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(54) Regenerative soot filter device and method for regenerating a soot filter

(57) The invention pertains to a regenerative soot filter (1) device applicable to exhaust gases from combustion engines, particularly diesel type engines. The device comprises a microwave generator (16) and a soot filter material (10), wherein a microwave absorbing material is provided on the soot filter material. The invention also pertains to a method of regenerating a soot filter comprising soot filter material applicable to exhaust gases from combustion engines, particularly diesel type engines. The method comprises applying microwave radiation to the soot filter by a microwave generator and passing a gas flow comprising oxygen over the soot filter, wherein the microwave radiation applied to the soot filter is absorbed by a microwave absorbing material that is provided on the soot filter material.

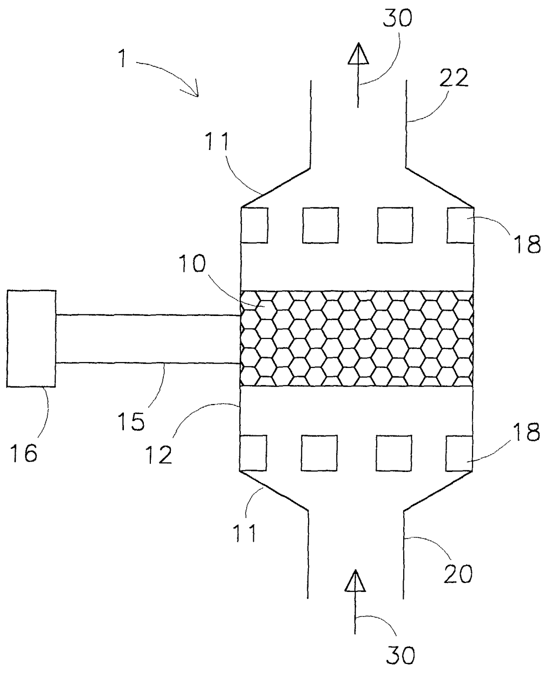

Fig 5

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Description

[0001] The invention pertains to a regenerative soot filter device applicable to exhaust gases from combustion engines, particularly diesel type engines, comprising a microwave generator and a soot filter material.

[0002] Such a device is known in the art, e.g. from EP-A1-0 635 625. This European patent application discloses a method of regenerating a ceramic soot filter, as well as a device for performing this method. Soot deposited on the ceramic soot filter used for diesel exhaust gases is oxidized by application of microwave radiation from a microwave generator. The soot is heated by the microwave radiation and oxidized in an oxygen excess present in the diesel exhaust gases. It is indicated that an ignition temperature of around 600°C is needed to completely oxidize soot which mainly comprises graphitic carbonaceous components, as well as aliphatic, alicyclic and aromatic hydrocarbons. A major concern is that all ceramic materials show a higher absorption of microwave radiation at higher temperatures, leading to a progressive heating when a constant radiation power is applied over longer periods of time. This effect is known as the thermal runaway and is to be avoided under all circumstances as it leads to overheating of the ceramic filter material and consequently to distortion and mechanical failure. In the known method this problem is solved by applying microwave radiation in pulses and by providing sufficient heat exchange by radiation and/or convection. The known method of regenerating a soot filter thus requires a delicate balancing of the heating by microwave radiation of the filter, because a temperature around 600°C is required to obtain a complete oxidation of soot, while, by contrast, any overheating of the filter is to be avoided. Optionally, a catalytically active material may be present to enhance CO oxidation. Noble metals, especially Pt, Rd and Pd, are generally known to be particularly suitable for this catalytic requirement. They are, for instance, widely used as so-called three way catalysts in automobiles.

[0003] This known method has some drawbacks. Firstly, pulsing of microwave radiation needs to be carefully monitored in order to maintain the delicate balance between the ignition temperature of the soot on the one hand and the temperature of the ceramic filter on the other hand. Secondly, complete oxidation of soot will be compromised, as the heating of soot is limited by the pulsing of the microwave radiation. Thirdly, the implementation of a heat exchange system renders the design of the device more complex and less easy to produce. Furthermore, noble metals are lately becoming more scarce and expensive, thereby increasing the total costs of catalysts in a soot filter that incorporate noble metals. Finally, distortion and mechanical failure of the filter will still occur to some extent rendering the device a shorter lifetime.

[0004] Apart from these drawbacks, it is a general desire to use microwave energy for the oxidation of soot in a more efficient way than by purely raising the temperature of the soot, as for example disclosed in EP-A1-0 635 625.

[0005] The object of the invention is to provide a regenerative soot filter device wherein the above mentioned drawbacks have been reduced or even eliminated, and a more efficient use of microwave energy is accomplished.

[0006] The regenerative soot filter device according to the invention is characterized in that a microwave absorbing material is provided on the soot filter material. The microwave absorbing material is provided on the soot filter material by deposition techniques known in the art so that the surface of the soot filter material is covered. Preferably, a deposition technique is chosen such that a physical or chemical bonding is obtained between the microwave absorbing material and the soot filter material, the bonding being capable of withstanding the working conditions. The term 'microwave absorbing material' is shortly explained hereinafter for clarity's sake.

[0007] In general, the microwave absorbing capacity of a material is indicated by its dielectric loss factor ε''. This can be deduced from the following formula 1 regarding the volume specific absorption of microwave radiation A of a microwave absorbing material:

$$A = n * f * \varepsilon'' * \tan \delta / E^2 \qquad (1)$$

wherein f is the frequency of the microwave radiation, E the mean strength of the electromagnetic field in the absorbing volume and tan 6 is the ratio of the dielectric loss factor ε'' to the dielectric constant ε' of the material.

[0008] Advantageously, the microwave absorbing material has a dielectric loss factor being a multiple of the dielectric loss factor of the soot filter material as well as the soot. For example, a known soot filter material like cordierite having a dielectric loss factor typically of 0.14, would be provided according to the invention with a microwave absorbing material having a dielectric loss factor being at least two times higher. The use of such a microwave absorbing material with a dielectric loss factor higher than the filter material and the soot reduces the need to apply a high electric field strength, still allowing the soot to reach ignition temperatures. Consequently, an important advantage of the device according to the invention is that the ignition temperature of soot can be reached by applying lower strengths of the electrical field, resulting in a decrease of the energy consumption.

[0009] Contrary to the known soot filter device according to EP-A1-0 635 625 wherein the soot itself is heated by microwave radiation and excessive heating of the soot filter material by microwave radiation is avoided by cooling through heat exchange and the pulsing of microwave radiation, the invention aims at supplying thermal energy for the oxidation of soot while protecting the soot filter material from being overheated by providing the soot filter material with a microwave absorbing material as defined above. Firstly, with regard to the oxidation of soot deposited on the soot filter material provided with microwave absorbing material according to the invention, it is evident that the thermal energy contained by the microwave absorbing material contributes to the heating of soot by heat exchange. As the microwave absorbing material is more efficiently heated by microwaves than the soot itself , the invention provides a regenerative soot filter device that uses microwave energy in a more efficient way, for instance a lower power of the applied microwave source may be employed. Moreover, a pulsing of microwave radiation to the extent according to the prior art is not required, so that the heating of the soot is less limited and more easily maintained in the device according to the invention, leading to a more complete oxidation of soot. Secondly, the microwave absorbing material provided on the soot filter material will also lower the penetration of microwave radiation into the soot filter material itself considerably. Thus the thermal runaway effect leading to overheating of the soot filter material is avoided, even more so because the heat energy contained by the microwave absorbing material being located on the surface of the soot filter material is easily exchanged with the gas passing through by radiation and/or convection, apart from the exchange with the soot deposited. Both beneficial effects of the microwave absorbing material, namely a more efficient use of microwave energy as well as a protection for the soot filter material from overheating, will elongate the lifetime of the device as the soot filter material itself is less subjected to microwave heating.

[0010] Advantageously, the greater part of the surface of the soot filter material is covered by a microwave absorbing material to obtain a good heat exchange with soot as well as with the gas passing through and a sufficient protecting effect from overheating. As the absorption of microwave radiation is proportional to the volume of the microwave absorbing material, the beneficial effects can further be controlled by adjusting the thickness of the microwave absorbing material provided on the soot filter material.

[0011] Advantageously, the device according to the invention comprises also an oxidation catalyst to completely oxidize any partially oxidized products like CO, as well as other hydrocarbons present in the gas phase. Preferably, the device comprises a microwave absorbing material that is an oxidation catalyst. By combining both functions in the device according to the invention, a complete oxidation of soot - via partially oxidized products like CO as well as other hydrocarbons present in the gas phase - into harmless CO_2 is further enhanced.

[0012] The device according to the invention preferably comprises a microwave absorbing material comprising a compound AMO_y having a perovskite structure (also referred to as 'a perovskite' hereinafter) wherein A is a rare earth metal, M is a transition metal, wherein the A atom is partly substituted by a different atom A', A' also being a rare earth metal and y is a numerator for the number of oxygen atoms O,the numerator y being dependent on the mean valency of the metalions included in the perovskite. These perovskites surprisingly show a good catalytic activity towards oxidation combined with a dielectric loss factor that is suitable for the purposes as explained above.

[0013] More preferably, the microwave absorbing material comprises a compound AMO_y having a perovskite structure wherein A is La or Sr, A' is La, Ce, Hf or Sr, and M is Mn, Co, Fe or Ti. Perovskites containing combinations of these specific elements surprisingly show a higher catalytic activity over perovskites in general. Among these, more preferred perovskites are compounds of formula AMO_y wherein A is La, A' is Ce, M is Mn, and y is 3. It is noted that the value of y, being dependent on the mean valency of the metalions included in the perovskite, should formally be interpreted as an approximate value.

[0014] The device according to the invention preferably comprises microwave absorbing material comprising a perovskite as described above, wherein the degree of substitution of the A atom is between 5 and 40%. Perovskites having these specific degrees of substitution surprisingly show a higher catalytic activity over perovskites in general. Most preferred perovskites are compounds wherein the degree of substitution of the A atom is 20%, in particular $La_{0.8}Ce_{0.2}MnO_3$.

[0015] The soot filter material in the soot filter device according to the invention can be a conventional one, such as ceramic cordierite, alumina, silica, zirconia or titania, preferably having a honeycomb structure design. A favorable application of the regenerative soot filter device according to the invention is in (stationary and instationary) diesel engines, because diesel engines are generally known to produce more soot than other combustion engines.

[0016] The invention also relates to a method of regenerating a soot filter, comprising a soot filter material, which method comprises applying microwave radiation to the soot filter by a microwave generator and passing a gas flow comprising oxygen over the soot filter, characterized in that the microwave radiation applied to the soot filter is absorbed by a microwave absorbing material that is provided on the soot filter material. The advantages mentioned earlier for the regenerative soot filter device, also apply to this method.

[0017] In a preferred embodiment of the method according to the invention the microwave absorbing material used is an oxidation catalyst.

[0018] In a more preferred method according to the invention a microwave absorbing material is used that comprises a compound AMO_y having a perovskite structure wherein A is a rare earth metal, M is a transition metal, wherein the A atom is partly substituted by a different atom A', A' also being a rare earth metal and y is a numerator for the number of oxygen atoms O.

[0019] More preferably, the microwave absorbing material comprises a compound AMO_y having a perovskite structure wherein A is La or Sr, A' is La, Ce, Hf or Sr, and M is Mn, Co, Fe or Ti. Among these, more preferred perovskites are compounds of formula $AMO_{\mathbf{y}}$ wherein A is La, A' is Ce, M is Mn, and y is 3 +/- d.

[0020] In a still more preferred method according to the invention a perovskite as defined above is used, wherein the degree of substitution of the A atom is between 5 and 40%, more preferably 20%.

[0021] The following examples are provided as a means of illustrating the present invention especially the microwave absorbing material that is a oxidation catalyst. These examples are not be construed as imposing a limitation to the present invention. Also, the present invention will be further elucidated by the accompanying drawings, wherein:

Fig. 1 shows the results referred to in Example 2 of measurements of catalytic activity of some microwave absorbing materials used in the regenerative soot filter device according to the invention.

Fig. 2 shows the results referred to in Example 3 of measurements of catalytic activity of a microwave absorbing material used in the regenerative soot filter device according to the invention with and without the addition of water to the gas stream.

Fig. 3 shows the results referred to in Example 4 of measurements of the dielectric constant and dielectric loss factor of a microwave absorbing material used in the regenerative soot filter device according to the invention comprising $La_{\mathbf{0.9}}Ce_{\mathbf{0.1}}MnO_{\mathbf{3}}$.

Fig. 4 shows the results referred to in Example 4 of measurements of the repeated microwave heating of a microwave absorbing material used in the regenerative soot filter device according to the invention comprising $La_{\mathbf{0.8}}Ce_{\mathbf{0.2}}MnO_{\mathbf{3}}$.

Fig. 5 shows a cross sectional view of a preferred embodiment of the regenerative soot filter device according to the invention.

Example 1

Preparation of perovskite-type oxide $La_{\mathbf{1-x}}A'_{\mathbf{x}}MnO_{\mathbf{3}}$

[0022] A series of perovskite-type oxide $La_{\mathbf{1-x}}A'_{\mathbf{x}}MnO_{\mathbf{3}}$ (A'=Ce and x being 0, 0.05, 0.1, 0.2 and 0.3) was prepared by a co-precipitation method. An aqueous solution of corresponding nitrates was used with sodium hydroxide solution and hydrogen peroxide as the precipitating agents.

[0023] During the precipitation the pH was kept at pH 9-9.4 at 50°C. The metal nitrates, the sodium hydroxide solution and hydrogen peroxide were added to 600 ml double distilled water while vigorously stirring the suspension. The precipitation time was about 90 minutes. The precipitate was aged for about 30 minutes, filtered off, washed three times with double distilled water to remove the remaining reactants, and dried over night at 120°C in air. After drying the precipitates were crushed and sieved to obtain a particle size between 125 and 212μm. Finally, the ground precipitates were calcined in technical air at different temperatures with heating rate of $5°C.min^{-1}$ (with the final temperature ranging from 500°C to 800°C, preferably 800°C) for 6 hours with an airflow of 100 ml/min.

Example 2

Catalytic activity - CO oxidation

[0024] The catalytic activity of the perovskites from Example 1 was investigated by using CO oxidation as model system. The experiments were carried out in a reactor coupled to a mass spectrometer (MS). A gas mixture with 1 vol% CO and 1 vol% O_2 in He was passed over the catalyst. As a catalyst 50 mg of perovskite was used having a particle size smaller than 0.2 mm. The conversion of CO was measured by MS in a temperature range of 75-300°C with a heating rate of 5°C/min. The results are given in fig. 1.

[0025] The results show that the highest activity of CO oxidation was obtained with the perovskite $La_{0.8}Ce_{0.2}MnO_3$. The increasing order of catalytic activity for this perovskite series, is $La_{0.8}Ce_{0.2}MnO_3 > La_{0.7}Ce_{0.3}MnO_3 > La_{0.9}Ce_{0.1}MnO_3 > La_{0.95}Ce_{0.05}MnO_3 > LaMnO_3$. In regard to $La_{0.8}Ce_{0.2}MnO_3$, the onset of oxidation occurred at 75°C with completion at 250°C. The catalytic properties of $La_{0.8}Ce_{0.2}MnO_3$ are totally different compared to the physical mixture of $LaMnO_3$ and CeO_2 also included in fig. 1. Fig. 1 also demonstrates that without catalyst, CO oxidation will not take place when the temperature is below 300°C.

Not only the catalytic activity was tested, but also the stability. The CO oxidation for each perovskite was repeated under the same reaction condition twice, a similar activity was observed, indicating a good temperature stability of these perovskites.

Example 3

Effects of water on catalytic activity of $La_{0.8}Ce_{0.2}MnO_3$

[0026] The impact of water vapor addition on catalyzed CO oxidation over $La_{0.8}Ce_{0.2}MnO_3$ prepared according to Example 1 is demonstrated in fig. 2. The addition of 3 vol.% water to the gas mixture used in Example 2 causes the decrease of the CO conversion. The CO conversion reduces instanteneously to about 72% of its original level, at which it stabilizes very quickly. After removal of the water-supply the activity is rapidly restored to about 100% of its original level. After repeated cycles the same activity levels are reached. This hydrothermal stability renders the perovskite more favorable to be applied to engine exhaust gases which normally contain some water vapor as well.

Example 4

Microwave heating

[0027] The microwave heating was carried out in a microwave system, which consisted of microwave source (2.45 GHz, 1kW, Muegge, MW-46029-850-01), a circulator (Philips, 27722.163.02071), a 3-stub tuner section (Muegge, MW-7614-0060), a monomode microwave cavity TE_{10} and water load.

[0028] The radiation is transported through the wave-guide, which is formed by a copper rectangular channel with the dimensions 0f 7.21 cm (width) by 3.61 cm (height), to the desired location. The circulator is placed to protect the microwave source from the reflected radiation. Any radiation reflected in the reactor is led to a water load located behind the circulator. The stub tuners are used to minimize the reflected radiation. A quartz tubular reactor with internal diameter of 18 mm is placed in the applicator section, which contains two power sensors (Rhode & Schwarz, 828.3818.02). The water load at the end of the wave-guide absorbs any microwave radiation after passing through the reactor. The microwave set-up system is operated in travelling wave mode; any radiation not absorbed after passing through the microwave cavity, is absorbed by the water load.

[0029] A sample with constant volume (10 ml) was placed in the reactor. The sample was first dried in the conventional oven at 150°C for one hour with heating rate of $5°C.min^{-1}$ in He flow ($GHSV=6.10^{-2}$ hr^{-1}). After cooling down to the room temperature in He flow, the sample was transferred to the microwave cavity under a high He flow. The temperature in the sample bed was controlled through a temperature control loop coupled to the microwave power and the optical fiber. The sample was heated up to 600°C and kept there for 24 hours in He flow ($GHSV=6.10^{-2}\ hr^{-1}$) with heating rate of $2°C.min^{-1}$ in the microwave field. Then the same sample was subsequently heated up at 200W repeatedly with the same gas hourly space velocity.

[0030] All the perovskites $La_{1-x}Ce_xMnO_3$ (x=0.0, 0.05, 0.2 and 0.3) show the microwave absorption, independent of the quantity of substitution of lanthanum by cerium. The perovskites also show high thermal stability upon the heating from 24 hours, because no gas release was measured by MS, and the weight before and after is comparable. The dielectric constant and the dielectric loss factor of $La_{0.9}Ce_{0.1}MnO_3$ were measured at room temperature and the frequency ranged from 30 MHz to 3000 MHz. The obtained results are shown in fig. 3. The real part of dielectric constant of this perovskite (ε') is about 4 and the dielectric loss factor, the imaginary part (ε") is about 0.91. Compared to the dielectric loss factor (ε") of cordierite and TiO_2, 0.14 and 0.43, respectively, the dielectric loss factor of this perovskite is about six times and two times higher. The results of the repeated heating over $La_{0.8}Ce_{0.2}MnO_3$ at 200W are presented in fig. 4.

[0031] A preferred embodiment of the device according to the invention is depicted in Figure 5, showing said device in cross sectional view. A regenerative soot filter device 1 is equipped with a soot filter material 10 of cordierite ceramic with a honeycomb structure on which microwave absorbing material is provided. The soot filter material 10 is positioned in a cylindrical filter chamber 12 having two narrowing conical ends 11. At the height of the soot filter material 10 at the outside of the filter chamber 12 is connected to one end of a microwave conducting channel 15 which is at the other end connected to a microwave generator 16, so that microwave radiation generated can enter the filter chamber 12. One narrowing conical end of the filter chamber 12 is connected to an inlet for exhaust gases 20 and the other narrowing conical end is connected to an outlet 22 for the gases after having passed through the soot filter material 10. The filter chamber 12 is further equipped with so-called Lambda quarter traps 18 to trap microwave radiation in order to prevent leakage of radiation out into other parts connected to the regenerative soot filter device 1. The direction of the gas stream is indicated by arrows 30.

Claims

1. Regenerative soot filter device applicable to exhaust gases from combustion engines, particularly diesel type engines, comprising a microwave generator and a soot filter material, **characterized in that** a microwave absorbing material is provided on the soot filter material.

2. Regenerative soot filter device according to claim 1, **characterized in that** the microwave absorbing material is an oxidation catalyst.

3. Regenerative soot filter device according to one of the preceding claims 1-2, **characterized in that** the microwave absorbing material comprises a compound AMO_y having a perovskite structure wherein A is a rare earth metal, M is a transition metal, wherein the A atom is partly substituted by a different atom A', A' also being a rare earth metal and y is a numerator for the number of oxygen atoms O.

4. Regenerative soot filter device according to claim 3, **characterized in that** A is La or Sr, A' is La, Ce, Hf or Sr, and M is Mn, Co, Fe or Ti.

5. Regenerative soot filter device according to claim 4, **characterized in that** A is La, A' is Ce, M is Mn, and y is 3.

6. Regenerative soot filter device according to one of the preceding claims 3-5, **characterized in that** the degree of substitution of the A atom is between 5 and 40%.

7. Regenerative soot filter device according to claim 6, **characterized in that** the degree of substitution of the A atom is 20%.

8. Method of regenerating a soot filter, comprising a soot filter material, which method comprises applying microwave radiation to the soot filter by a microwave generator and passing a gas flow comprising oxygen over the soot filter, **characterized in that** the microwave radiation applied to the soot filter is absorbed by a microwave absorbing material that is provided on the soot filter material.

9. Method of regenerating a soot filter according to claim 8, **characterized in that** the microwave absorbing material is also an oxidation catalyst.

10. Method of regenerating a soot filter according to one of the preceding claims 8-9, **characterized in that** the microwave absorbing material comprises a compound AMO_y having a perovskite structure wherein A is a rare earth metal, M is a transition metal, wherein the A atom is partly substituted by a different atom A', A' also being a rare earth metal and y is a numerator for the number of oxygen atoms O.

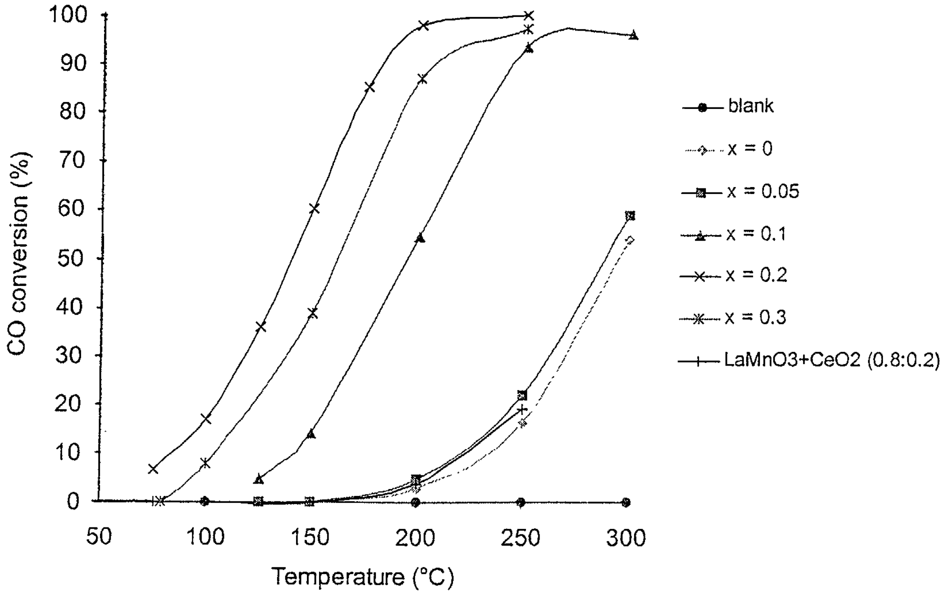
11. Method for regenerating a soot filter according to claim 10, **characterized in that** A is La or Sr, A' is La, Ce, Hf or Sr, and M is Mn, Co, Fe or Ti.

12. Method for regenerating a soot filter according to claim 11, **characterized in that** A is La, A' is Ce, M is Mn, and y is 3.

13. Method for regenerating a soot filter according to one of the preceding claims 10-11, **characterized in that** the degree of substitution of the A atom is between 5 and 40%.

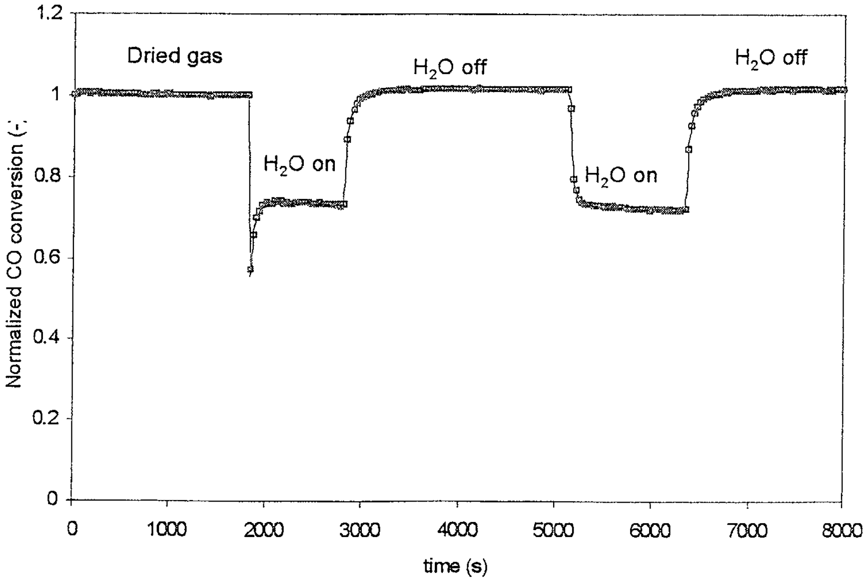
14. Method for regenerating a soot filter according to claim 13, **characterized in that** the degree of substitution of the A atom is 20%.

Fig. 1



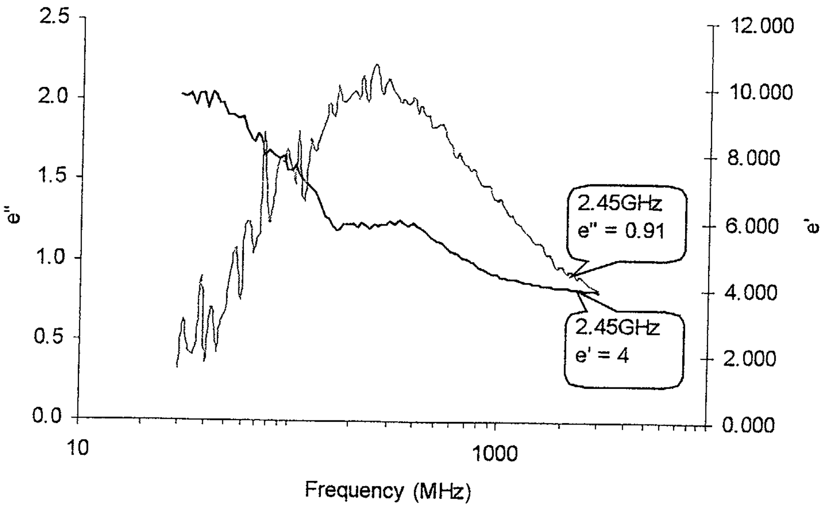
Relative conversion of CO over a blank reactor, $LaMnO_3$, $La_{0.95}Ce_{0.05}MnO_3$, $La_{0.9}Ce_{0.1}MnO_3$, $La_{0.8}Ce_{0.2}MnO_3$, $La_{0.7}Ce_{0.3}MnO_3$ and a physical mixture of $LaMnO_3$ + CeO_2.

Fig. 2



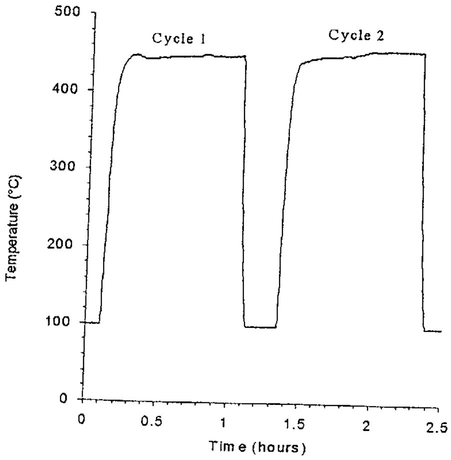
Normalized CO conversion over $La_{0.8}Ce_{0.2}MnO_3$ as a function of time at T=200°C; 2 times 3 vol% H_2O is added (H_2O on) to a gas mixture of 1% O_2 and 1% CO in He.

Fig. 3



Dielectric constant (e″) and dielectric loss factor (e′) versus frequency.

Fig. 4



Two subsequent dielectric heating cycles in He at 200 W for $La_{0.8}Ce_{0.2}MnO_3$.

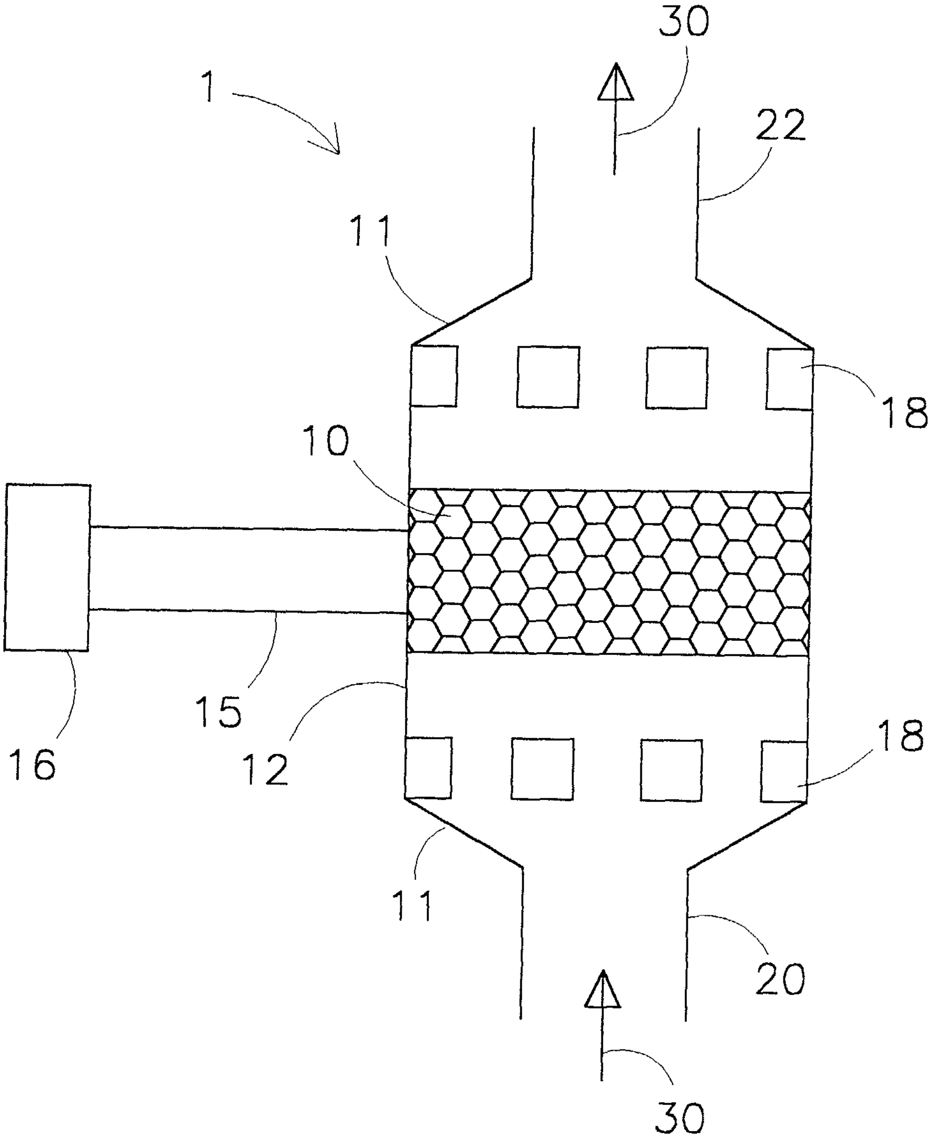


Fig 5

European Patent
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EUROPEAN SEARCH REPORT

Application Number
EP 01 20 3954

DOCUMENTS CONSIDERED TO BE RELEVANT

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TECHNICAL FIELDS SEARCHED (Int.Cl.7)
F01N H05B

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	20 February 2002	Torle, E

CATEGORY OF CITED DOCUMENTS

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