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(54) A procedure for the metallisation conditioning of radioactive graphite

(57) The procedure for the metallisation conditioning of radioactive graphite from nuclear plants or nuclear plant dismantling consists of the metallisation of the irradiated graphite through a double and thin layer of copper-nickel for water-tightness purposes, minimizing the lixivation of the radionucleids found inside and facilitating its handleability by eliminating the unctuosity problem. The metallisation is carried out by an electrodeless self-catalyst reduction of the metal salts found on the surface of the graphite where previously the catalyst has been deposited and activated.

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Description

NAME OF THE INVENTION

A procedure for the metallisation conditioning of radioactive graphite from nuclear plants or nuclear plant dismantling.

TECHNICAL SCOPE

The invention is included within the field of nuclear power, since graphite is used in nuclear reactors as a moderator and neutron reflector inside the nucleus.

During its stay inside the nucleus, the graphite is subject to an intense neutron flow which activates its structural carbon and the impurities it contains. Furthermore, during the operation of the reactor, it suffers a radioactive pollution process which adds to the activation process. As a consequence, the graphite becomes radioactive graphite and when it is extracted from the nucleus it must be conditioned to allow its storage.

Most of the existing radioactive graphite comes basically from four kinds of power reactors the nuclei of which is formed by a stack of prismatic graphite rods with several cavities where combustible elements are housed.

Gas refrigerated graphite reactors with unenriched uranium belong to the first generation of reactors and, therefore, it seems logical to think that the useful lives of a vast majority of them are nearly exhausted.

The graphite is a solid, quite inert element, with good mechanical properties. The graphite used in nuclear reactors is amorphous and formed by microcrystals with a low content of impurities.

In the case of graphite crystal, the carbon atoms form structured layers of regular hexagon sheets. Each layer, which is joined to the others by means of the force of Van der Waals, forms a gigantic bi-dimensional molecule.

This kind of structure confers the graphite the following characteristic properties which influence the behaviour of radioactive graphite:

- A structure of carbon atoms bonded by means of covalent bondings, very stable and inert.
- A number of chemical elements and compounds can be introduced between the layers, forming the so-called sandwiching compounds. Therefore, many of the graphite impurities are not adhered to the surface by mere physical bondings but, in certain cases, are true chemical compounds.
- The structure of graphite, which is basically covalent, makes it a hydrofugous substance which only enables the lixiviators, ordinarily hydrous to actuate on surface layers.
- Since the layers are bonded by means of weak forces, they can be easily separated, and some surface graphite particles are shed by rubbing. When the graphite is of the radioactive type, this property represents a handling problem, since it contaminates easily everything that comes into contact whit it.

It has a very irregular surface with many pores and cavities that make it to easily absorb environmental hydrocarbonated compounds.

The kind and concentration of radionuclides present in radioactive graphite depend on the content of impurities, the neutronic flow to which it has been subject and the pollution suffered, either at the nucleus or subsequently.

In general terms, the most important radionucleids present in graphite are H-3, C-14 and Co-60.

The fixation and lixiviation capabilities depend on the aforementioned properties.

H-3 is mainly generated from fuel, and it comes off as a gas. It is introduced in the graphite by fixing itself to active points of its structure, by means of chemical bonds.

The H-3 found at the outer layers interchanges isotopically with the hydrogen of hydrocarbonated compounds and water absorbed from the atmosphere. The graphite escapes when the former come off. Therefore, the outer layers are less tritium rich than the adjacent inner ones.

The C-14 comes from the activation of the carbon found in the graphite structure and therefore, it is very stable, and it only comes off by contact due to the unctuousness of the surface.

The Co-60 comes from the external activation and pollution. That coming from the activation can be found throughout the graphite, and that from the pollution is found on the surface and it is much more lixiviable.

STATE OF THE ART

The conditioning techniques used until present consist of the graphite coating with epoxy resins, bitumen and hydraulic binder.

The ultimate aim of the graphite conditioning by means of metallic coating, which is the object of this invention, is to seal the radioactive graphite obtained in nuclear plants, to minimise the lixiviation of the radionucleids that it contains and eliminate the unctuousness of its surface, to prevent any pollution during its handling.

The coating application system is based on the placement of two overlapped metallic layers with a thickness of a few microns, the first one made of copper and the second one of nickel, by means of the method known as "electroless", or electrodeless catalytic reduction.

Basically, it consists of reducing to a metal the ions present on a solution of metal salt, depositing it on the graphite surface, which has been previously catalysed so that this reaction can take place preferably on it, instead of in the bulk of the solution. To control this process, the ions present at the metal salt are sequestered, releasing them as needed.

Free copper ions are converted into metallic copper, preferably on the catalysed surface, where the activating energy of the reaction is smaller, thanks to the action of the reducer.

The procedure is based on the self-catalytic coating principles normally used in the industry under the licence of many patents.

The technique used to coat the radioactive graphite, although being similar, has to solve the specific problems posed by the own nature of graphite.

- The graphite has a porous and hydrofugous surface which cannot be mechanically treated, as the pieces to be coated, in the case of the electroless industry, since the pollution generated would be very high, thus impairing the surface preparation procedure.
- The coating has to be water-tight and therefore, the cavities existing between metal crystallization granules are more important than the granules themselves.

To achieve a water tight coating on the irregular porous heterogeneous surface, it has been necessary to thoroughly study the different stages of the process.

The problem has been solved by applying an initial semi-permeable copper layer which serves as a backbone to smooth the surface irregularities and where an amorphous nickel layer is deposited to make the previous layer water tight.

Copper has been used for the first layer since it is a metal easy to deposit and that serves to adjust the size of the metallic granules. The second layer is made of nickel since it is amorphous, very compact and resistant to corrosion.

In the case of copper, an intermediate granule size has been chosen, that enables to cover the surface cavities and pores, leaving small intergranular spaces that can be sealed by means of a nickel coating. The size of the granule has been achieved through an adequate catalysation process and the incorporation of an additive to the solution. The higher the number of active points is, the more nucleation points will be obtained and more copper crystals will be formed during the metallization, thus decreasing its size with the number.

During the depositing of the nickel layer, the concentration of nickel cations present in the solution has been limited to reduce the deposition speed and to prevent an excessively quick growth which might cause cracks.

In order to obtain the minimum quantity of radioactive residues, a series of metallisation solutions to deposit a metallic layer at the maximum speed have been looked for, in an effort to decrease the lixiviation time.

The metallisation through self-catalytic reduction is a technique which has been used since 1844. In 1916, F.A. Roux registered the first patent for Nickel depositing. In the fifties, the technique was industrialised, and in the period 1959-1973, a series of more complex industrial techniques were developed, including the alloy depositing. Since 1973 onwards, the industrial competition has increased, and the methods to evaluate the coatings have been standardized.

This kind of coatings is used to modify the properties of the surfaces, to make it more corrosion-resistant, to incorporate new properties or to improve their aesthetic aspect. When applied to metals, it competes with electroplating, advantageously in some cases due to the possibility of modifying the characteristics of the deposit with additives. The possibility of depositing metals on non-conducting surfaces enables the technique to obtain metal-coated plastics.

The polymeric surfaces are coated with a thin metal coat to make them more attractive, to protect them against corrosion or to modify its electric or magnetic properties. With this in mind, there is a significant activity focused on the home electric appliances, motor cars, chemical and electronics sector, etc.

The possibility of depositing many types of pure metals or metal alloys in very thin layers has paved the way to new fields of application.

In all cases, the aim is to change the aspect of the surface, to modify its electric or magnetic properties, or to protect it against external agents, but it has never been used as a barrier to prevent leakages of the products contained inside it, and never in the case of radioactive products. The interest has been focused on the composition of the deposited layer and on the characteristics of the metallic granules, but not on the room left between granules and the water tight coating which is the basis of this patent.

DESCRIPTION OF THE INVENTION

METALLIC COATING THROUGH SELF-CATALYTIC REACTION

The coating application system is based on the depositing of a metallic layer through the method known as "electroless" or electrodeless self-catalytic reaction.

The process for the complete conditioning consists of four basic stages: catalysation, metallisation with copper, activation and metallisation with nickel.

If the graphite is to be metallised with the aim to eliminate its unctuousness, it suffices to submit it only to the two first stages.

In the different stages, the parts are introduced and extracted from one bath to another, either draining them or not, as indicated below.

The operations are performed at a room temperature between 18 and 25 °C.

When the pieces are gripped to introduce them in the bath, attention should be paid to the fact that the gripping points are not coated. Therefore, the gripping surface must be reduced to a minimum extent.

The bath tanks do not have necessarily to comply with any special requisite except, obviously, that they should not react with the solution.

Surface catalysis

The object of the first stage consists of depositing on the active points of the surface a series of small particles of a catalyst metal that will serve as focuses for the copper deposition.

For this purpose, the piece is subject to three operations:

- Regeneration of active points.
- Adsorption of the catalyst ions.
- Reduction of the catalyst ions.

a) Regeneration of active points

To regenerate the active points, the graphite piece is preferably submersed into a acetone bath for a period exceeding one minute -depending on the graphite- although sometimes the period should be of ten minutes, which is the recommended time.

Afterwards, the piece is taken out from the bath and dried at open air during five minutes, to eliminate the acetone.

b) Adsorption of the catalyst ions

After the regeneration, the piece is introduced inside a sensitization bath (Table 1, formula 2) of cobalt salts or nickel during 10 minutes.

c) Reduction of the catalyst ions.

The piece is removed from the previous bath and drained for 1 minute to avoid that the remnants of the solution are reduced, forming metal particles suspended in the solution.

The dried piece is introduced inside a reducer bath of potassic borohydride (Table 1 formula 3) during 30 minutes where the chemically absorbed cations are reduced to metal.

Deposition of the copper layer through self-catalytic reduction

During the second stage, a layer of crystalline compact metallic copper is deposited in two stages.

a) Rinsing

To eliminate the borohydride adhered to the piece, which would damage the metallization solution, the piece is submersed into an aqueous solution bath formed by $Co(CH_3COO)2$ or $CoCl2$ (table I, formula 4) for 5 minutes.

b) Copper plating

After it has been taken out from the rinsing solution, the piece is submersed into the plating bath for a period ranging between 0.5 and 10 hours, and the pH value is set to 12 throughout the whole process.

The plating solution contains a copper salt, a salt sequester, a reducer and additives (Table I, formula 5).

Deposition of the nickel layer through self-catalytic reduction

Once the copper layer has been deposited, to deposit a nickel layer it is necessary to introduce the piece into a nickel plating bath (Table I, formula 6) for a minute, and then, in a reducing solution identical to the one previously used to reduce the catalyst (Table I, formula 3).

Then, the piece is taken to the nickel plating bath, where the sequester is sodium citrate and the reducer, hypophosphite (Table I, formula 6) for a period ranging between 5 and 10 hours. Finally, the piece is rinsed under abundant water.

Throughout the whole plating process, the pH should be kept between 8.7 and 9, by means of NaOH.

Plating baths

Composition of the baths

a) Activation bath (formula I)

To achieve that the catalyst is fixed on the graphite surface, it is necessary to regenerate the active points which are saturated with molecules and atoms adhered by chemical absorption. This is done by eliminating the molecules or atoms by chemical attack or dissolution.

The best results have been achieved by submersing the graphite in very volatile and water soluble solvents, especially acetone. After eliminating the saturating product, the solvent shall be released from the surface, mainly through evaporation and the rest is eliminated by solving them in the aqueous plating bath.

b) Catalysation bath (Formula 2)

The catalyst solution can be Ni, Pd, Co and, in general, any transitional metal. In this stage, the metal cations are adsorbed through chemical adsorption at the active surface points.

c) Reducer bath (formula 3)

The reducer bath consists of an alkaline solution of potassic borohydride with pH = 13, which has to be prepared immediately before using it since it is spontaneously decomposed.

d) Rinsing bath (formula 4)

The only task of the rinsing bath is to carry the borohydride remnants.

e) Plating baths (formulæ 5 and 6)

Many metal salts of Cu and Ni can be used, and those appearing in the formula have been chosen for performance reasons.

An increase in the concentration of plating cation of the plating solution increases in turn the reduction reaction speed which, on the other hand, is also catalysed with the deposited metal itself. When the concentration exceeds a predetermined value and there are solid particles suspended, the speed of the bulk solution increases and a generalized reaction takes place, thereby decomposing the solution. The solution will not be decomposed during the plating, provided that the decomposing time is longer than the plating time.

The plating concentrations and times defined in the invention are optimised for a room temperature ranging between 18 and 25 °C. At higher temperatures, the invention can also work provided that the concentrations and times are adjusted accordingly. However, this would entail the additional evaporation problems that the patent intends to avoid.

Table I
Baths for the self-catalytic plating process

Formula Nº	PRODUCTS	BATHS					
		Regeneration	Catalysis		Rinsing	Copper plating	Nickel plating
			Sensitivization	Reduction			
1	Acetone	99%					
2	$CoCl_2$ or $NiCl_2$		0.2 M				
3	KBH_4 NaOH			0.1 M pH 13			
4	$CoCl_2$ or $Cu(CH3COO)_2$				0.01 M		
5	$CuCl_2$ or $Cu(CH3COO)_2$ $CoCl_2$ or $Co(CH3COO)_2$ $[Cu]+[Co]/[Na_2EDTA]$ HCHO Polyethylengycol 400 NaOH					0.02-0,06M 0.015 M 1 0.2 M 15 ml/l pH 12	
6	$NiCl_2$ $CoCl_2$ [Ni]+[Co]/[Sodium citrate] NH_4Cl NaH_2PO_2 NH_3 NaOH						0.15-0.38 M 0.02 M 1 1.0 M 0.6 M 0.5 M pH 8.7 - 9.0

Preparation of baths

The solution must be prepared in accordance with the sequence of operations defined in Table 2, in order to achieve a good dissolution of the products.

Table 2

Sequence of solution preparation		
Order of addition	**Copper plating bath**	**Nickel plating bath**
1	Copper salt Cobalt salt Na_2EDTA (solved with NaOH)	Nickel salt Cobalt salt Sodium citrate
2	Add water and solve by agitation	Add water and solve by agitation
3	Polyethylenglycol 400	NaH_2PO_2 and NH_3
4	HCHO (at the plating stage)	Set pH = 9 through NaOH (at the plating stage)
5	Set pH = 12 through NaOH	Adjust volume by adding water
6	Adjust volume by adding water	

Regeneration

The baths used throughout the whole plating process should be replaced or regenerated after several uses, since they get damaged or consumed.

a) Rinsing bath

The rinsing bath used before submersing the piece inside the copper plating bath gets corrupted by exhaustion of CO^{2+}, which is reduced to metallic Cobalt dispersed within the solution. It is regenerated by adding $Co(CH_3\text{-}COO)_2$ or $CoCl_2$ and filtering it.

b) Copper and nickel plating bath

The copper plating bath gets exhausted as the plating progresses. When the plating speed decreases, a regeneration process must be implemented. It is done by filtering, by adding the consumed copper salt and the necessary amount of Na_2EDTA to dissolve all the copper, and by adding the consumed HCHO and setting the pH to 12 by means of NaOH.

The nickel plating bath undergoes a process similar to the one of the copper bath. therefore, it must also be regenerated by filtering it and adding the nickel salt and the necessary NaH_2PO_2 and NH_3 to restore the consumed products.

Advantages of the invention

The metal coating offers the following advantages when compared with the previously existing methods:

1. The metal coating does not increase the volume of the residues, as it happens in the case of concrete. Therefore, the same stacking system used in the nucleus can be applied.

2. It is a reversible process, i.e., the coating can be eliminated in the future, if deemed appropriate, or another coating can be applied over the first one. This enables to check its condition at any time.

3. It is possible to decrease the lixiviation over a 90% with thicknesses of 4 to 40 µm while in the case of other coatings, a thickness of several centimetres is needed.

4. A humid process is applied, thus eliminating the gas and aerosol issues.

5. The application can be easily automated through a conventional technology.

6. It is applied at room temperature and pressure.

PROCEDURE FOR THE EXECUTION OF THE INVENTION

The essays described hereinafter help to illustrate the process:

Example No. 1

Complete example of plating

The solutions 1, 2, 3, 4, 5 (this bath with a concentration of copper of 0.06 M) and 6 (this bath with a concentration of nickel of 0.28 M) were prepared (Table I) according to the procedure described above.

A specimen of graphite sheath of approx. 75 x 20 x 20 mm of a fuel element which had remained inside the nucleus of the reactor during a full period was taken.

It was handled by means of a plastic clamp holding it by two points, hung from a nylon yarn.

Six beakers of 250 ml were prepared and numbered from 1 to 6, and filled with 225 ml of solutions 1, 2, 3, 4, 5 and 6, respectively.

The whole test was carried out maintaining the bath tanks thermostatted at 25 °C.

The piece was submitted to the following successive operations:

a) The piece was introduced into bath no. 1 for 10 minutes.
b) The piece was removed from bath 1 and dried at open air for 5 minutes.
c) The piece was introduced in the catalyst bath no. 2 for 10 minutes.
d) The piece was removed from bath no. 2 and drained for 1 minute
e) The piece was introduced into the reducing solution bath no. 3 for 30 minutes.
f) The piece was removed from bath no. 3 and drained for 1 minute.
g) The piece was introduced into the rinsing solution bath no. 4 for 5 minutes.
h) The pH of the plating solution bath no. 5 was adjusted to value 12 and the piece was introduced into it for 4 hours.
i) The piece was removed from bath no. 5 and drained for 1 minute. It looked as copper plated.
j) The pH of the plating solution bath no. 5 was adjusted to value 9 and the piece was introduced into it for 1 minute.
k) The piece was removed from bath no. 5 and introduced into the reducing solution bath no. 3 for 10 minutes.
l) The piece was removed from bath no. 3 and introduced in the plating solution bath no. 5 for 4 hours. When removed, the piece looked nickel coated.
m) The piece was rinsed under abundant water.

The piece was dried inside a lab drier with silica gel and was subsequently weighted. It was appreciated that the weight had increased some 8 µm.

Example No. 2

Verification of the efficiency of the plating

Two specimens of irradiated graphite sheath of approx. 75 x 20 x 20 mm obtained from the same area were used to verify the efficiency of the plating.

The content of Co-60 present at the graphite dust collected from cutting area was analyzed.

One of the pieces was plated following the procedure of Test No. 1, and the other was left unplated.

They were handled by means of a plastic clamp holding them by two points, hung from a nylon yarn.

2 beakers of 250 ml were prepared and filled with 225 ml of a NaCl solution 0.1 M and the pieces were submersed in it by means of the nylon yarn.

The whole test was carried out maintaining the beaker tanks thermostatted at 40 °C.

After 1, 3, 7, 10, 14, 21 and 28 days after having introduced the piece, it was removed from the solution and replaced by another new one with the same characteristics.

The content of Co-60 present on each solution contained in the beakers was analyzed each time the solution was renewed.

The total content of Co-60 lixiviated with each probe was calculated according to the following formula:

$$A_T = 225\,\Sigma\,A_n$$

Where:
A_T = Total activity lixiviated at the probe, in Bq.
A_n = Activity in Bq/ml of the solution taken after n days.
n = number of days (1, 3, 7, 10, 14, 21 and 28)
The efficiency of the coating was expressed as a percentage of Co-60 not lixiviated thanks to the coating, and it was calculated using the following formula:

$$\text{Efficiency} = \frac{A_{SC} - A_C}{A_{SC}} \times 100$$

Where:
A_{SC} is the total lixiviated activity of the non-coated piece, in Bq
A_C is the total lixiviated activity of the coated piece, in Bq.
The efficiency was around 90 and 97%.

Example no. 3

Need for a prior treatment

Two specimens of non irradiated graphite of the same characteristics as that used in the reactor were obtained and numbered as pieces 1 and 2.

They were handled by means of a plastic clamp holding them by two points, hung from a nylon yarn.

5 baths of 225 ml were prepared numbered 1 to 5 in a beaker of 250 ml which contained the solutions nos. 1, 2, 3, 4 and 5 (this beaker had a copper concentration of 0,06 M) respectively.

The whole test was carried out keeping the beaker tanks thermostatted at 25 °C.

The pieces were submitted to the following successive operations:

a) Piece 1 was introduced into bath no. 1 for 10 minutes.
b) The piece was removed from bath 1 and dried at open air for 5 minutes.
c) Pieces 1 and 2 were introduced in the catalyst bath no. 2 for 10 minutes.
d) Pieces 1 and 2 were removed from bath no. 2 and drained for 1 minute
e) Pieces 1 and 2 were introduced into the reducing solution bath no. 3 for 30 minutes.
f) Pieces 1 and 2 were removed from bath no. 3 and drained for 1 minute.
g) Pieces 1 and 2 were introduced into the rinsing solution bath no. 4 for 5 minutes.
h) Pieces 1 and 2 were introduced into the plating solution bath no. 5 for 4 hours.

When the pieces were taken out, piece 1 - which had been treated with acetone - showed a metal coating while piece 2 - which has not been subject to this treatment - had no coating.

Example no. 4

Need for a reactivation to deposit the second layer

Two specimens of non irradiated graphite of the same characteristics of that used in reactor were taken and numbered as pieces 1 and 2.

They were handled by means of a plastic clamp holding them by two points, hung from a nylon yarn.

6 beakers of 250 ml were prepared numbered 1 to 6 with 225 ml of solutions nos. 1, 2, 3, 4, 5 (this bath had a copper concentration of 0,06 M) and 6 (this with a nickel concentration of 0,28 M) respectively.

The whole test was carried out keeping the beaker tanks thermostatted at 25 °C.

The pieces were submitted to the following successive operations:

a) The pieces were introduced into bath no. 1 for 10 minutes.
b) They were removed from bath 1 and dried at open air for 5 minutes.
c) They were introduced in the catalyst bath no. 2 for 10 minutes.
d) They were removed from bath no. 2 and drained for 1 minute
e) They were introduced into the reducing solution bath no. 3 for 30 minutes.
f) They were removed from bath no. 3 and drained for 1 minute.
g) They were introduced into the rinsing solution bath no. 4 for 5 minutes.
h) The pH of the plating solution bath no. 5 was adjusted to value 12 and the pieces were introduced into it for 4 hours.
i) They were removed from bath no. 5 and drained for 1 minute. It looked as copper plated.

j) The pH of the plating solution bath no. 6 was adjusted and the piece 1 was introduced into it for 1 minute.

k) The piece 1 was introduced into the reducing solution bath no.3 for 10 minutes.

l) The piece 1 was removed from bath no. 3 and piece 2 from bath no. 5 and drained for 1 minute. Then, both pieces were introduced into the plating solution bath no. 6 for 4 hours.

When the pieces were taken out, piece 1 seemed to have a nickel coating and piece 2 only a copper one.

The aim of the implementation of the above examples is to evidence the feasibility and limitations of the conditions of the patent and can not represent a limitation to the claims.

Claims

1. A procedure for the metallisation conditioning of radioactive graphite from nuclear plants or nuclear plant dismantling the aim of which is to minimize the lixiviability of the radionucleids contained and characterized by the following successive operations: a prior conditioning of the graphite surface by submersing the piece to be water-tighten in a bath with a chemical solvent that releases its active points; the piece is dried at open air to eliminate the solvent; a subsequent treatment of catalyst activation of the graphite surface so treated through a chemical absorption of cations of a catalyst metal on the active points of the surface and the subsequent reduction of such points with an aqueous solution of an appropriate chemical reductor; the rinsing of the piece with a cobalt salt solution; an electrolytic deposition without electrode of a copper coating by submersing a metallization bath that contains a copper salt, a reductor, a sequester and additives; immersion into a solution that contains a nickel salt, a reductor, a sequester and additives, similar to that to be used later on to deposit the nickel layer; a reactivation treatment of the metallized surface of the graphite and rinsed by submersion in a bath with an aqueous solution of an appropriate chemical reductor; an electrolytic deposition without electrode of a second nickel layer through submersion in a metallization bath containing a nickel salt, a reductor, a sequester and additives; and the rinsing through a final cleansing of the water-tighten graphite piece.

2. A procedure for the metallisation conditioning of radioactive graphite according to claim 1, characterized because all the successive operations are carried out at a room temperature ranging between 18º and 25º C and at room pressure and because the graphite piece will be dried at open air once it has been taken out of the prior conditioning bath and dried once it has been taken out of the remaining baths.

3. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 2, characterized because the chemical agent where the piece is introduced between 1 and 10 minutes for the prior conditioning of the surface is an organic solvent of a high volatility, polar and water soluble, preferably acetone.

4. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 2 characterized because the copper metallisation bath is an aqueous solution consisting of 0.02 and 0.06 M of $CuCl_2$ or $Cu(CH_3COO)_2$, 0.015 of $CoCl_2$ or $Co(CH_3COO)_2$, 0.2 M of HCHO, 1,5% volume of Polyethylenglycol 400 and Na_2EDTA in a solution equimolar to the total cobalt and copper concentrations, prepared according to the following sequence of operations: first of all, the necessary quantities of copper salt, cobalt salt and Na_2-EDTA - according to the previous formulation - are introduced in a container along with the minimum quantity of water necessary to solve them by agitation; secondly, the Polyethylenglycol 400 is poured into this solution; thirdly, the HCHO is poured when the metallization starts; fourthly, the pH is adjusted to 12 through an aqueous solution of NaOH and finally the necessary volume is completed with water to reach the specific concentrations.

5. A procedure for the metallisation conditioning of radioactive graphite according to claims 1, 2 and 4 characterized because the time that the piece should be kept in the copper metallization bath shall be of over 30 minutes.

6. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 4 characterized because the copper metallization solution can be generated through filtering and by adding the $CuCl_2$, Na_2EDTA and HCHO consumed.

7. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 2 characterized because the chemical reductor of the superficial reactivation bath of the copper plated graphite is an aqueous solution recently prepared with a composition of 0.1 M of KBH_4 and 0.1 M of NaOH and a reactivation time of 5 minutes.

8. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 2 characterized because the nickel metallization bath is an aqueous solution consisting of between 0.15 and 0.38 M of $NiCl_2$, 0.02 of $CoCl_2$, 1.0 M of NH_4Cl, 0.5 M of NH_3, 0.6 M of NaH_2PO_2 and sodium citrate in a concentration equimolecular to

the total cobalt and nickel concentrations, prepared according to the following sequence of operations: first of all, the necessary quantities of nickel salt, cobalt salt and sodium citrate - according to the previous formulation - are introduced in a container along with the minimum quantity of water, while agitating them, necessary to solve them; secondly, successively pour over this solution the NH_4Cl, the NaH_2PO_2 and the NH_3; thirdly, adjust the pH to 9 through NaOH at the time of the metallization; fourthly, fill in with the necessary volume of water to reach the specified concentrations.

9. A procedure for the metallisation conditioning of radioactive graphite according to claims 1, 2 and 4 characterized because the piece is kept in the nickel metallization bath during a period of 0.5 to 10 hours.

10. A procedure for the metallisation conditioning of radioactive graphite according to claims 1, 2 and 4 characterized because while the piece is kept in the nickel metallisation bath, the pH shall be adjusted through NaOH to a value ranging between 7 and 10.

11. A procedure for the metallisation conditioning of radioactive graphite according to claims 1 and 8, characterized because nickel metallization solution can be generated by filtering and adding $NiCl_2$, NaH_2PO_2 and NH_3 to restore the consumed products.

12. A procedure for the metallisation conditioning of radioactive graphite according to claim 1 characterized because the $CoCl_2$ solution can be generated filtering and restoring the cobalt consumed by adding the necessary $CoCl_2$.