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(12) (19) (CA) **Brevet-Patent**

(11) (21) (C) **2,142,003**
(22) 1995/02/07
(43) 1995/08/25
(45) 2000/06/27

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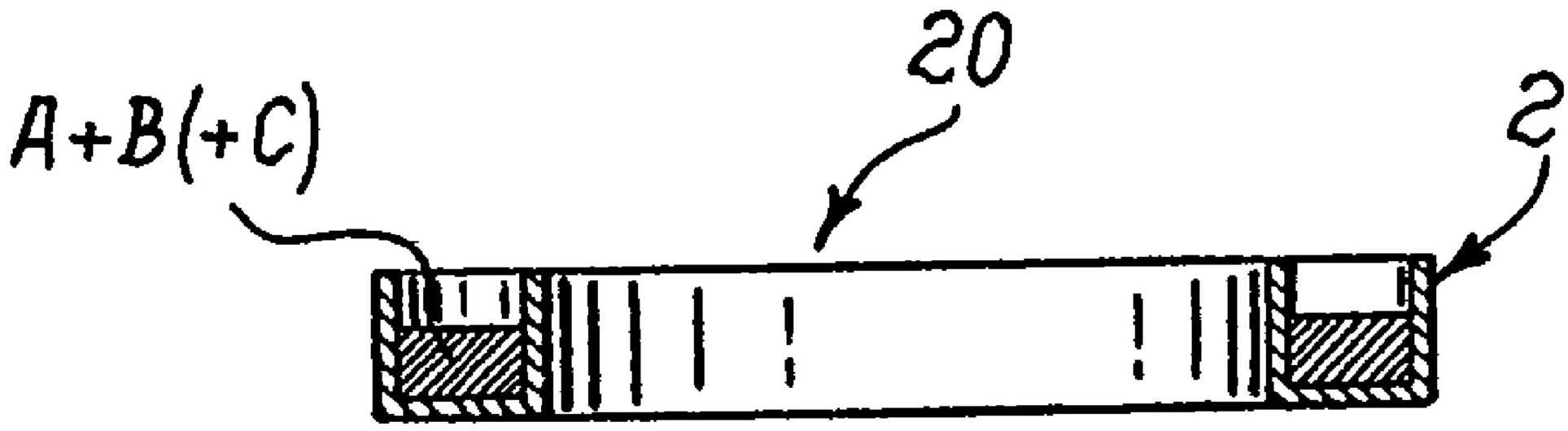
(51) Int.Cl.[6] H01J 7/10
(30) 1994/02/24 (MI94A000341) IT

(54) **COMBINAISON DE MATERIAUX SERVANT DANS DES DISPOSITIFS D'ALIMENTATION EN MERCURE, METHODE D'OBTENTION ET DISPOSITIFS OBTENUS**

(54) **COMBINATION OF MATERIALS FOR MERCURY-DISPENSING DEVICES, METHOD OF PREPARATION AND DEVICES THUS OBTAINED**



(57) A mercury-dispensing combination suitable to release an amount of mercury higher than 60% during the activation step, even after partial oxidation, includes a mercury-dispensing intermetallic compound A with Hg and a second metal selected among Ti, Zr and mixtures thereof, as well as a promoting alloy or intermetallic compound B including Cu and a second metal selected among Sn, In or Ag or combinations thereof. There is also disclosed a mercury-dispensing device containing a combination of materials A and B, in addition to a process for introducing mercury into electron tubes consisting in the introduction of one of said devices inside the open tube and then heating thereof at a temperature between 550 and 900 °C after the tube sealing in order to get Hg free.

"A COMBINATION OF MATERIALS FOR MERCURY-DISPENSING DEVICES, METHOD OF PREPARATION AND DEVICES THUS OBTAINED"

ABSTRACT

A mercury-dispensing combination suitable to release an amount of mercury higher than 60% during the activation step, even after partial oxidation, includes a mercury-dispensing intermetallic compound A with Hg and a second metal selected among Ti, Zr and mixtures thereof, as well as a promoting alloy or intermetallic compound B including Cu and a second metal selected among Sn, In or Ag or combinations thereof. There is also disclosed a mercury-dispensing device containing a combination of materials A and B, in addition to a process for introducing mercury into electron tubes consisting in the introduction of one of said devices inside the open tube and then heating thereof at a temperature between 550 and 900°C after the tube sealing in order to get Hg free.

"A COMBINATION OF MATERIALS FOR MERCURY-DISPENSING DEVICES, METHOD OF PREPARATION AND DEVICES THUS OBTAINED"

The present invention relates to a combination of materials for the production of mercury-dispensing devices, to the mercury-dispensing devices thus produced and to a process for the introduction of mercury inside electron tubes.

The use of small amounts of mercury in electron tubes such as, for example, mercury-arc rectifiers, lasers, various kinds of alphanumeric displays and, particularly, fluorescent lamps is well known in the art.

A precise dosage of mercury inside these devices is extremely important for the quality of the devices and most of all for ecological reasons. In fact, the high toxicity of this element implies serious problems of ecological nature upon end-life disposal of the devices containing it, or in case of accidental break-up of the devices. These problems of ecological nature impose the use of amounts of mercury as small as possible, compatibly with the functionality of the tubes. These considerations have been lately included also in the legislative sphere, and the trend of the recent international regulations is to establish top limits for the amount of mercury which can be introduced into the devices: for example, for standard fluorescent lamps the use of a total amount of Hg not greater than 10 mg per lamp has been suggested.

In the past mercury was introduced into the tubes in liquid form. However, the use of liquid mercury first of all poses problems concerning the storing and handling in the plants for the production of tubes, due to its high vapor pressure also at room temperature. Secondly, a common drawback of the techniques for the introduction into the tubes of mercury in liquid form is the difficulty in precisely and reproducibly dosing volumes of mercury in the order of microliters, which difficulty usually takes to the introduction of amounts of the element much higher than needed.

These drawbacks have taken to the development of various techniques in alternative to the use of liquid mercury in free form.

The use of liquid mercury contained in capsules is disclosed in several prior art documents. This method is described, for example, in US patents nos.4.823.047 and 4.754.193, referring to the use of metallic

capsules, and in US patents nos.4.182.971 and 4.278.908 wherein the mercury container is made of glass. After closing the tube, the mercury is released by means of a heat treatment which causes the breakage of the container. These methods generally have some drawbacks. First of all, the production of the capsules and their mounting inside the tubes may be complicated, especially when they have to be introduced inside small-size tubes. Secondly, the breakage of the capsule, particularly if it is made of glass, may produce fragments of material which can jeopardize the tube quality, so much that US patent no.4.335.326 discloses an assembly wherein the mercury-containing capsule is in turn located inside a capsule acting as a shield for the fragments. Moreover, the release of the mercury is often violent, with possible damages to the inner structure of the tube. Finally, these systems still have the drawback of employing liquid mercury, and therefore they do not completely solve the problem of the precise and reproducible dosage of few milligrams of mercury.

US patent no.4.808.136 and the European patent application EP-568.317 disclose the use of tablets or small spheres of porous material soaked with mercury which is then released by heating once the lamp is closed. However, also these methods require complicated operations for the loading of mercury into the tablets, and the released amount of mercury is difficult to be reproduced.

These problems are overcome by US patent no.3.657.589 in the name of the Applicant, which discloses the use of intermetallic compounds of mercury having the general formula $Ti_xZr_yHg_z$, wherein x and y may vary between 0 and 13, the sum (x+y) may vary between 3 and 13 and z may be 1 or 2.

These compounds have a temperature of mercury-release start variable according to the specific compound, however they are all stable up to about 500°C both in the atmosphere and in evacuated volumes, thus resulting compatible with the operations for the assembly of the electron tubes, during which the mercury-dispensing devices may reach temperatures of about 400°C. After closing the tube, the mercury is released from the above-cited compounds by an activation operation, which is usually carried out by heating the material between 750°C and 900°C for about 30 seconds. This heating may be accomplished by laser radiation, or by induction heating of the metallic support of the

Hg-dispensing compound. The use of the Ti_3Hg compound, manufactured and sold by the Applicant under the trade name St505 results particularly advantageous; in particular, the St505 compound in sold in the form of compressed powder in a ring-shaped container or of compressed powder in pills or tablets, under the trademark "STAHGSORB", or in the form of powders laminated on a metallic strip, under the trademark "GEMEDIS".

These materials offer various advantages with respect to the prior art:

- as mentioned above, they avoid the risks of mercury evaporation during the cycle of production of the tubes, in which temperatures of about 350-400°C may be reached;
- as described in the cited US patent no.3.657.589, a getter material can be easily added to the mercury-dispensing compound with the purpose of chemisorption of gases such as CO, CO_2, O_2, H_2 and H_2O, which would interfère with the tube operation; the getter is activated during the same heat treatment for the release of mercury;
- the released amount of mercury is easily controllable and reproducible.

Despite their good chemical-physical characteristics and their great ease of use, these materials have the drawback that the contained mercury is not completely released during the activation treatment. In fact, the processes for the production of mercury-containing electron tubes include a tube-closing operation performed by glass fusion (e.g. the sealing of fluorescent lamps) or by frit sealing, i.e. welding two pre-shaped glass members by means of a paste of low-melting glass. During these operations, the mercury-dispensing device may undergo an indirect heating up to about 350-400°C; in this step the device is exposed to gases and vapours emitted by the melted glass and, in almost all industrial processes, to air. In these conditions, the mercury-dispensing material undergoes a surface oxidation, whose final result is a yield of about 40% of the total mercury content during the activation process.

The mercury not released during the activation operation is then slowly released during the life of the electron tube.

This characteristic, together with the fact that the tube must obviously work from the beginning of its life cycle, leads to the necessity of introducing into the device an amount of mercury which is about double

than that which would theoretically be necessary.

In order to overcome these problems, patent application EP-A-091.297 suggests the addition of Ni or Cu powders to the Ti_3Hg or Zr_3Hg compounds. According to this document, the addition of Ni and Cu to the mercury-dispensing compounds causes the melting of the combination of materials thus obtained, favouring the release of almost all the mercury in few seconds. The melting takes place at the eutectic temperatures of the systems Ni-Ti, Ni-Zr, Cu-Ti and Cu-Zr, ranging from about 880°C for the Cu 66% - Ti 34% composition to 1280°C for the Ni 81% - Ti 19% composition (atomic percent), though the document erroneously gives a melting temperature of 770°C for the Ni 4% - Ti 96% composition. The document acknowledges that the mercury-containing compound is altered during the tube working treatments, and it needs a protection; to this purpose, there is suggested to close the powder container by means of a steel, copper or nickel sheet which is broken during the activation by the pressure of the mercury vapor generated inside the container. This solution is not completely satisfactory: in fact, same as it happens in the methods employing capsules, mercury bursts out violently and can cause damages to portions of the tube; the manufacturing of the container is quite complicated, since it requires the welding of small-size metallic members. Furthermore, this document does not contain experimental data to support the assessed good mercury-release characteristics of the combinations indicated.

Therefore, the object of the present invention is to provide an improved combination of materials for dispensing mercury in the electron tubes, which allows to overcome one or more drawbacks of the prior art.

In particular, the object of the present invention is first of all to provide an improved combination of materials for dispensing mercury which is capable of releasing amounts of mercury higher than 60% during the activation step, even after partial oxidation, so as to be able to reduce the total amount of employed mercury.

Another object of the present invention is to provide mercury-dispensing devices containing the combination of materials of the invention.

Still another object is to provide a process for introducing mercury by means of the devices of the invention into the electron tubes which require

said element.

According to the present invention, these and other objects are achieved by using a mercury-dispensing combination of materials made up of:

- a mercury-dispensing intermetallic compound A including mercury and a second metal selected among titanium, zirconium and mixtures thereof; and
- an alloy or an intermetallic compound B including copper, a second metal selected among tin, indium, silver or combinations thereof, and possibly a third metal selected among the transition elements, wherein the transition metal is present in an amount not greater than 10% of the overall weight of component B.

A mercury-dispensing device of the invention contains a combination of said materials A and B, possibly further containing a getter material C, while a process according to the invention shows the features of claim 23.

Further objects and advantages of the present invention will be apparent from the following detailed description referring to the annexed drawings wherein:

Fig.1 is a perspective view of a mercury-dispensing device of the present invention according to a possible embodiment thereof;

Figs.2 and 2a are, respectively, a top plan view and a sectional view along II-II of a device of the invention according to another possible embodiment;

Figs.3, 3a and 3b are, respectively, a top plan view and two sectional views along III-III of a device of the invention according to a further embodiment, in two possible variations.

Component A of the combination of the present invention, hereafter also defined mercury dispenser, is an intermetallic compound corresponding to formula $Ti_xZr_yHg_z$, as disclosed in the cited US patent no.3.657.589, to which reference is made for further details. Among the materials corresponding to said formula, Zr_3Hg and, particularly, Ti_3Hg are preferred.

Component B of the combination of the present invention has the function of favouring the release of mercury from component A, and hereafter will also be defined promoter. This component is an alloy or an intermetallic compound including copper, a second metal selected among

tin, indium, silver or combinations thereof, and possibly a third metal selected among the transition elements.

The atomic ratios between the elements of the binary or ternary compositions making up component B of the combinations of the present invention vary according to the constituting elements.

In the case of binary alloys of copper with tin or indium, the optimum ranges are the following:

- Cu-Sn: from about 3% to about 63% of copper on a weight basis
- Cu-In: from about 40% to about 60% of copper on a weight basis

It is also possible to use alloys of three or more metals obtained from the preceding ones by adding an element selected among the transition metals in an amount not greater than 10% of the overall weight of component B.

In the case of Cu-Ag binary alloys, the ratio between the two components may range from about 10% to about 80% of Cu on a weight basis, and preferably between 20% and 50% of Cu on a weight basis.

Among the above-mentioned compositions, those including Sn-Cu are particularly preferred for the easy preparation and the good mechanical characteristics, and most of all the composition containing from 54,5% to 56,5% (atomic percent) of copper, corresponding to the non-stechiometric compound Cu_6Sn_5.

The weight ratio between components A and B of the combination of the invention may vary within a wide range, but it is generally included between 20:1 and 1:20, and preferably between 10:1 and 1:5.

Components A and B of the combination of the invention may be employed in various physical forms, not necessarily the same for the two components. For example, component B may be present in the form of a coating of the metallic support, and component A as a powder adhered to component B by rolling. However, the best results are obtained when both components are in the form of a fine powder, having a particle size lower than 250 µm and preferably between 10 and 125 µm.

The present invention, in a second aspect thereof, relates to the mercury-dispensing devices which use the above-described combinations of A and B materials.

As previously mentioned, one of the advantages of the materials of the invention with respect to prior art systems is that they do not need a

mechanical protection from the environment, thus not posing the limit of a closed container. Consequently, the mercury-dispensing devices of the present invention can be manufactured with the most different geometric shapes, and materials A and B of the combination can be employed without support or on a support, usually metallic.

Some classes of electron tubes for which the mercury dispensers are intended further require, for their correct operation, the presence of a getter material C which removes traces of gases such as CO, CO_2, H_2, O_2 or water vapor: it is the case, for example, of fluorescent lamps. For these applications, the getter can be advantageously introduced by means of the same mercury-dispensing device, according to the manners described in the cited US patent no.3.657.589.

Examples of getter materials include, among the others, metals such as titanium, zirconium, tantalum, niobium, vanadium and mixtures thereof, or alloys thereof with other metals such as nickel, iron, aluminum, like the alloy having a weight percentage composition Zr 84% - Al 16%, manufactured by the Applicant under the name St101, or the intermetallic compounds Zr_2Fe and Zr_2Ni, manufactured by the Applicant respectively under the name St198 and St199. The getter is activated during the same heat treatment by which mercury is released inside the tube.

The getter material C may be present in various physical forms, but it is preferably employed in the form of a fine powder, having a particle size lower than 250 μm and preferably between 10 and 125 μm.

The ratio between the overall weight of the A and B materials and that of the getter material C may generally range from about 10:1 to 1:10, and preferably between 5:1 and 1:2.

Some possible embodiments of the devices of the invention are illustrated hereunder with reference to the drawings.

In a first possible embodiment, the devices of the invention can simply consist of a tablet made up of compressed and unsupported powders of the A and B (and possibly C) materials, which for ease of production generally has a cylindrical or parallelepipedal shape; this latter possibility is shown in fig.1.

In the case of supported materials, the device may have the shape of a ring 20 as shown in fig.2, which represents a top plan view of the device, and in fig.2a which represents a cross-section along II-II of device 20. In

this case, the device is made up of a support 21 having the shape of a toroidal channel containing the A and B (and possibly C) materials. The support is generally metallic, and preferably of nickel-plated steel.

Alternatively, the device may be made in the shape of a strip 30 as shown in fig.3, which represents a top plan view of the device, and in figs.3a and 3b wherein a section along III-III of device 30 is depicted. In this case, support 31 consists of a strip, preferably made of nickel-plated steel, onto which the A and B (and possibly C) materials are adhered by cold compression (rolling). In this case, whenever the presence of the getter material C is required, materials A, B and C may be mixed together and rolled on one or both faces of the strip (fig.3a), but in a preferred embodiment materials A and B are placed on one surface of the strip and material C on the opposite surface, as shown in fig.3b.

The invention, in a further aspect thereof, relates to a method for introducing mercury into the electron tubes by using the above-described devices.

The method includes the step of introducing inside the tube the above-described mercury-dispensing combination of materials and preferably in one of the above-described devices 10, 20 or 30, and then the combination heating step to get mercury free. The heating step may be carried out with any suitable means such as, for example, by radiation, by high-frequency induction heating or by having a current flow through the support when the latter is made of a material having a high electric resistivity. The heating is effected at a temperature which causes the release of mercury from the mercury-dispensing combination, comprised between 500 and 900°C for a time of about 10 seconds to one minute. At temperatures lower than 500°C mercury is almost not dispensed at all, whereas at temperatures higher than 900°C there is the danger of the development of noxious gases by outgassing from the portions of the electron tube adjacent to the device or of the formation of metal vapors.

The invention will be further illustrated by the following examples. These non-limiting examples illustrate some embodiments intended to teach to those skilled in the art how to put in practice the invention and to show the accomplishment of the invention which is considered the best.

Examples 1 to 9 concern the preparation of the releasing and promoting materials, while examples 10 to 23 concern the tests for the mercury

release after the heat treatment simulating the sealing operation. All the metals used for the preparation of alloys and compounds for the following tests have a minimum pureness of 99,5%. In the compositions of the examples all percentages are on a weight basis if not differently specified.

EXAMPLE 1

This example illustrates the synthesis of the mercury-dispensing material Ti_3Hg.

143,7 g of titanium are placed in a steel cradle and degassed by a furnace treatment at a temperature of about 700°C and a pressure of 10^{-6} mbar for 30 minutes. After cooling the titanium powder in an inert atmosphere, 200,6 g of mercury are introduced in the cradle by means of a quartz tube. The cradle is then closed and heated at about 750°C for 3 hours. After cooling, the product is ground until a powder passing through a 120µm mesh-size standard sieve is obtained.

The resulting material essentially consists of Ti_3Hg, as confirmed by a diffractometric test carried out on the powder.

EXAMPLES 2-10

These examples concern the preparation of the promoting alloys which make part of the combinations of the invention. The alloys are prepared by loading weighed amounts of the starting metals into alumina cradles which are then introduced in a vacuum induction furnace. The metal mixtures are heated at a temperature about 100°C higher than the melting temperature of the corresponding alloy, kept at that temperature for 5 minutes to encourage the homogeneity thereof, and finally cast into a steel ingot-mould. Each ingot is ground in a blade mill and the powder is sieved like in example 1. The respective amounts in grams of the metals used to produce the alloys are indicated in table 1. In the table, TM refers to a transition metal.

Table 1

EXAMPLE N.	Cu	Sn	In	Ag	TM
2	41	59	0	0	0
3	62	38	0	0	0
4	56	0	44	0	0
5	41	43	10	0	0
6	31	39	0	0	7 (Mn)
7	31	39	0	0	7 (Ti)
8	31	39	0	0	7 (Ni)
9	31	39	0	0	7 (Fe)
10	28	0	0	72	0

EXAMPLES 11-26

Examples 11 to 26 concern the tests for the mercury release from the mixtures after a heat treatment in air which simulates the conditions to which the device is subjected during the tube closing (hereafter generally referred to as sealing).

For the simulation of the sealing, 150 g of each powder mixture have been loaded in a ring-shaped container like in fig.2 and have been subjected to the following thermal cycle in air:

- heating from room temperature to 400°C in about 5 seconds;
- isotherm at 400°C for 30 seconds;
- cooling from 400°C to 350°C, requiring about 1 second;
- isotherm at 350°C for 30 seconds;
- spontaneous cooling to room temperature, requiring about 2 minutes.

Thereafter, the mercury release tests have been carried out on the thus treated samples by induction heating thereof at 850°C for 30 seconds inside a vacuum chamber and by measuring the mercury remained in the dispensing device through the method of the complexometric titration according to Volhart.

The results of the tests are summarized in examples 17-26 of table 2, which show the mercury-dispensing compound A, the promoting material B (the combination referring to examples 2-10 is indicated in brackets), the weight ratio between components A and B and the mercury yield.

The comparative examples are marked by a star.

Table 2

EXAMPLE N.	A	B	A/B	Hg
11*	Ti_3Hg	-	-	35,2
12*	Ti_3Hg	Cu	5/1	45,7
13*	Ti_3Hg	Cu	7/3	34,0
14*	Ti_3Hg	Sn	5/1	25,0
15*	Ti_3Hg	In	5/1	27,0
16*	Ti_3Hg	Ag	5/1	49,1
17	Ti_3Hg	Cu-Sn (2)	7/3	85,2
18	Ti_3Hg	Cu-Sn (2)	1/1	83,6
19	Ti_3Hg	Cu-Sn (3)	7/3	81,7
20	Ti_3Hg	Cu-In (4)	7/3	83,4
21	Ti_3Hg	Cu-Sn-In (5)	7/3	83,8
22	Ti_3Hg	Cu-Sn-Mn (6)	7/3	67,8
23	Ti_3Hg	Cu-Sn-Ti (7)	7/3	60,4
24	Ti_3Hg	Cu-Sn-Ni (8)	7/3	64,1
25	Ti_3Hg	Cu-Sn-Fe (9)	7/3	71,2
26	Ti_3Hg	Cu-Ag (10)	7/3	65,3

It may be noted from the data of table 2 that the combinations with promoter of the present invention allow mercury yields higher than 60% during the activation step, thus permitting the reduction of the overall mercury amount introduced in the electron tubes.

Furthermore, the combinations with promoter of the present invention offer another important advantage, consisting in the possibility of carrying out the activation operation at temperatures or with times lower than those allowed by prior art materials. In fact, in order to have industrially acceptable activation times, Ti_3Hg alone requires an activation temperature of about 900°C, whereas the present combinations allow the reduction of this temperature to about 850°C for the same time, or alternatively the reduction of the operation time at the same temperature; in both cases a double advantage is achieved of causing less pollution inside the tube due to the outgassing of all the materials present therein and of reducing the amount of energy required for the activation.

CLAIMS

1. A mercury-dispensing combination comprising a mercury-dispensing intermetallic compound A including mercury and a second metal selected among titanium, zirconium and mixtures thereof, characterized in that it further comprises a promoting compound B including copper, and a second metal selected among tin, indium or silver or combinations thereof.

2. A mercury-dispensing combination according to claim 1, characterized in that the promoting compound B includes copper, a second metal selected among tin or indium or combinations thereof, and a third metal selected among the transition elements, and in that the third metal is present in an amount not greater than 10% of the overall weight of the promoting compound B.

3. A mercury-dispensing combination according to claim 1, characterized in that the intermetallic compound A is Ti_3Hg.

4. A mercury-dispensing combination according to claim 1, characterized in that the promoting compound is a Cu-Sn alloy containing from 3% to 63% of Cu on a weight basis.

5. A mercury-dispensing combination according to claim 4, characterized in that the promoting compound is the non-stechiometric phase Cu_6Sn_5.

6. A mercury-dispensing combination according to claim 1, characterized in that the promoting compound is a Cu-In alloy containing from about 40% to about 60% of Cu on a weight basis.

7. A mercury-dispensing combination according to claim 6, characterized in that the promoting compound is a Cu-In alloy containing 44% of Cu on a weight basis.

8. A mercury-dispensing combination according to claim 1, characterized in that the promoting compound is a Cu-Ag alloy containing from about 10% to about 80% of Cu on a weight basis.

9. A mercury-dispensing combination according to claim 1, characterized in that the weight ratio between components A and B ranges from 20:1 to 1:20.

10. A mercury-dispensing combination according to claim 9, characterized in that the weight ratio between components A and B ranges from 10:1 to 1:5.

11. A mercury-dispensing device characterized in that it contains a combination of materials A and B of claim 1.

12. A mercury-dispensing device according to claim 11 characterized in that it further contains a getter material C.

13. A mercury-dispensing device according to claim 12, characterized in that the getter material C is selected among titanium, zirconium, tantalum, niobium, vanadium and mixtures thereof, or alloys of these metals with nickel, iron or aluminum.

14. A mercury-dispensing device according to claim 13, characterized in that the mercury dispenser A is Ti_3Hg, the promoting compound B is the non-stechiometric phase Cu_6Sn_5 and the getter material C is an alloy having the composition Zr 84% - Al 16% on weight basis.

15. A mercury-dispensing device according to claim 12, characterized in that the mercury dispenser A, the promoting compound B and the getter material C are in the form of powder.

16. A mercury-dispensing device according to claim 15, characterized in that it consists of a tablet of compressed powders of materials A, B and C.

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17. A mercury-dispensing device according to claim 15, characterized in that materials A, B and C are contained in a metallic support having a ring shape.

18. A mercury-dispensing device according to claim 15, characterized in that the combination of materials A and B is rolled on the surface of a support having the shape of a strip, and material C is rolled on the opposite surface of the same strip.

19. A mercury-dispensing device according to claim 12, characterized in that the ratio between the overall weight of materials A and B and the weight of the getter material C is between 10:1 and 1:10.

20. A mercury-dispensing device according to claim 19, characterized in that the ratio between the overall weight of materials A and B and the weight of the getter material C is between 5:1 and 1:2.

21. A mercury-dispensing device according to claim 15, characterized in that the mercury-dispensing material, the promoter and the getter are in the form of powders having a particle size lower than 250 μm.

22. A mercury-dispensing device according to claim 21, characterized in that the mercury-dispensing material, the promoter and the getter are in the form of powders having a particle size between 10 and 125 μm.

23. A process for introducing mercury inside electron tubes, characterized in that it consists in introducing into the tube one of the devices of claims 11 to 22, and heating the device to get mercury free at a temperature between 550°C and 900°C for a time between about 10 seconds and one minute after the tube sealing.

24. A process according to claim 23, characterized in that the electron tube consists of a fluorescent lamp.

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Fig.1

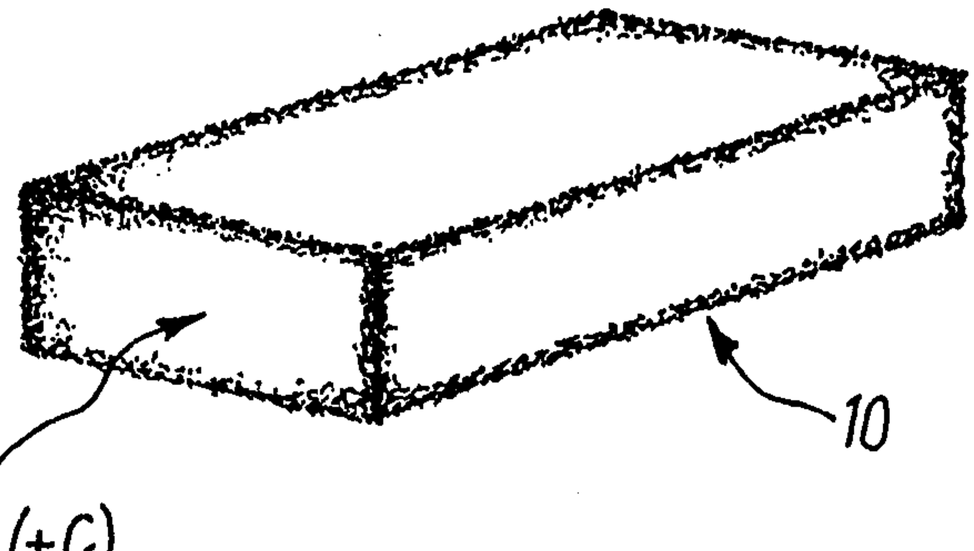
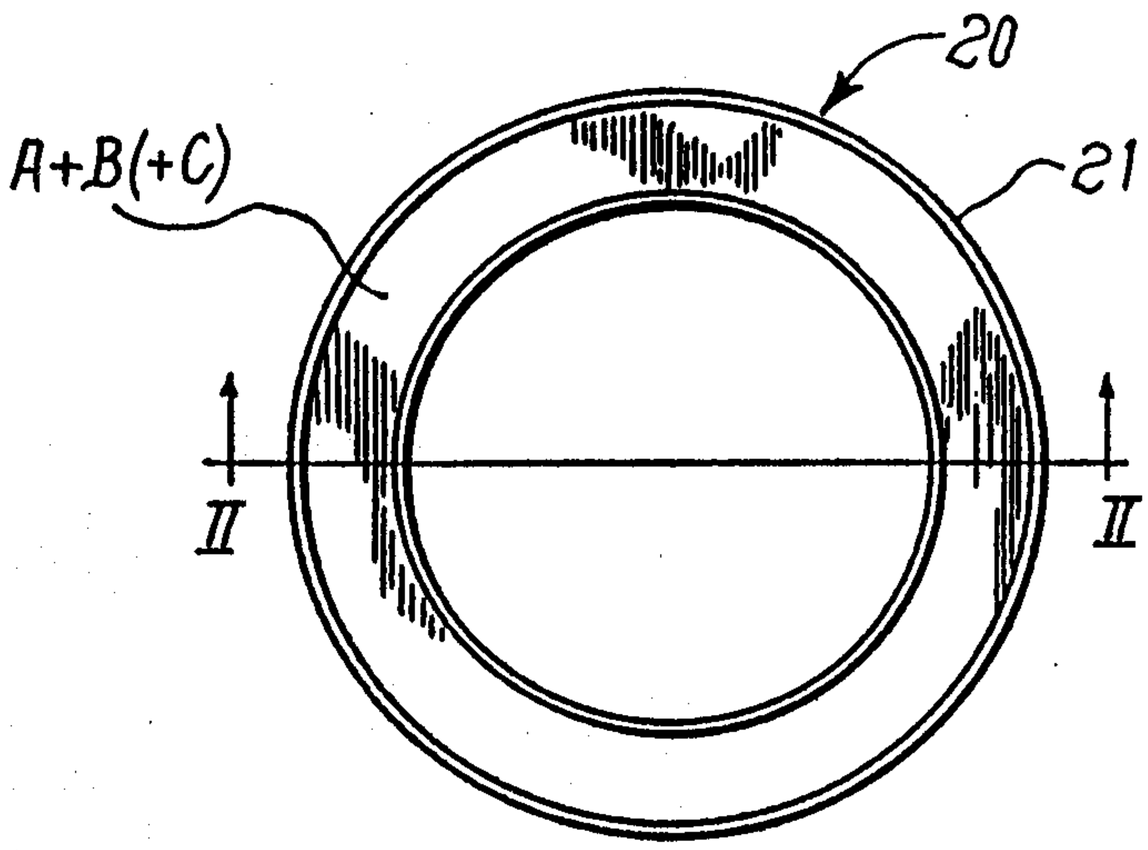


Fig.2



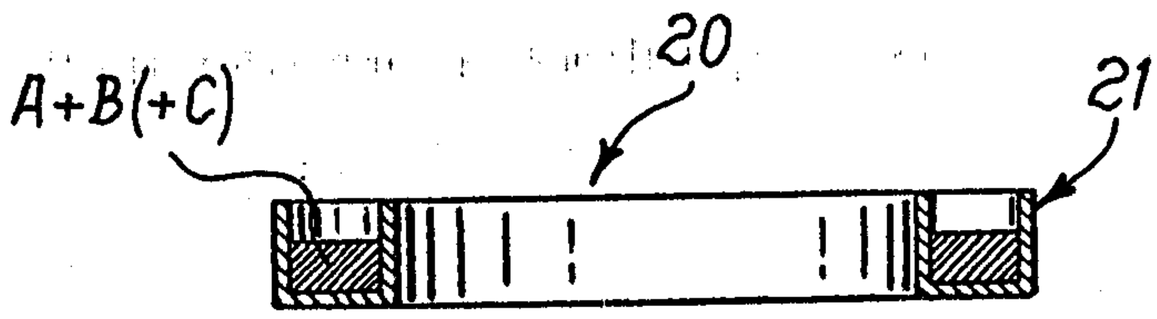


Fig.2a

Gowling, Strathy & Henderson

Fig. 3

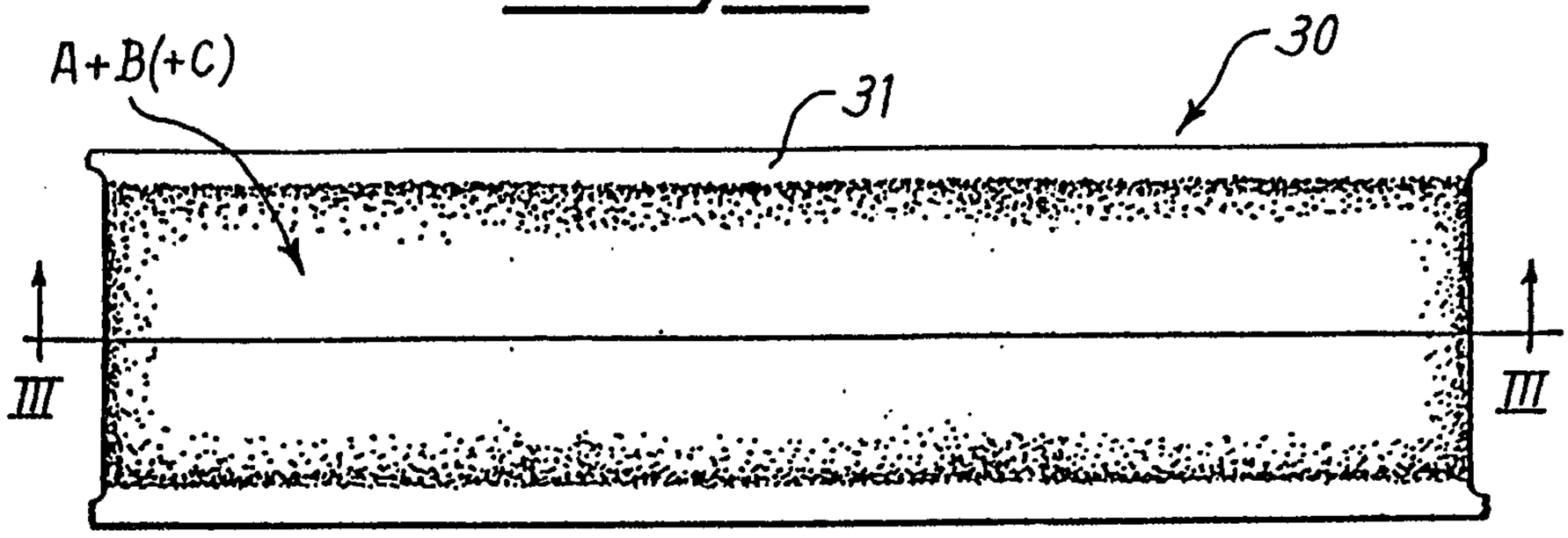


Fig. 3a

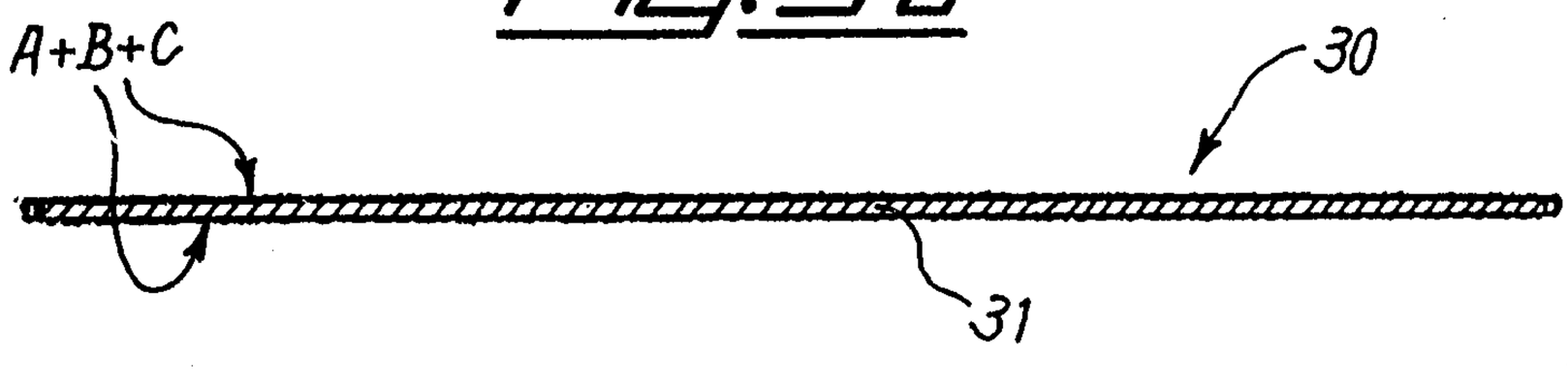


Fig. 3b