, although it is sufficiently expected that the optimum blending ratio varies depending on types of the waste, it is understood from those examples that the blend wherein the weight ratio of the water glass solid content and the hydride solid content is 60 : 40 is most preferred.

COMPARATIVE EXAMPLES 23 AND 24 AND EXAMPLES 129 TO 133

A crude solution of the sodium silicate aqueous solution (I) and a potassium carbonate aqueous solution (solid content: 36.6%) at 40°C were mixed in the blending ratio as shown in Table 15 to prepare a treating material. 20 g of water was added to each of the treating material, and kneaded such that the sum (solid content) of the solid content of the water glass aqueous solution and the solid content of potassium carbonate contained in the treating material aqueous solution was 5.0 g per 50 g of the fly ash (V). The kneaded product (the solid content of the treating material was 15 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours. After aging, the product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test. The results obtained are shown in Table 15 below.

TABLE 15

Elution Test Result			
Type of treating material	Treating material (parts by weight)		Pb elution amount (ppm)
	Water glass solid content	Na ₂ CO ₃ solid content	
Comparative Example 23	0	100	150
Example 129	20	80	8.3
Example 130	40	60	2.7
Example 131	60	40	1.4
Example 132	90	10	2.8
Example 133	95	5	7.8
Comparative Example 24	100	0	10

EXAMPLE 134

The treating material of Example 131 was evaluated for the storage stability at 0 to 30°C. The evaluation was conducted using two methods of evaluation method 1 and evaluation method 2 in the same manner as in Example 121. The results obtained are shown in Table 16 below.

TABLE 16

Storage Stability of Treating Material		
Temperature (°C)	Evaluation method 1	Evaluation method 2
0	Precipitation of salt observed	Precipitation of salt increased
5	Slight precipitation of salt, and slight formation of gel	No precipitation of salt, and no formation of gel
10	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel
20	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel
30	No precipitation of salt, and no formation of gel	No precipitation of salt, and no formation of gel

From Comparative Examples 18, 23 and 24 and Examples 129 to 133, it is understood that the treating material comprising a mixed solution of the water glass aqueous solution and potassium carbonate is extremely effective for preventing the Pb elution in the waste. Further, it is understood that a blend wherein the weight ratio of the water glass solid content and the potassium carbonate solid content in the mixed solution is 90 : 10 to 40 : 60 shows particularly excellent Pb elution preventing performance.

On the other hand, from the results of the evaluation method 1 of Example 134, precipitation of salt and formation of gel were slightly observed at 5°C or less by allowing to stand for 40 days, but in the evaluation method 2, the results were substantially the same as in the results at 10°C or more. Where the treating material is industrially used, it is preferred to maintain the same at a temperature of 10° or more, but even if the temperature is 5°C, disadvantage does not particularly occur.

COMPARATIVE EXAMPLE 25

An aqueous solution prepared by dissolving 1.58 g of hydrochloric acid (36% HCl) (hydrogen ion (H⁺) of hydrochloric acid was 75 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) in

26.0 g of water was added to the sodium silicate aqueous solution (I). Gel was instantaneously formed, and it was impossible to add the resulting solution to the fly ash (I) and knead those.

COMPARATIVE EXAMPLE 26

An aqueous solution prepared by dissolving 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution, and 0.75 g of sodium gluconate (30 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was added thereto. Gel was instantaneously formed, and it was impossible to add the resulting solution to the fly ash (I) and knead those.

COMPARATIVE EXAMPLE 27

Hydrochloric acid (4.20 g of 36% HCl was dissolved in 20.0 g of water) and a tripotassium phosphate aqueous solution (1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was dissolved in 6.0 g of water), which were separately stored, were simultaneously added to 50 g of the fly ash (I), and mixed, and 6.5 g of the sodium silicate solution (I) (solid content: 2.5 g, the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was further added thereto, and kneaded. The resulting kneaded product was aged, solidified, classified with a sieve, and then subjected to the elution test in the same manner as in Comparative Example 2.

COMPARATIVE EXAMPLE 28

An aqueous solution prepared by dissolving 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I). After 5 minutes from the addition, the resulting solution was added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 120°C for 6 hours to solidify. The solidified product was in a dry state, that is, in the state that the product was liable to scatter. The solidified product was classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and the subjected to the elution test.

The elution test results obtained in Comparative Examples 27 and 28 are shown in Table 17 below.

TABLE 17

Pb Elution Amount (ppm)	
Comparative Example 27	140
Comparative Example 28	78

EXAMPLE 135

An aqueous solution prepared by dissolving 1.05 g of hydrochloric acid (36% HCl) (hydrogen ion (H^+) of hydrochloric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution. The resulting solution (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product was aged, solidified, classified with sieve, and then subjected to the elution test in the same manner as in Comparative Example 2.

EXAMPLE 136

The elution test was conducted in the same manner as in Example 135 except that 0.48 g of ethanol was added in

place of hydrochloric acid.

EXAMPLE 137

5 The elution test was conducted in the same manner as in Example 135 except that 0.97 g of phenol was added in place of hydrochloric acid.

EXAMPLE 138

10 A calcium chloride solution (0.57 g of CaCl_2 was dissolved in 20.0 g of water) as the additive A was added to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I), and the resulting solution (the product of the amount of calcium ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) was allowed to stand for 5 minutes. A tripotassium phosphate aqueous solution (1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was dissolved in 6.0 g of water) as the additive B was added to the solution to prepare a treating material. The treating material was added to 50 g of the fly ash (I) and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 139 TO 146

25 The elution test was conducted in the same manner as in Example 131 except that the following compound was used in place of hydrochloric acid as the additive A.

Example 139: 0.57 g of ethylene glycol diacetate

Example 140: 1.00 g of ethylene carbonate

Example 141: 1.00 g of γ -butyrolactone

30 Example 142: 1.00 g of succinic acid dimethyl ester

Example 143: 1.00 g of glyoxal

Example 144: 1.07 g of nitric acid (61% HNO_3)

Example 145: 0.40 g of phosphoric acid (85% H_3PO_4) (hydrogen ion (H^+) of phosphoric acid was 50 parts by mol per 100 parts by mol of cation of the water glass aqueous solution)

35 Example 146: 1.00 g of dry ice as a carbonic acid source

EXAMPLE 147

40 An aqueous solution prepared by dissolving sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate aqueous solution (I), and the resulting solution was allowed to stand for 5 minutes to prepare a treating material. The treating material was added to 50 g of the fly ash (I) and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above. Further, gelation of the treating material was visually observed to examine the one part stability. The results obtained are shown in Table 23 below.

EXAMPLE 148

50 The elution test was conducted in the same manner as in Example 135 except that a treating material prepared by adding 0.62 g of acetic acid in place of hydrochloric acid as the additive A was used (hydrogen ion (H^+) of acetic acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution).

EXAMPLE 149

The elution test was conducted in the same manner as in Example 135 except that a treating material prepared by

adding 0.47 g of oxalic acid (98% $(\text{COOH})_2$) in place of hydrochloric acid as the additive A was used (hydrogen ion (H^+) of oxalic acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution).

The elution test results obtained in Examples 135 to 149 above are shown in Table 18 below.

TABLE 18

Pb Elution Amount (ppm)			
Example 135	2.5	Example 136	18
Example 137	27	Example 138	5.3
Example 139	18	Example 140	13
Example 141	21	Example 142	20
Example 143	16	Example 144	2.1
Example 145	6.2	Example 146	21
Example 147	3.4	Example 148	1.7
Example 149	2.7		

EXAMPLE 150

An aqueous solution prepared by dissolving 1.05 g of hydrochloric acid (36% HCl) (hydrogen ion (H^+) of hydrochloric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution. The resulting solution was immediately added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged, solidified, classified with sieve, and then subjected to the elution test in the same manner as in Comparative Example 2.

EXAMPLES 151 TO 156

The elution test was conducted in the same manner as in Example 150 except that the treating material was prepared using the following material in place of hydrochloric acid as the additive A.

Example 151: 1.07 g of nitric acid (61% HNO_3) (hydrogen ion (H^+) of nitric acid was 50 parts by mol per 100 mol of cations of the water glass aqueous solution)

Example 152: 0.40 g of phosphoric acid (85% H_3PO_4) (hydrogen ion (H^+) of phosphoric acid was 50 parts by mol per 100 mol of cations of the water glass aqueous solution)

Example 153: 1.00 g of dry ice as a carbonic acid source

Example 154: 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 mol of cations of the water glass aqueous solution)

Example 155: 0.62 g of acetic acid (hydrogen ion (H^+) of acetic acid was 50 parts by mol per 100 mol of cations of the water glass aqueous solution)

Example 156: 0.47 g of oxalic acid (98% $(\text{COOH})_2$) (hydrogen ion (H^+) of oxalic acid was 50 parts by mol per 100 mol of cations of the water glass aqueous solution)

EXAMPLE 157

A zinc sulfate aqueous solution (0.83 g of zinc sulfate was dissolved in 20.0 g of water) as the additive A was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I), and the resulting solution (the product of the amount of zinc ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) was allowed to stand for 5 minutes. A tripotassium phosphate aqueous solution (1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was dissolved in 6.0 g of water) as the additive B was added to the solution to prepare a treating material. The treating material was

added to 50 g of the fly ash (I) and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 158 TO 169

The elution test was conducted in the same manner as in Example 154 except that the following solution was used in place of the zinc sulfate aqueous solution as the additive A.

Example 158: Magnesium chloride aqueous solution (0.49 g of magnesium chloride was dissolved in 20.0 g of water) (the product of the magnesium ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 159: Iron (II) chloride aqueous solution (0.65 g of iron (II) chloride was dissolved in 20.0 g of water) (the product of the iron (II) ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 160: Iron (III) chloride aqueous solution (0.56 g of iron (III) chloride was dissolved in 20.0 g of water) (the product of the iron (III) ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 161: Aluminum chloride aqueous solution (0.46 g of aluminum chloride was dissolved in 20.0 g of water) (the product of the aluminum ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 162: Magnesium nitrate aqueous solution (0.76 g of magnesium nitrate was dissolved in 20.0 g of water) (the product of the magnesium ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 163: Calcium nitrate aqueous solution (0.84 g of calcium nitrate was dissolved in 20.0 g of water) (the product of the calcium ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 164: Iron (II) nitrate aqueous solution (0.92 g of iron (II) nitrate was dissolved in 20.0 g of water) (the product of the iron (II) ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 165: Iron (III) nitrate aqueous solution (0.83 g of iron (III) nitrate was dissolved in 20.0 g of water) (the product of the iron (III) ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 166: Aluminum nitrate aqueous solution (0.73 g of aluminum nitrate was dissolved in 20.0 g of water) (the product of the aluminum ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 167: Magnesium sulfate aqueous solution (0.62 g of magnesium sulfate was dissolved in 20.0 g of water) (the product of the magnesium ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 168: Iron (II) sulfate aqueous solution (0.78 g of iron (II) sulfate was dissolved in 20.0 g of water) (the product of the iron (II) ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

Example 169: Aluminum sulfate aqueous solution (0.59 g of aluminum sulfate was dissolved in 20.0 g of water) (the product of the aluminum ion and the number of valency is 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution)

EXAMPLE 170

An aqueous solution prepared by dissolving 0.03 g of strontium carbonate as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution. The resulting solution was added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLE 171

The elution test was conducted in the same manner as in Example 170 except that a treating material obtained by adding 0.03 g of barium phosphate was used in place of strontium carbonate.

The elution test results obtained in Examples 150 to 171 above are shown in Table 19 below.

TABLE 19

Pb Elution Amount (ppm)			
Example 150	3.2	Example 151	2.9
Example 152	8.1	Example 153	29
Example 154	4.7	Example 155	2.6
Example 156	3.9	Example 157	22
Example 158	4.9	Example 159	3.2
Example 160	2.4	Example 161	1.3
Example 162	14	Example 163	11
Example 164	8.5	Example 165	7.8
Example 166	5.0	Example 167	14
Example 168	10	Example 169	4.7
Example 170	28	Example 171	27

EXAMPLE 172

Hydrochloric acid (2.10 g of 36% HCl was dissolved in 20.0 g of water) as the additive A and a tripotassium phosphate aqueous solution (1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was dissolved in 6.0 g of water) as the additive B were simultaneously added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I). The resulting solution was added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 173 TO 179

The elution test was conducted in the same manner as in Example 172 except that the following material was used in place of hydrochloric acid as the additive A.

Example 173: Ethanol (0.96 g of ethanol was dissolved in 20.0 g of water)

Example 174: Calcium chloride aqueous solution (1.04 g of calcium chloride was dissolved in 20.0 g of water)

Example 175: Ethylene glycol diacetate aqueous solution (1.04 g of ethylene glycol diacetate was dissolved in 20.0 g of water)

Example 176: Ethylene carbonate aqueous solution (2.00 g of ethylene carbonate was dissolved in 20.0 g of water)

Example 177: γ -Butyrolactone aqueous solution (2.00 g of γ -butyrolactone was dissolved in 20.0 g of water)

Example 178: Succinic acid dimethyl ester aqueous solution (2.00 g of succinic acid dimethyl ester was dissolved in 20.0 g of water)

Example 179: Glyoxal aqueous solution (2.00 g of glyoxal was dissolved in 20.0 g of water)

The elution test results obtained in Examples 172 to 179 above are shown in Table 20 below.

TABLE 20

Pb Elution Amount (ppm)			
Example 172	2.5	Example 173	25
Example 174	4.7	Example 175	14
Example 176	15	Example 177	21
Example 178	21	Example 179	17

EXAMPLE 180

Hydrochloric acid (4.20 g of 36% HCl was dissolved in 20.0 g of water) as the additive A and a tripotassium phosphate aqueous solution (1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was dissolved in 6.0 g of water) as the additive B were simultaneously added to a mixture prepared by adding 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I) to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 181 TO 187

The elution test was conducted in the same manner as in Example 180 except that the following material was used in place of hydrochloric acid as the additive A.

Example 181: Ethanol (1.92 g of ethanol was dissolved in 20.0 g of water)

Example 182: Calcium chloride aqueous solution (2.08 g of calcium chloride was dissolved in 20.0 g of water)

Example 183: Ethylene glycol diacetate aqueous solution (2.08 g of ethylene glycol diacetate was dissolved in 20.0 g of water)

Example 184: Ethylene carbonate aqueous solution (4.00 g of ethylene carbonate was dissolved in 20.0 g of water)

Example 185: γ -Butyrolactone aqueous solution (4.00 g of γ -butyrolactone was dissolved in 20.0 g of water)

Example 186: Succinic acid dimethyl ester aqueous solution (4.00 g of succinic acid dimethyl ester was dissolved in 20.0 g of water)

Example 187: Glyoxal aqueous solution (4.00 g of glyoxal was dissolved in 20.0 g of water)

The elution test results obtained in Examples 180 to 187 above are shown in Table 21 below.

TABLE 21

Pb Elution Amount (ppm)			
Example 180	3.2	Example 181	2.2
Example 182	5.7	Example 183	23
Example 184	18	Example 185	17
Example 186	25	Example 187	19

EXAMPLE 188

An aqueous solution prepared by dissolving 0.52 g of sulfuric acid (97% H₂SO₄) (hydrogen ion (H⁺) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous

solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I), and 0.03 g of sodium gluconate (1 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution) was further added thereto, to prepare a treating material. The treating material was added to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above. Further, gelation of the treating material was visually observed to examine the one part stability.

EXAMPLES 189 TO 196

The elution test was conducted in the same manner as in Example 188 except that the following material was used in place of sodium gluconate as the additive E. Further, gelation of the treating material was visually observed to examine the one part stability.

Example 189: 0.03 g of potassium tartrate (1 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 190: 0.03 g of ammonium benzoate (1 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 191: 0.03 g of sodium ligninsulfonate (1 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 192: 0.03 g of starch (1 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 193: 0.50 g of lithium hydroxide (20 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 194: 0.50 g of sodium hydroxide (20 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 195: 0.50 g of potassium hydroxide (20 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 196: 0.50 g of 25% aqueous ammonia (20 part by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

The elution test results obtained in Examples 188 to 196 are shown in Table 22 below. Further, the results of the one part stability are shown in Table 23 below.

TABLE 22

Pb Elution Amount (ppm)			
Example 188	2.2	Example 189	3.5
Example 190	3.6	Example 191	3.1
Example 192	3.7	Example 193	3.4
Example 194	3.0	Example 195	3.0
Example 196	3.9		

TABLE 23

One Part Stability of Treating Material (gelation time (min) of treating material)			
Example 147	320	Example 188	650
Example 189	670	Example 190	620
Example 191	700	Example 192	680
Example 193	720	Example 194	1,020
Example 195	980	Example 196	890

EXAMPLE 197

The elution test was conducted in the same manner as in Example 147 except that a treating material obtained using 6.3 g (solid content: 2.5 g) of a potassium silicate aqueous solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: A POTASSIUM SILICATE, solid content: 40.0%, $\text{SiO}_2/\text{K}_2\text{O}$ compositional ratio = 3.1) was used in place of sodium silicate B.

EXAMPLE 198

The elution test was conducted in the same manner as in Example 147 except that a sodium silicate aqueous solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: 30% LIQUID ORTHO, solid content: 31.5%, $\text{SiO}_2/\text{K}_2\text{O}$ compositional ratio = 0.50) was used in place of sodium silicate (I), and the amount of sulfuric acid (97% H_2SO_4) added was 0.69 g.

The elution test results obtained in Examples 197 and 198 above are shown in Table 24 below.

TABLE 24

Pb Elution Amount (ppm)			
Example 197	5.2	Example 198	8.5

EXAMPLE 199

An aqueous solution prepared by dissolving 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of tripotassium phosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I), and the resulting solution was allowed to stand for 5 minutes to prepare a treating material. The treating material was added to 50 g of the fly ash (I) and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 40°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 200 TO 204

The elution test was conducted in the same manner as in Example 198 except for changing the aging conditions as follows.

Example 200: The aging temperature was 60°C

Example 201: The aging temperature was 80°C

Example 202: The aging temperature was 60°C, and the aging time was 2 hours

Example 203: The aging temperature was 60°C, and the aging time was 6 hours

Example 204: The aging temperature was 60°C, and the aging time was 48 hours

The elution test results obtained in Examples 199 to 204 above are shown in Table 25 below.

TABLE 25

Pb Elution Amount (ppm)			
Example 199	1.6	Example 200	0.27
Example 201	0.11	Example 202	1.8
Example 203	0.30	Example 204	0.14

EXAMPLE 205

An aqueous solution prepared by dissolving 1.05 g of hydrochloric acid (36% HCl) (hydrogen ion (H⁺) of hydrochloric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) as the additive A and 1.0 g of pentapotassium triphosphate (40 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) as the additive B in 20.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I), and the resulting solution was allowed to stand for 5 minutes to prepare a treating material. The treating material was added to 50 g of the fly ash (I) and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 206 TO 226

The elution test was conducted in the same manner as in Example 205 except for using the following material in place of pentapotassium triphosphate as the additive B.

Example 206: 1.0 g of hexasodium tetrphosphate (sodium tetrapolyphosphate)

Example 207: 1.0 g of sodium hexametaphosphate

Example 208: 1.0 g of potassium carbonate

Example 209: 1.0 g of ammonium sulfate

Example 210: 1.0 g of potassium oxalate

Example 211: 1.0 g of potassium ethylene diamine tetraacetate

Example 212: 1.0 g of sodium ethylene diamine tetraacetate

Example 213: 1.0 g of sodium dimethyldithiocarbamate

Example 214: 1.0 g of sodium 1-hydroxyethane-1,1-diphosphonate

Example 215: 1.0 g of trisodium phosphate

Example 216: 1.0 g of tetrapotassium diphosphate (potassium pyromellitate)

Example 217: 1.0 g of sodium carbonate

Example 218: 1.0 g of potassium sulfate

Example 219: 1.0 g of sodium oxalate

Example 220: 1.0 g of potassium aminotrimethylenephosphonate

Example 221: 3.5 g of potassium carbonate (140 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 222: 7.0 g of potassium carbonate (280 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 223: 3.5 g of potassium oxalate (140 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 224: 7.0 g of potassium oxalate (280 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 225: 3.5 g of sodium ethylene diamine tetraacetate (140 parts by weight per 100 parts by weight of the

solid content of the aqueous solution)

Example 226: 7.0 g of sodium ethylene diamine tetraacetate (280 parts by weight per 100 parts by weight of the solid content of the aqueous solution)

The elution test results obtained in Examples 205 to 226 above are shown in Table 26 below.

TABLE 26

Pb Elution Amount (ppm)			
Example 205	2.7	Example 206	2.1
Example 207	2.0	Example 208	2.5
Example 209	2.8	Example 210	3.0
Example 211	3.7	Example 212	4.0
Example 213	3.8	Example 214	3.7
Example 215	3.2	Example 216	4.2
Example 217	3.8	Example 218	4.1
Example 219	3.9	Example 220	4.4
Example 221	1.8	Example 222	1.4
Example 223	2.4	Example 224	2.1
Example 225	3.5	Example 226	3.3

EXAMPLE 227

An aqueous solution prepared by dissolving 1.8 g of dipotassium hydrogen phosphate (hydrogen ion (H^+) of dipotassium hydrogen phosphate was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of dipotassium hydrogen phosphate was 72 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution) in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I). After 5 minutes, the resulting solution was added to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and then subjected to the elution test in the same manner as above.

EXAMPLES 228 TO 234

The elution test was conducted in the same manner as in Example 227 except for replacing dipotassium hydrogen phosphate with the following material.

Example 228: 0.70 g of potassium dihydrogen phosphate (hydrogen ion (H^+) of potassium dihydrogen phosphate was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of potassium dihydrogen phosphate was 28 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 229: 1.03 g of potassium hydrogen carbonate (hydrogen ion (H^+) of potassium hydrogen carbonate was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of potassium hydrogen carbonate was 41 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 230: 1.40 g of potassium hydrogen sulfate (hydrogen ion (H^+) of potassium hydrogen sulfate was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of potassium hydrogen sulfate was 56 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 231: 1.32 g of potassium hydrogen oxalate (hydrogen ion (H^+) of potassium hydrogen oxalate was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of potassium

hydrogen oxalate was 53 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 232: 0.40 g of phosphoric acid (85% H_3PO_4) (hydrogen ion (H^+) of phosphoric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of phosphoric acid was 16 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 233: 0.52 g of sulfuric acid (97% H_2SO_4) (hydrogen ion (H^+) of sulfuric acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of sulfuric acid was 21 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 234: 0.47 g of oxalic acid (98% $(\text{COOH})_2$) (hydrogen ion (H^+) of oxalic acid was 50 parts by mol per 100 parts by mol of cations of the water glass aqueous solution, and the amount of oxalic acid was 19 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

The elution test results obtained in Examples 227 to 234 above are shown in Table 27 below.

TABLE 27

Pb Elution Amount (ppm)			
Example 227	4.2	Example 228	3.8
Example 229	2.7	Example 230	4.5
Example 231	3.2	Example 232	16
Example 233	7.3	Example 234	5.8

From the comparison between Examples 135 to 198 and 205 to 234 and Comparative Example 2, it is apparent that the treating material according to the present invention comprising the water glass aqueous solution and having added thereto the additive A and the additive B has excellent Pb stabilizing performance as compared with the case of using only the water glass aqueous solution.

From the comparison between Example 135 and Comparative Example 25, it is apparent that the preferred embodiment as the treating material is such that the amount of the acid added which is the additive A in the treating material according to the present invention is such that hydrogen ion (H^+) of the acid is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

From the comparison between Examples 135 and 144 to 156 and Comparative Example 2, it is apparent that by using any of hydrochloric acid, nitric acid, phosphoric acid, sulfuric acid, carbonic acid, acetic acid and oxalic acid as the acid which is the additive A in the treating material according to the present invention, the treating material having excellent Pb stabilizing performance can be obtained.

From the comparison between Example 138 and Comparative Example 4, it is apparent that the preferred embodiment as the treating material is such that the amount of the polyvalent metal salts added which are the additive A in the treating material according to the present invention is such that the product of the part by mol of the polyvalent metal salts and the number of valency is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.

From the comparison between Examples 138 and 157 to 171 and Comparative Example 2, it is apparent that by using any of chlorides, nitrates, sulfates, carbonates and phosphates of magnesium, calcium, strontium, barium, iron, aluminum and zinc, preferably at least one of magnesium chloride, potassium chloride, iron (II) chloride, iron (III) chloride, aluminum chloride, magnesium nitrate, calcium nitrate, iron (II) nitrate, iron (III) nitrate, aluminum nitrate, magnesium sulfate, iron (II) sulfate and aluminum sulfate as the polyvalent metal salts which are the additive A in the treating material according to the present invention, the treating material having excellent Pb stabilizing performance can be obtained as compared with the case of using only the water glass aqueous solution.

From the comparison between Examples 205 to 226 and Comparative Example 2, it is apparent that by using materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water, or ion sealing agents which seal calcium ions, particularly at least one of tripotassium phosphate, pentasodium triphosphate, hexasodium tetrakisphosphate, sodium hexametaphosphate, potassium carbonate, ammonium sulfate, potassium oxalate, potassium 1-hydroxyethane-1,1-diphosphonate, sodium 1-hydroxyethane-1,1-diphosphonate, potassium ethylene diamine tetraacetate, sodium ethylene diamine tetraacetate and sodium dimethylthiocarbamate, which are the additive B in the treating material according to the present invention, the treating material having excellent Pb stabilizing performance can be obtained as compared with the case of using only the water glass aqueous solution.

From the comparison between Example 147 and Example 197, it is apparent that the water glass aqueous solution

which contains sodium silicate ($\text{Na}_2\text{O} \cdot n\text{SiO}_2$) as the main structural component has excellent Pb stabilizing performance as compared with potassium silicate ($\text{K}_2\text{O} \cdot n\text{SiO}_2$), and is the preferred embodiment as the treating material.

From the comparison between Example 147 and Example 198, it is apparent that where the compositional ratio of sodium silicate which is the main structural component of the water glass aqueous solution is compared between $\text{SiO}_2/\text{Na}_2\text{O} = 3.15$ and 0.50, the ratio of 3.15 has excellent Pb stabilizing performance, and is the preferred embodiment as the treating material.

From the comparison between Example 147 and Examples 188 to 196, it is apparent that if any of gluconates, tartrates, benzoates, ligninsulfonates, polysaccharides and bases comprising monovalent cations and hydroxide ions is additionally added as the additive E in the treating material of the present invention, the one part stability of the treating material is apparently improved without decreasing the Pb stabilizing performance; and more preferably, if sodium hydroxide, potassium hydroxide or aqueous ammonia is used as the additive E, the one part stability of the treating material is further improved as compared with other additives.

From the comparison between Example 188 and Comparative Example 26, it is apparent that where the amount of the one part stabilizing agent added exceeds 20 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution, gel is instantaneously formed, and the one part stability is markedly decreased as compared with the treating material to which a stabilizing agent is not added.

From the comparison between Examples 172 to 179 and Comparative Example 2, it is apparent that the Pb stabilizing performance of the method comprising separately storing the main structural components of the treating material according to the present invention, mixing those immediately before adding to the fly ash, and adding the mixture is superior to the performance of the water glass aqueous solution.

From the comparison between Examples 180 to 187 and Comparative Examples 2 and 27, it is apparent that the method for treating waste, which comprises separately storing the structural components of the treating material according to the present invention, adding the water glass aqueous solution to the fly ash, and then adding other components thereto shows excellent Pb stabilizing performance as compared with the method of adding components other than the water glass aqueous solution to the fly ash, and then adding the water glass aqueous solution thereto.

From the comparison between Examples 199 to 201 and Example 147, it is apparent that the Pb stabilizing performance is improved by aging at 40°C or more.

From the comparison between Examples 147 and 202 to 204 and Comparative Example 28, it is apparent that by conducting the aging for 6 hours or more, the Pb stability is markedly improved, and also by setting the aging temperature to higher temperature, excellent Pb stabilizing performance can be maintained despite that the aging time is short; however, if the aging is conducted at a temperature exceeding 100°C, the solidified product becomes dry, and the Pb stabilizing performance is markedly decreased; therefore, the aging is preferably conducted at a temperature of 100°C or less.

From Examples 227 to 234, it is apparent that hydrogen phosphate, hydrogen carbonates, hydrogen sulfates, hydrogen oxalates, phosphoric acid, sulfuric acid, oxalic acid, and the like act as the acid which is the additive A in the treating material according to the present invention, and also act as materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water, as the additive B, and therefore are effective for Pb stabilization.

COMPARATIVE EXAMPLE 29

An aqueous solution prepared by adding 26.0 g of water to 9.1 g (solid content: 3.5 g) of the sodium silicate (I) was added to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 7 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and the subjected to the elution test.

EXAMPLE 235

An aqueous solution prepared by dissolving 1.0 g of polyaluminum chloride (reagent) (40 parts by weight as the additive C per 100 parts by weight of the solid content of the water glass aqueous solution) in 26.0 g of water was added to 6.5 g (solid content: 2.5 g) of the sodium silicate solution (I). The resulting solution was added as a treating material to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged, solidified, pulverized, classified with a sieve, and the subjected to the elution test in the same manner as in Comparative Example 29.

EXAMPLES 236 TO 243

The elution test was conducted in the same manner as in Example 235 except for using the following material in

place of polyaluminum chloride as the additive C.

Example 236: 1.0 g of carbamate (SIMIFLOCK HM-2000, trade name, a product of Sumitomo Chemical Co., Ltd.)

Example 237: 1.0 g of polyacrylamide (SIMIFLOCK FA-50, trade name, a product of Sumitomo Chemical Co., Ltd.)

Example 238: 0.25 g of sodium polyacrylate (ARON A-20P, trade name, a product of TOA Gosei Chemical Industry Co., Ltd.) (10 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 239: 0.03 g of sodium polyacrylate (ARON A-20P, trade name, a product of TOA Gosei Chemical Industry Co., Ltd.) (1.2 parts by weight per 100 parts by weight of the solid content of the water glass aqueous solution)

Example 240: 1.0 g of polyvinyl alcohol (POVAL PA-05GP, trade name, a product of Shin-Etsu Chemical Industries, Ltd.)

Example 241: 1.0 g of methyl cellulose (METHOLOSE SM-25, trade name, a product of Shin-Etsu Chemical Industries, Ltd.)

Example 242: 1.0 g of carboxymethyl cellulose (reagent)

Example 243: 1.0 g of sulfuric band (reagent)

EXAMPLE 244

The elution test was conducted in the same manner as in Example 234 except that 7.9 g (solid content: 2.5 g) of a sodium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: 30% LIQUID ORTHO, solid content: 31.5%, $\text{SiO}_2/\text{Na}_2\text{O}$ compositional ratio = 0.50) was used in place of the sodium silicate solution (I).

EXAMPLE 245

The elution test was conducted in the same manner as in Example 234 except that 6.3 g (solid content: 2.5 g) of a potassium silicate solution manufactured by Nippon Chemical Industrial Co., Ltd. (trade name: A POTASSIUM SILICATE, solid content: 40.0%, $\text{SiO}_2/\text{K}_2\text{O}$ compositional ratio = 2.0) was used in place of the sodium silicate solution (I).

The elution test results obtained in Comparative Example 29 and Examples 235 to 245 are shown in Table 28 below.

TABLE 28

Elution Test Result	
	Pb elution amount (ppm)
Comparative Example 29	34
Example 235	8.3
Example 236	3.9
Example 237	13
Example 238	10
Example 239	15
Example 240	12
Example 241	18
Example 242	16
Example 243	21
Example 244	12
Example 245	13

It is understood from Examples 235 to 245 and Comparatives 1 and 29 that the Pb elution amount is decreased by even only addition of the water glass aqueous solution as compared with the case of non-treatment, and the water glass aqueous solution has the harmful metal (Pb) stabilizing performance. It is also understood that if the additive C is added, the stabilizing performance is further improved. Further, it is apparent from the comparison between Example 235 and Examples 244 and 245 that sodium silicate ($\text{Na}_2\text{O} \cdot n\text{SiO}_2$) has excellent Pb stabilizing performance as com-

pared with potassium silicate ($K_2O \cdot nSiO_2$), and is the preferred embodiment as the treating material. Furthermore, it is apparent that sodium silicate having SiO_2/Na_2O compositional ratio of 2.0 or more is more preferred embodiment as the treating material.

5 COMPARATIVE EXAMPLE 30

A water glass aqueous solution was prepared by adding 26.0 g of water to 13.0 g (solid content: 5.0 g) of the sodium silicate (I). This water glass aqueous solution was added to 50 g of the fly ash (I), and then kneaded. The resulting kneaded product (the solid content of the water glass aqueous solution was 5 parts by weight per 100 parts by weight of the ash) was aged at 20°C for 24 hours to solidify. After solidification, the solidified product was pulverized and classified with a sieve having an opening of 5 mm. 30 g of particles was dispensed from the particles which passed through the sieve, and the subjected to the elution test.

COMPARATIVE EXAMPLE 31

15 An aqueous solution prepared by dissolving 3.8 g of sodium chloride (monovalent metal ion of sodium chloride was 150 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) in 26.0 g of water was added to 13.0 g (solid content : 5.0 g) of the sodium silicate aqueous solution (I). The solid component was precipitated, and the solution was in the form which was impossible to use as a liquid treating material.

20 EXAMPLE 246

An aqueous solution prepared by dissolving 0.08 g of sodium chloride (monovalent metal ion of sodium chloride was 3 parts by mol per 100 parts by mol of cations of the water glass aqueous solution) in 26.0 g of water was added to 13.0 g (solid content : 5.0 g) of the sodium silicate aqueous solution (I). The aqueous solution was added to 50 g of the above fly ash (I). The elution test was conducted in the same manner as in Comparative Example 30.

EXAMPLES 247 TO 252

30 The elution test was conducted in the same manner as in Example 246 except that the following material was used in place of sodium chloride as the additive D.

- Example 247: 0.13 g of sodium chloride (monovalent metal ion of sodium chloride was 5 parts by mol per 100 parts by mol of cation of the water glass aqueous solution)
- 35 Example 248: 2.5 g of sodium chloride (monovalent metal ion of sodium chloride was 100 parts by mol per 100 parts by mol of cation of the water glass aqueous solution)
- Example 249: 2.0 g of sodium chloride
- Example 250: 2.0 g of potassium chloride
- Example 251: 2.0 g of sodium nitrate
- 40 Example 252: 2.0 g of ammonium chloride

The elution test results obtained in Comparative Example 30 and Examples 246 to 252 above are shown in Table 29 below.

TABLE 29

Elution Test Result	
	Pb elution amount (ppm)
Comparative Example 30	10
Example 246	9.5
Example 247	5.0
Example 248	2.1
Example 249	2.5
Example 250	2.8
Example 251	4.1
Example 252	2.9

It was proved from the comparison between Comparative Examples 1 and 30 that the Pb elution amount was decreased by the addition of the water glass aqueous solution as compared with the non-treatment, and the water glass aqueous solution had the harmful metal (Pb) stabilizing performance. However, the Pb stabilizing performance is not sufficient by the above-mentioned fly ash treatment method. Next, it is apparent from the comparison between Examples 246 to 252 and Comparative Example 30 that by adding the additive D, such a treating material has excellent Pb stabilizing performance as compared with the water glass aqueous solution. Further, it is apparent that where sodium chloride or potassium chloride is selected as the monovalent metal salt, it shows excellent Pb stabilizing performance as compared with other materials. Furthermore, it is apparent that if the amount of the monovalent metal salt added as the additive D is 3 parts by mol per 100 parts by mol of cations in the water glass aqueous solution, the improvement of the Pb stabilizing performance is not obtained as compared with the water glass aqueous solution, and if the amount thereof is 150 parts by mol, the resulting solution does not show the embodiment preferred as the treating material. Therefore, it is apparent that the monovalent metal ion of the monovalent metal salt is 5 to 100 parts by mol per 100 parts by mol of cations in the water glass aqueous solution, and this is the preferred embodiment as the treating material aqueous solution. In addition, it is apparent that ammonium chloride is preferred as the ammonium salt of the additive D.

By treating industrial wastes containing harmful metals using the waste-treating material and the treatment method according to the present invention, harmful metals, particularly Pb contained in the waste incineration fly ash, are stabilized, and the elution amount thereof is decreased. Further, a blend of waste and the treating material, which is obtained as a result of treating the waste according to the present invention can be reused as roadbed materials, aggregates for cement, and the like as materials that the elution amount of harmful metals is extremely small, and therefore can be valuable resources.

Claims

1. A waste-treating material comprising a water glass aqueous solution, and at least one of the following additive A and additive B, as the main structural components:

Additive A: At least one member selected from the group consisting of acids, alcohols, polyvalent metal salts, polyhydric alcohol esters, carbonates, intramolecular esters, dibasic acid esters and dialdehydes.

Additive B: At least one of materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water, and ion sealing agent which seals calcium ions.

2. The waste-treating material as claimed in claim 1, wherein the acids are added as the additive A in an amount such that an amount of hydrogen ions (H^+) of the acid is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.
3. The waste-treating material as claimed in claim 1 or 2, wherein the acids are at least one member selected from the group consisting of hydrochloric acid, nitric acid, phosphoric acid, sulfuric acid, carbonic acid, acetic acid and oxalic acid.

4. The waste-treating material as claimed in claim 1, wherein polyvalent metal salts are added as the additive A in an amount such that the product of mol part of polyvalent metal ions of the polyvalent metal salts and the number of valency thereof is 50 parts by mol or less per 100 parts by mol of cations in the water glass aqueous solution.
- 5 5. The waste-treating material as claimed in claim 4, wherein the polyvalent metal salts are at least one member selected from the group consisting of chlorides, nitrates, sulfates, carbonates and phosphates of magnesium, calcium, strontium, barium, iron, aluminum and zinc.
- 10 6. The waste-treating material as claimed in claim 5, wherein the polyvalent metal salts are at least one member selected from the group consisting of magnesium chloride (MgCl_2), calcium chloride (CaCl_2), iron (II) chloride (FeCl_2), iron (III) chloride (FeCl_3), aluminum chloride (AlCl_3), magnesium nitrate ($\text{Mg}(\text{NO}_3)_2$), calcium nitrate ($\text{Ca}(\text{NO}_3)_2$), iron (II) nitrate ($\text{Fe}(\text{NO}_3)_2$), iron (III) nitrate ($\text{Fe}(\text{NO}_3)_3$), aluminum nitrate ($\text{Al}(\text{NO}_3)_3$), magnesium sulfate (MgSO_4), iron (II) sulfate (FeSO_4) and aluminum sulfate ($\text{Al}_2(\text{SO}_4)_3$).
- 15 7. The waste-treating material as claimed in claim 1, wherein the additive B is added in an amount of 10 to 400 parts by weight per 100 parts by weight of a water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) in the water glass aqueous solution.
- 20 8. The waste-treating material as claimed in claim 1 or 7, wherein the materials which react with calcium ions as the additive B to produce calcium compounds which are insoluble or sparingly soluble in water are at least one member selected from the group consisting of phosphates, carbonates, sulfates, carboxylates and hydroxides of potassium, sodium and ammonium.
- 25 9. The waste-treating material as claimed in claim 8, wherein the materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water are at least one member selected from the group consisting of tripotassium phosphate, pentasodium triphosphate (sodium tripolyphosphate), hexasodium tetraphosphate (sodium tetrapolyphosphate), sodium hexametaphosphate, potassium carbonate, sodium carbonate, sodium hydroxide, potassium hydroxide, ammonium sulfate and potassium oxalate.
- 30 10. The waste-treating material as claimed in claim 8, wherein the materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water are potassium carbonate.
- 35 11. The waste-treating material as claimed in claim 10, wherein potassium carbonate is added in an amount such that a ratio of water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) : potassium carbonate is in the range of 90 : 10 to 40 : 60.
- 40 12. The waste-treating material as claimed in claim 10 or 11, wherein the potassium carbonate is blended with the waste-treating material in a manner such that an aqueous solution of potassium carbonate having a concentration of 40% or more is prepared, and the resulting solution is mixed with the water glass aqueous solution, followed by stirring.
13. The waste-treating material as claimed in claim 8, wherein the materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water are sodium carbonate.
- 45 14. The waste-treating material as claimed in claim 13, wherein sodium carbonate is added in an amount such that a ratio of water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) : sodium carbonate is in the range of 90 : 10 to 40 : 60.
- 50 15. The waste-treating material as claimed in claim 13, wherein sodium carbonate is added in an amount such that a ratio of water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) : sodium carbonate is in the range of 60 : 40 to 50 : 50.
- 55 16. The waste-treating material as claimed in claim 13, 14 or 15, wherein the sodium carbonate is blended with the waste-treating material in a manner such that an aqueous solution of sodium carbonate having a concentration of 20% or more is prepared, and the resulting solution is mixed with the water glass aqueous solution, followed by stirring.
17. The waste-treating material as claimed in claim 8, wherein the materials which react with calcium ions to produce calcium compounds which are insoluble or sparingly soluble in water are sodium hydroxide.

18. The waste-treating material as claimed in claim 17, wherein sodium hydroxide is added in an amount such that a ratio of water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) : sodium hydroxide is in the range of 90 : 10 to 40 : 60.
- 5 19. The waste-treating material as claimed in claim 17, wherein sodium hydroxide is added in an amount such that a ratio of water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) : sodium hydroxide is 60 : 40.
- 10 20. The waste-treating material as claimed in claim 1 or 7, wherein the ion sealing agents which seal calcium ions, as the additive B are at least one member selected from the group consisting of phosphates of potassium or sodium, and organic chelating agents having carboxyl group ($-COOX$, wherein X represents hydrogen, potassium or sodium) or dithiocarbamic acid group ($>NCSSY$, wherein Y represents hydrogen, potassium or sodium).
- 15 21. The waste-treating material as claimed in claim 20, wherein the ion sealing agents which seal calcium ions are at least one member selected from the group consisting of potassium 1-hydroxyethane-1,1-diphosphonate, sodium 1-hydroxyethane-1,1-diphosphonate, potassium ethylene diamine tetraacetate, sodium ethylene diamine tetraacetate, sodium dimethyldithiocarbamate, sodium diethyldithiocarbamate, and sodium dibutyldithiocarbamate.
- 20 22. The waste-treating material as claimed in claim 1, further comprising the following additive E:
Additive E: At least one member selected from the group consisting of gluconates, tartarates, benzoates, ligninsulfonates, polysaccharides, and bases comprising monovalent cations and hydroxide ions.
- 25 23. The waste-treating material as claimed in claim 22, wherein the additive E is added in an amount of 20 parts by weight or less per 100 parts by weight of a water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) in the water glass aqueous solution.
- 30 24. The waste-treating material as claimed in claim 22 or 23, wherein the bases comprising monovalent cations and hydroxide ions are at least one member selected from the group consisting of sodium hydroxide ($NaOH$), potassium hydroxide (KOH) and ammonium hydroxide (NH_4OH).
- 35 25. A waste-treating material comprising a treating material which comprises a water glass aqueous solution as the main structural component, and an additive C comprising a coagulating sedimentator added thereto.
- 40 26. The waste-treating material as claimed in claim 25, wherein the additive C is added in an amount of 1 to 40 parts by weight per 100 parts by weight of a water glass solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) in the water glass aqueous solution.
- 45 27. The waste-treating material as claimed in claim 25 or 26, wherein the additive C is at least one member selected from the group consisting of polyaluminum chlorides, carbamates, polyacrylamides, sodium polyacrylates, polyvinyl alcohols, methylcellulose, and carboxymethyl cellulose.
- 50 28. A waste-treating material comprising a treating material which comprises a water glass aqueous solution as the main structural component, and an additive D which comprises at least one member selected from the group consisting of monovalent metal salts and ammonium salts, which do not produce calcium compounds which are insoluble or sparingly soluble in water by reacting with calcium ions.
- 55 29. The waste-treating material as claimed in claim 28, wherein the additive D is added in an amount such that the amount of at least one of monovalent metal ions of the monovalent metal salts and ammonium ions of the ammonium salts is 5 to 100 parts by mol per 100 parts by mol of cations in the water glass aqueous solution.
30. The waste-treating material as claimed in claim 28 or 29, wherein the monovalent metal salts as the additive D are at least one member selected from the group consisting of chlorides and nitrates of sodium or potassium.
31. The waste-treating material as claimed in claim 29, wherein the monovalent metal salts as the additive D are at least one of sodium chloride ($NaCl$) and potassium chloride (KCl).
32. The waste-treating material as claimed in claim 29, wherein the ammonium salt as the additive D is ammonium

chloride (NH_4Cl).

- 5 **33.** The waste-treating material as claimed in claim 1, 26 or 28, wherein the main structural component of the water glass aqueous solution is water glass ($\text{M}_2\text{O} \cdot n\text{SiO}_2$) having $\text{SiO}_2/\text{M}_2\text{O}$ compositional ratio (wherein M represents cation in the water glass aqueous solution) of 2.00 or more.
- 10 **34.** The waste-treating material as claimed in claim 1, 26 or 28, wherein the main structural component of the water glass aqueous solution is sodium silicate.
- 15 **35.** A method for treating waste, which comprises kneading waste and the waste-treating material as claimed in any of claims 1 to 34, and aging the resulting mixture.
- 20 **36.** The method for treating waste as claimed in claim 35, wherein the additive A is added to the water glass aqueous solution, and after 5 minutes, the resulting treating material is added to the waste.
- 25 **37.** The method for treating waste as claimed in claim 35, wherein the water glass aqueous solution and the additive A are separately stored, those are mixed immediately before adding to the waste, and the resulting treating material is added to the waste.
- 30 **38.** The method for treating waste as claimed in claim 35, wherein the water glass aqueous solution and other additives are separately stored, the water glass is added to the waste, if necessary followed by mixing, the other additives are added to the mixture and kneaded, and the resulting mixture is aged.
- 35 **39.** The method for treating waste as claimed in claim 35, wherein the waste-treating material is heated at 5 to 30°C and then added to the waste.
- 40 **40.** The method for treating waste as claimed in claim 35, wherein the waste contains calcium compounds such as calcium hydroxide, calcium oxide or calcium chloride.
- 45 **41.** The method for treating waste as claimed in claim 35 or 40, wherein the waste is waste incineration fly ash.
- 50 **42.** The method for treating waste as claimed in claim 35, wherein the aging is conducted at a temperature of 40 to 100°C.
- 55 **43.** The method for treating waste as claimed in claim 42, wherein the aging is conducted for 6 hours or more.
- 44.** The method for treating waste as claimed in claim 42, wherein the heating is conducted utilizing heat generated in a waste incinerator.
- 45.** The method for treating waste as claimed in claim 35, wherein the waste-treating material is added in an amount such that the amount of solid component (the sum of the weight of M_2O and the weight of SiO_2 , wherein M represents a cation in the water glass aqueous solution) in the water glass aqueous solution is 1 to 20 parts by weight per 100 parts by weight of the waste.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP95/01966

A. CLASSIFICATION OF SUBJECT MATTER		
Int. C1 ⁶ B09B3/00, A62D3/00, C09K3/00		
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED		
Minimum documentation searched (classification system followed by classification symbols)		
Int. C1 ⁶ B09B3/00, A62D3/00, C09K3/00		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP, 52-109482, A (Naojiro Oshita), September 13, 1977 (13. 09. 77) (Family: none) Claim, line 18, lower left column, page 2 to line 8, upper left column, page 3	1, 4-6, 33-35, 38, 40, 41, 43, 45
X	JP, 52-84174, A (Kyokado Engineering K.K.), July 13, 1977 (13. 07. 77) (Family: none) Claim, line 2, lower right column, page 2 to line 20, upper right column, page 4	1 - 9, 28 - 34
P	JP, 7-39847, A (Entec K.K.), February 10, 1995 (10. 02. 95) (Family: none) Claim, line 35, column 2 to line 40, column 3	1, 2, 22, 23, 35-37, 40, 41
A	JP, 57-204276, A (Nihon Kaihatsu Consultant K.K.), December 14, 1982 (14. 12. 82) (Family: none) Claim, line 10, upper left column to line 15, upper right column, page 2	1, 35
☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier document but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search February 7, 1996 (07. 02. 96)		Date of mailing of the international search report February 27, 1996 (27. 02. 96)
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Form PCT/ISA/210 (second sheet) (July 1992)