(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 980 077 A1

(12) EUROPEAN PATENT APPLICATION

(43) Date of publication:
16.02.2000 Bulletin 2000/07

(51) Int Cl.[7]: **G21G 4/06**

(21) Application number: **99500116.1**

(22) Date of filing: **09.07.1999**

(84) Designated Contracting States:
AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU MC NL PT SE
Designated Extension States:
AL LT LV MK RO SI

(30) Priority: **10.08.1998 ES 9801720**

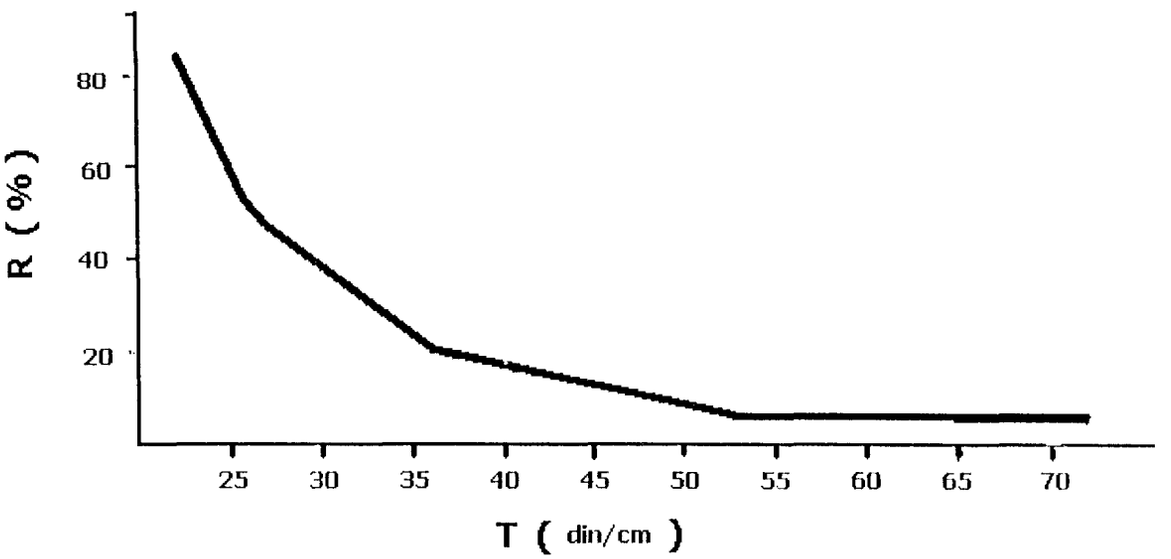
(71) Applicant: **CENTRO DE INVESTIGACIONES ENERGETICAS MEDIOAMBIENTALES Y TECNOLOGICAS (C.I.E.M.A.T.)**
E-28040 Madrid (ES)

(72) Inventor: **Gascon Murillo, Jose Luis D.**
28040 Madrid (ES)

(74) Representative: **Del Santo Abril, Natividad**
Oficina Garcia Cabrerizo, S.L.,
Vitruvio, 23
28006 Madrid (ES)

(54) Procedure for the incorporation of hazardous or costly onto aluminium supports with very low thickness of anodising

(57) It comprises, in general, the following stages: preparation of a solvent-additive mixture which complies to the conditions: surface tension less than 25 din cm^{-1}, steam pressure greater than 40 mm of Hg at 20°C and solid residue below 4.5 10^{-3} g/l, preparation of a 10^{-10} M solution of the substance to be incorporated into the solvent - additive mixture, removal of the excess and rubbing with paper soaked in the solvent - additive mixture; using a water - ethanol mixture for the platting of ^{241}Am on an anodising thickness of 4 μm in the manufacture of extensive radioactive sources.



EP 0 980 077 A1

Description

[0001] The subject matter of the present invention is a procedure to deposit hazardous or costly substances onto the anodised layer of an aluminium support.

[0002] The applications of the subject matter of the invention are, mainly the preparation of extensive radioactive sources and in general the deposit onto anodised aluminium supports of hazardous (radioactive, toxic, medical, etc.) or costly products (silver salts for photography, noble elements for decoration, antithermal ceramic elements, etc.)

[0003] The Spanish standard UNE 21-72-90 and international one ISO 8769 specify the characteristics that radioactive sources used for the calibration of superficial contamination monitors must fulfil. According to this standard, the area of the radioactive source must have a surface of at least 100 cm^2 and, as to the homogeneity or distribution of the activity of the radionuclide on the source, there should not be a variation above 10% in the specific surface activity measured at different parts of the source.

[0004] The preparation of radioactive sources of large superficial area cannot be done with the electroplatting methods used in the preparation of small sources (less than 5 cm^2) due to the problems that appear on the homogeneity of the electrical fields applied which give rise to an irregular distribution of the activity of the radionuclide on the surface, which causes a series of dark spots that deteriorate the appearance of the mentioned source.

[0005] An alternative to the electroplatting methods are the methods of direct platting of the solution of the radionuclide onto the support of the source as described in Lally et al. Nucl. Instr. Meth. 223 (1984)

[0006] 259. Among these methods, the simplest one is the one consisting in the evaporation of the solution deposited on the support. The homogeneity of the activity on the surface of the source is quite acceptable, but has the drawback that the deposit is unprotected and may readily come off.

[0007] The procedure of the invention is based on the direct platting of the radionuclide solution onto an anodised aluminium support. The radionuclide, being absorbed onto the aluminium oxide porous layer, is platted in a homogeneous way and protected.

[0008] Conventional procedures for the incorporation of stable elements (Co, Mn, Cr, etc.) onto an anodised aluminium layer are well known, and are based on the use of aqueous solutions of the elements to be incorporated under diverse conditions of temperature, pH, concentration, etc.

[0009] The application of the above mentioned procedures to the preparation of extensive radioactive sources has not produced good results, obtaining very low yields (around 2%) for the deposit of the radionuclides. Besides, as a consequence of other stable elements being incorporated to the deposit, causes an increase in the thickness of the latter which degrades the radiation energy, making more difficult the calibration of the source.

[0010] The possible causes that may produce these poor results, are thought to be:

a) The thickness of the anodised aluminium layer of the supports of the extensive radioactive sources must be at most 4 μm, so that there is no important degradation in the radiation energy, much thinner, therefore, than the minimum 10 μm used in conventional anodising procedures.

b) Conventional procedures use stable element solutions, whose concentrations are in the 10^{-1} M range, much above those of the radionuclide which are 10^{-10} M. The concentrations of the radionuclide cannot be in the same range as those of the stable elements due to the problems that working with very high radioactive activities would originate.

[0011] The process for the incorporation of the radionuclide to the aluminium oxide layer is ruled by the superficial liquid-solid interaction. It is this interaction that controls the solution volume absorbed and, therefore, the amount of radionuclide that deposits onto the porous aluminium oxide layer.

[0012] The procedure subject matter of the invention manages to increase this volume by modifying the surface tension between the liquid-solid phases; i.e., by varying the composition of the radionuclide solution it is possible to obtain a better absorption onto the anodised porous layer.

[0013] In order not to hinder the calibration of the source, it is necessary to limit to the maximum the contamination of the anodised layer, and thus it is essential to operate with a low solid residue rate.

[0014] On the other hand, a high steam pressure will allow to operate at room temperature which, besides providing easiness and economy, makes possible the platting of substances which would not endure high temperatures.

[0015] A series of tests were carried out with solutions of a radionuclide with different composition and surface tension values, and it was found that when the surface tension value was 72 din cm^{-1} (versus air and at 20°C) very low yields were obtained (around 2%), whilst when the surface tension value was around 20 din cm^{-1} (versus air and at 20°C) the yield was excellent (around 80%) .

[0016] The procedure for the incorporation of hazardous or costly substances onto an anodised aluminium support subject matter of the invention consists in the use of solutions with small surface tension values (around 20 din cm^{-1} versus air and at 20°C) in solutions which must present determined characteristics.

[0017] Components

- The substance to be incorporated; which must be

soluble in the solvent which accompanies it.
- A solvent which has a low residue rate.
- Various additives; in order to obtain the adequate surface tension and steam pressure values, in such a way that it is not necessary to use high temperatures.

[0018] Parameters

- Surface tension; less than 25 din cm^{-1} versus air and at 20°C
- Steam pressure; greater than 40 mm of Hg at 20°C
- Solid residue; less than 4.5 10^{-3} g/l

[0019] In order to help to a better understanding of the invention, a detailed description of a preferred embodiment will follow, with an orienting and not limiting character, supported by the figure attached, being an integral part of this descriptive report and which represents the incorporation yield of a 10^{-10} M solution of ^{241}Am onto an aluminium plate with an anodised thickness of 4 μm as a function of surface tension of the water-ethanol mixture used in the solution.

[0020] The reckoning of the deposit yield of the 10^{-10} M solution of ^{241}Am was done in the following way: Preparation of a sample with a solution volume which is a function of the plate surface, as will be stated below. Measurement of the radioactive activity.

[0021] Preparation of a solution volume identical to the preceding one and deposition of the same onto the anodised aluminium plate in the conditions that will be described below. Measurement of the radioactive activity of the anodised aluminium plate impregnated in this manner.

$$R = \frac{\text{Activity of the anodised aluminium plate}}{\text{Activity of the solution sample}} \times 100$$

[0022] The procedure for the preparation of extensive radioactive sources comprises the following stages:

A solvent - additive mixture is prepared that complies to the following conditions: surface tension below 25 din cm^{-1}, steam pressure greater than 40 mm of Hg at 20°C and solid residue below 4.5 10^{-3} g/l which in this case will be made up of a water ethanol mixture with a considerable larger rate of this latter product in a proportion at least of approximately 20/80.

The 10^{-10} M solution of the radionuclide is prepared with the previously prepared mixture from a solution of ^{241}Am in 4M nitric acid, by drying and change of medium.

The solution thus prepared is deposited by successive drops onto an aluminium support with a thickness of anodising of 4 μm. A volume of 1 ml is sufficient for one plate of 100 cm^2.

The excess is removed after one minute, by means of blotting paper.

The deposit which may have remained on the anodised layer is removed by rubbing with paper soaked in the solvent - additive mixture used for the dissolution of the radionuclide.

[0023] Once the example of a preferred embodiment of the invention has been described, the generalisation of the procedure for the incorporation of other substances onto anodised plates will be evident for any person skilled in the art.

[0024] The solvent shall be chosen as a function of the substance that is to be incorporated. Given that the solution thus prepared will not comply, in general, to the limits established for the surface tension, steam pressure and solid residue parameters which ensure the success of the procedure, it will be necessary to include surfactant additives which must also be compatible, in terms of solubility, with the solvent used.

[0025] The last removal stage, by rubbing, of the deposit which could have remained on the anodised layer, is essential in the application of the procedure to the preparation of extensive radioactive sources, in order to ensure the homogeneity of the radioactive activity and its endurance against mechanical aggressions, but may not be essential in other applications.

[0026] As regards the industrial performance of the procedure subject matter of the invention, it will be evident for any person skilled in the art that the removal of the excess solution by means of blotting paper is an operation whose purpose is the same as the high steam pressure requirement. Thus, for the performance of the method by hand, it will be possible to operate with a lower steam pressure as a function of the ability of the operator with the blotting paper, whilst in a mechanised production installation, it will be mandatory to respect the steam pressure values specified and simplify or even leave out the stage of removal of the excess by means of blotting paper.

Claims

1. Procedure for the incorporation of hazardous or costly substances onto aluminium supports with very low thickness of anodising, characterised in that it comprises the following stages:

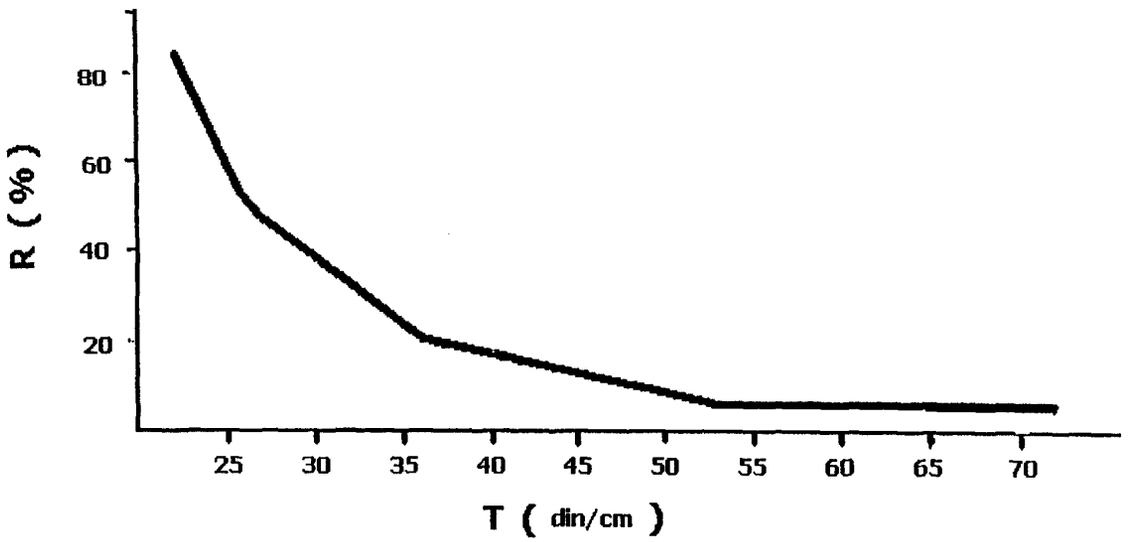
   preparation of a solvent-additive mixture which complies to the conditions: surface tension below 25 din cm^{-1}, steam pressure greater than 40 mm of Hg at 20°C and solid residue below 4.5 10^{-3} g/l,

   preparation of a 10^{-10} M solution of the substance to be incorporated with the mentioned solvent - additive mixture,

   deposition of the solution thus prepared by successive drops onto an aluminium support with

an thickness of anodising of 4 μm,
removal of the excess, after one minute, by means of blotting paper,
removal of the deposit which may have remained on the anodised layer by rubbing with paper soaked in the solvent - additive mixture mentioned before.

2. Procedure for the incorporation of hazardous or costly substances onto aluminium supports with very low thickness of anodising, according to claim 1, characterised in that the substance to be incorporated mentioned is a radionuclide, preferably ^{241}Am, and the solvent - additive mixture is made up of water and ethanol.


R (%)
80
60
40
20
25
30
35
40
45
50
55
60
65
70
T (din/cm)

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 99 50 0116

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
A	GARCIA-TORANO E ET AL: "Preparation and standardization of large-area sources for calibration of alpha-particle contamination monitors" NUCLEAR INSTRUMENTS & METHODS IN PHYSICS RESEARCH, SECTION - A: ACCELERATORS, SPECTROMETERS, DETECTORS AND ASSOCIATED EQUIPMENT, vol. 396, no. 1-2, 1 September 1997 (1997-09-01), page 130-134 XP004097889 ISSN: 0168-9002 * page 131, last paragraph - page 132, right-hand column, paragraph 1 * ---	1,2	G21G4/06
D,A	LALLY ET AL. : "Source preparation in alpha spectrometry" NUCLEAR INSTRUMENTS AND METHODS., vol. 223, 1984, pages 259-265, XP002121026 NORTH-HOLLAND PUBLISHING COMPANY. AMSTERDAM., NL * page 261, last paragraph - page 263, paragraph 1 * ---	1	TECHNICAL FIELDS SEARCHED (Int.Cl.7) G21G C23C
A	FUSCHINI ET AL.: "A new method for preparing radioactive sources" NUCLEAR INSTRUMENTS AND METHODS., vol. 26, no. 2, March 1964 (1964-03), pages 301-304, XP002121134 NORTH-HOLLAND PUBLISHING COMPANY. AMSTERDAM., NL * page 301, paragraph 1 - page 302, last paragraph * ---	1	
	-/--		

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	15 November 1999	Capostagno, E

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P04C01)

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 99 50 0116

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.7)
A	PATENT ABSTRACTS OF JAPAN vol. 013, no. 189 (M-821), 8 May 1989 (1989-05-08) & JP 01 014090 A (FUJI PHOTO FILM CO LTD), 18 January 1989 (1989-01-18) * abstract *	1	
A	EP 0 015 807 A (COMMISSARIAT ENERGIE ATOMIQUE) 17 September 1980 (1980-09-17)		

TECHNICAL FIELDS SEARCHED (Int.Cl.7)

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	15 November 1999	Capostagno, E

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P04C01)

ANNEX TO THE EUROPEAN SEARCH REPORT ON EUROPEAN PATENT APPLICATION NO. EP 99 50 0116

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on
The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

15-11-1999

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
JP 01014090 A	18-01-1989	NONE	
EP 0015807 A	17-09-1980	FR 2450498 A	26-09-1980
		JP 55119100 A	12-09-1980

EPO FORM P0459

For more details about this annex : see Official Journal of the European Patent Office, No. 12/82