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(54) **Title**: NUCLEAR FUEL ELEMENT AND SOL-GEL PROCESS FOR ITS PRODUCTION

(57) **Abstract**: The present invention relates to a new kind of nuclear fuel elements in which one or more radioactive isotopes are comprised in a quartz matrix, either in the form of a solid solution or in the form of a dispersion of powders of compounds of said radioactive isotopes in said matrix. The invention also relates to sol-gel processes for the production of the above nuclear fuel elements.

WO 2011/036210 A1

NUCLEAR FUEL ELEMENT AND SOL-GEL PROCESS FOR ITS PRODUCTION

Field of the invention

The present invention relates to nuclear a fuel element and to a sol-gel process for producing it.

Prior art

Nuclear fuel elements are the active elements of plants for the production of energy that exploit nuclear reactions. In the present text, by nuclear reaction is meant nuclear fission (that is, the present invention does not regard nuclear fusion reactions). The chemical elements that can give rise to nuclear reactions are generally referred to as radioactive elements and are the natural or synthetic heavy ones. These elements have generally a remarkable number of isotopes, which may not all be radioactive, or not all in the same way; for instance, radioactive elements may have fissile and fertile isotopes. Fissile isotopes are those that can undergo fission upon capture of neutrons of suitable energy; examples are uranium-233 (^{233}U), uranium-235 (^{235}U) or plutonium-239 (^{239}Pu). Fertile isotopes are those that cannot undergo fission themselves, but that upon neutron capture are transformed into fissile isotopes; examples are thorium-232 (^{232}Th), uranium-234 (^{234}U) and uranium-238 (^{238}U), which transform respectively into ^{233}U, ^{235}U and ^{239}Pu. The definition “radioactive elements” will be used for the sake of brevity in some instances to mean chemical elements that have at least one fissile or fertile isotope. Besides, as both fissile and fertile isotopes may be used in nuclear plants, the definition “nuclear fuel elements” as used in the rest of the description is intended to cover both kinds of isotopes.

There are several forms of nuclear fuel elements, the most common of which is rods containing the active material. The rods are generally arranged in assemblies grouping a few tens of them, and in which these are separated by a moderator and a coolant; the moderator may be e.g. water, heavy water or graphite, and has the function of reducing the velocity of particles (normally neutrons) circulating in the system to values at which their capture by radioactive elements is high, so as to guarantee the propagation of the nuclear reactions; the coolant is a fluid for transporting the heat generated by the nuclear reaction to a part of the plant where it is transformed into power, and is commonly water but may also be water vapour, a molten metal (e.g., sodium) or a molten salt.

A very common form of nuclear fuel element is a zirconium alloy tube containing particles of a fuel compound that is generally an oxide, such as e.g. uranium dioxide, UO_2, plutonium dioxide, PuO_2, or, less commonly, thorium dioxide, ThO_2; some reactors (so-called MOX reactors) use mixtures of oxides.

The known nuclear fuel elements have some drawbacks.

First, the oxide pellets presently used may generate dusts due to the high temperatures reached during the nuclear reactions; these dusts may accumulate in the rods and create problems of spreading in the environment when the spent rods are removed from the plant for disposal.

Besides, the oxide pellets need encasement, generally in a metallic alloy cladding (typically made of a Zircaloy, a family of alloys containing more than 98% b.w. of zirconium); the cladding is however subject to wear with time, due to corrosion at the high operating temperatures and embrittlement as a consequence of absorption in the alloy structure of hydrogen atoms and consequent formation of fragile hydride phases. These phenomena may result in breaking of the cladding, with the possible release of nuclear isotopes in the coolant and consequent pollution with radioactive material of the whole plant and possibly of the surrounding areas.

Summary of the invention

It is thus an object of the present invention to provide a new kind of nuclear fuel elements which do not suffer from the above mentioned problems; another object of the invention is to provide a process for the production of said elements.

These and other objects are reached through the present invention that, in a first aspect thereof, relates to nuclear fuel elements.

According to the invention, the nuclear fuel elements are formed as a monolithic quartz body in which are comprised one or more fissile or fertile radioactive elements.

The quartz bodies forming the nuclear fuel elements of the invention, further to resolving the prior art problems of dust generation and need of encasing, thanks to the thermal conductivity of quartz also afford better thermal management of the nuclear fuel element.

Detailed description of the invention

As will be apparent from the following description, the nuclear fuel elements of the present invention are more versatile than traditional ones and, also thanks to the ease of production in tiny dimension (well below one millimeter), allow expansion of the range of possible use of nuclear energy to applications in which this was first excluded due to safety reasons.

A nuclear fuel element of the invention may be realized according to several different embodiments. In a first case, it may be in the form of a solid solution of one or more of said radioactive elements in a quartz matrix; alternatively, it may be in the form of a matrix essentially consisting of pure silica in which "islands" of a compound of the radioactive element are dispersed; finally, it may be in a mixed form, in which said islands of compound of the radioactive element are dispersed in a matrix of the first kind.

In the first embodiment, the atoms of the radioactive element are dispersed in the quartz matrix; in this condition, said atoms replace silicon atoms in the quartz structure, and linked through oxygen bridges to silicon atoms of the quartz matrix.

In the second embodiment, the nuclear fuel elements are formed by a quartz matrix embedding particles of compounds of one or more radioactive elements. Said particles have preferably dimensions comprised lower than about 5 μm, and even more preferably lower than about 1 μm; particles of higher dimensions would be too big and thus produce too much heat during the nuclear reactions, thus leading to localized overheating and possible breaking of the fuel element.

The third embodiment is a mixed situation between the ones above.

The chemical elements that may be used are all those having at least one fissile or fertile isotope. It is not necessary that the isotope employed in the preparation of the fuel element is fissile; it is sufficient that it is fertile, so that it can be made fissile upon suitable activation, generally by neutron absorption; the use of fertile isotopes may even be preferred, because their handling in the production of fuel elements is less problematic, and the storing and transporting of the so obtained elements are not subjected to the severe regulations valid for elements containing fertile isotopes. The amount of fissile or fertile atoms in the quartz matrix is lower than about 15% (atomic percentage); at higher percentages the quartz monolith loses its structural integrity so that the advantages of the nuclear fuel elements of the invention, linked to mechanical characteristics of said monolith, are lost. At too low percentages however the fissile or fertile atoms are too diluted for sustaining a nuclear reaction; for the scopes of the invention, preferred are monoliths in which the amount of radioactive elements varies between about 0.5% and 10% atomic.

The preferred nuclear fuel elements of the invention are those in which the radioactive element is one of uranium, plutonium, and particularly thorium; the reason is that this element is present in nature essentially only as its fertile isotope ^{232}Th, lending itself thus to the production of elements with the handling, storing and transporting advantages described above. Besides, thorium has another important advantage: the oxide ThO_2 has a melting point of 3390 °C and a boiling point of 4400 °C; so, when present in the form of particles in the quartz bodies, it offers the safety feature that it cannot evaporate in case of possible accidents in consequence of which the plant catches fire; if, under such circumstances, the silica of the fuel element melts, ThO_2 particles simply precipitate at the bottom of the melt thus self-confining in a small, localized volume, without spreading around.

The nuclear fuel elements of the invention may have any shape and dimensions, depending on the target use of the same.

For use in traditional nuclear power plants, the elements of the invention have preferably the usual shape and dimensions of known rods, and in particular may be shaped as cylindrical bodies of diameter up to 20 cm and length up to 100 cm; preferably, said elements have diameter in the range 8-10 cm and length below 50 cm. These bodies may anyway assume more complex shapes, for instance shapes in which the body has voids and cavities, in order to allow the flow of the coolant into the body or in order to accommodate a moderator.

For use in applications different from nuclear power plants, the nuclear fuel elements of the invention may be easily produced in very small dimensions, even down to the micrometers range, and with low concentrations of the radioactive elements in the quartz matrix. Thanks to the combination of these two features, the elements of the invention can be produced in forms that can fit into very small volumes and greatly reduce the risk of nuclear pollution, thus relaxing the safety requirements and opening new fields of possible application.

A first possible application is in miniaturized or even “micro” power plants, that may be adapted for use in micro-powering systems of transport means (ships, submarines, but also trucks or even cars), for “local” plants for heating buildings, blocks or small towns, or for the production of transportable systems for production of energy; depending on the specific use, the nuclear fuel elements of the invention may nave dimensions ranging between about of a few centimeters (or a few tens of centimeters) for heating small towns, down to less than one centimeter for cars.

Another possible application is in Stirling engines, in which the nuclear fuel elements of the invention can be used as heat source in the hot portion of the engine; in this case, the dimensions of the element of the invention vary depending on those of the engine, but may typically be comprised between about 1 and 15 cm.

Finally, another very interesting application possible with the elements of the invention is the localized treatment of site-specific diseases, in particular tumors, in the human (or animal) body; this can be done by mounting a tiny element of the invention as the tip of a probe or catheter and bringing said tip through blood vessels or cavities of the body close to the part to be treated, so as to irradiate with nuclear radiation only the part needing treatment. In a possible variant, the same effect can be obtained by producing the element of the invention as a coating of, or anyway associated to, a small part of a magnetic metal or alloy (e.g., iron), and guiding the element towards the part to be treated by means of magnets from outside the body.

In its second aspect, the invention relates to a sol-gel process for producing nuclear fuel elements.

The process of the invention may be carried out in two alternative ways.

In the first alternative, the process is adapted for the production of the first kind of nuclear fuel elements described above, and comprises:

- preparing an aqueous or hydroalcoholic dispersion containing silica, at least one alkoxysilane and a hydrolizable chemical compound of at least a radioactive element, said dispersion having a pH not higher than 4 and in which the molar ratio silica:alkoxysilane is between 6 and 50, preferably between 6.5 and 25, more preferably between 7 and 15;

- modifying the dispersion pH to obtain a sol;
- pouring the sol thus obtained into a mould having a shape corresponding to that of the desired nuclear fuel element;
- allowing the sol to gel to hence obtain a hydrogel;
- drying the hydrogel to obtain a dried gel;
- sintering the dried gel to obtain the vitreous silica article.

In the present description and in the claims which follow, except where otherwise indicated, the numerical ranges include all values comprised within the range, including the ends of the range.

The sol-gel process of the present invention is characterized, with respect to similar processes of the prior art, by a higher ratio of silica to alkoxysilane; applied to pure silica, this process is the object of co-pending International patent application no. PCT/EP2009/060155, still secret at the filing date of the present application.

The first step, preparing the dispersion, may be realized following several different orders of addition of the components. For instance, it is possible to prepare an acidic aqueous of hydroalcoholic solution of said hydrolizable compound of radioactive element(s) and add first silica to this solution and then the alkoxysilane(s) to the resulting dispersion; or, it is possible to prepare the silica dispersion first and to this add the said hydrolizable compound(s) and alkoxysilane(s) together or in any order.

The silica is preferably pyrogenic silica. Preferably, said alkoxysilane is a tetraalkoxysilane in which each alkoxy group has from 1 to 6, preferably from 1 to 4, carbon atoms. More preferably, the tetraalkoxysilane is chosen from: tetraethoxysilane (TEOS), tetramethoxysilane (TMOS), methoxytriethoxysilane (MTEOS). Preferred for the specific application is tetraethoxysilane (TEOS). The hydrolizable compound of the radioactive element may be, for instance, an organometallic compound, e.g., an alkoxide, of the element; or, it may be a soluble salt of the element, such as an halogenide (preferably the chloride) or a nitrate. The preferred compounds for this step are MCl_4 and $M(-OR)_4$, wherein M is a radioactive element, preferably U, Pu or more preferably Th, and R is an alkyl radical, preferably methyl or ethyl. The compound is added to the sol in the necessary stoichiometric ratio, taking into account that the metal M is completely taken up by the final quartz composition.

Whichever order of addition is chosen, when the silica dispersion is produced, its pH must be not higher than 4 and is preferably from 1.5 to 3.0, more preferably from 2.0 to 2.5; this may be achieved by adding an acid, in particular an inorganic acid, for example hydrochloric acid, phosphoric acid, sulphuric acid or the like, or an organic acid, for example acetic acid. The acidity may be conferred by the solution of said hydrolysable compound of radioactive element(s), as these solutions have generally pH lower than 2. The preparation is generally conducted by adding the silica to the aqueous or hydroalcoholic medium and dispersing it by mechanical stirring, for example with a mixer of Ultra-Turrax® type or similar devices.

According to a preferred embodiment, in said dispersion at acid pH, the molar ratio of water to alkoxysilane is between 40 and 200, preferably between 50 and 160, more preferably between 60 and 120. The Applicant has in fact surprisingly found that such a high molar ratio of water to alkoxysilane enables a different and better quality of the final product to be obtained, significantly reducing the formation of defects and/or breakages.

After completing the dispersion preparation and before modifying the pH, the dispersion temperature is preferably increased to a value between 30 and 50 ºC for a time variable between 6 and 18 hours. The dispersion pH is then modified, in particular raised to a value generally between 4.0 and 6.0, preferably between 4.7 and 5.2. This pH modification can be achieved, for example, by adding a water-soluble inorganic or organic basic compound, for example ammonium hydroxide or amines, in particular cyclohexylamine.

Alternatively, it is possible to use an aminoalkyl-alkoxysilane as the basic compound; examples of products suitable for the purpose are: 3-aminopropyl-trimethoxysilane, 3-aminopropyl-triethoxysilane, 2-aminoethyl-3-aminopropyl-dimethoxysilane, 2-aminoethyl-3-aminopropyl-diethoxysilane, [3-(2-aminoethyl)aminopropyl]-trimethoxysilane, [3-(2-aminoethyl)aminopropyl]-

triethoxysilane, or their mixtures. The use of an aminoalkyl-alkoxysilane enables sol gelling to be achieved without leaving in the final product ammonium salts or other by-products which can reduce the quality of the finished product.

The basic compound is preferably added under stirring so as to prevent excessive local pH increase which can cause premature gelling with consequent non-homogeneity in the final product.

Before gelling becomes apparent, the sol is poured into a mould and allowed to gel to hence obtain a hydrogel. The mould must have a shape corresponding to the final shape of the desired nuclear fuel element; as known, at the end of a sol-gel process, when the manufact is brought to complete densification by thermal treatments, its dimensions are about one half than the dimensions of the starting gel, so in defining the dimensions of the mould this shrinking must be taken into account. In this manner, at the end of the process, a self-sustaining nuclear fuel element is obtained having the necessary shape to be fit in the reactor, without the need for a cladding. Evidently the mould is generally filled such as to avoid the inclusion of air, and the consequent defects formation in the final product. The mould can be made of various materials, for example plastic materials, metals, glass fibre, carbon fibre, ceramic, etc. Preferably the mould comprises plastic materials such as: polypropylene, polyethylene, polystyrene, fluorinated polymers (such as polytetrafluoroethylene), silicones. Gelling is preferably but not exclusively carried out at room temperature.

When gelling has been achieved, the hydrogel is dried to remove as much as possible the water present inside the gelled structure. According to a preferred embodiment, this drying is conducted by introducing the hydrogel into a thermal and humidity cycle oven. In this manner both the temperature and the degree of humidity under which the drying takes place can be controlled to hence prevent formation of internal stresses within the vitreous material due to disuniformities in the drying process.

In general, the drying time can vary within wide limits, depending on the various process parameters involved, in particular the dimensions and/or transfer area of the nuclear fuel element to be produced. In general, the drying time can vary from a minimum of a few hours to 1200 hours, preferably between 150 and

900 hours, depending on the final dimensions of the nuclear fuel element; for tiny elements, drying times of about one day may be enough. The temperature at which the drying step is conducted is preferably varied during the process, in general between 15 °C and 120 °C, preferably between 20 °C and 100 °C. The degree of humidity is also preferably varied during the drying process, in general between 30% and 100%. Preferably, the degree of humidity is maintained, for a time at least equal to 50% of the overall drying time, at values on an average greater than 70%, to then be gradually reduced to average values between 60% and 30%.

With regard to the process of sintering the dried gel, this can be carried out by overall known methods. In general, this comprises a step of calcining in an oxidizing atmosphere (generally oxygen, air or synthetic oxygen-nitrogen blends), followed by consolidation in an inert atmosphere; on the other hand, it is absolutely to be avoided the dehydration and purification in the presence of at least one chlorinated product that is conducted in most sol-gel processes directed to the production of pure silica, because this step removes from the dried gel essentially any metal that is not silica, while for the object of the present invention it is necessary to retain the radioactive compounds in the final sintered quartz body. The calcining step is generally but not exclusively carried out in a kiln at a temperature between 30 °C and 60 °C, with repeated steps of drawing off and feeding oxygen to eliminate the combustion products formed. Consolidation, leading to material densification, is generally carried out in a helium atmosphere, possibly with traces of oxygen, at a temperature between 800 °C and 1400 °C.

In its second alternative, the process is adapted for the production of the second kind of nuclear fuel elements described above, namely those in which the radioactive elements are present particles embedded in a quartz matrix. This process comprises:

- preparing an aqueous or hydroalcoholic dispersion of silica having a pH not higher than 4;
- adding to said dispersion at least one alkoxysilane in a quantity such as to obtain a silica:alkoxysilane molar ratio between 6 and 50, preferably between 6.5 and 25, more preferably between 7 and 15;

- modifying the dispersion pH to obtain a sol while adding particles of at least one oxide of a radioactive element said particles having dimensions below about 1 µm;
- pouring the sol thus obtained into a mould;
- allowing the sol to gel to hence obtain a hydrogel;
- drying the hydrogel to obtain a dried gel;
- sintering the dried gel to obtain the vitreous silica article.

This alternative process is in many steps and adopted conditions similar to the one of the first alternative.

The preparation of the dispersion of silica in water or hydroalcoholic solution is carried out similarly to how described before for the first alternative of the process; the pH must not be higher than 4, preferably comprised between 1.5 and 3.0 and more preferably from 2.0 to 2.5, by addition of the acid cited before. The silica is preferably pyrogenic silica. The preparation is generally conducted by adding the silica to the aqueous medium and dispersing it by mechanical stirring, for example with a mixer of Ultra-Turrax® type or similar devices.

Similarly, the alkoxysilanes that may be employed are the same as described before; and preferably, the molar ratio of water to alkoxysilane in the dispersion is between 40 and 200, preferably between 50 and 160, more preferably between 60 and 120.

After adding the alkoxysilane, the dispersion temperature is preferably increased to a value between 30 and 50 °C for a time variable between 6 and 18 hours. The dispersion pH is then modified, in particular raised to a value generally between 4.0 and 6.0, preferably between 4.7 and 5.2. This pH modification is achieved with the same basic compounds mentioned before, or with the cited aminoalkyl-alkoxysilanes. The basic compound is preferably added under stirring to avoid non-homogeneity in the final product s discussed above.

The compound of the radioactive element, in the form of powders, is added the dispersion not earlier than the addition of the basic compound. The reason is that these compounds have generally density values of about 3, higher than the sol or gel systems they are added to, so, if added to a sol of relatively low density and viscosity, the powders could give rise to separation through precipitation towards the bottom of the dispersion, eventually resulting in a strongly non-homogeneous nuclear fuel element. The addition of said powders during the addition of the basic compounds, and thus during the gelation step, has the advantage that the medium to which they are added is much more viscous and thus capable of suspending powders. In order to improve homogeneity, such addition takes place with stirring, preferably with a Ultra-Turrax® mixer. Further, still in order to favour homogeneity of the system, the powders have a grain size lower than about 1 µm.

The steps of adding the basic system and adding said powders preferably take place with the dispersion already in the final mould, having a shape corresponding to the final nuclear fuel element, so as to guarantee homogeneity of the system up to gelation; said additions can also take place in a different container and the sol under gelling can then be poured in the final mould.

The next steps of the process, drying of the wet gel, calcining in an oxidizing atmosphere and consolidation in an inert atmosphere, take place in the same manner as described above with reference to the first alternative process of the invention.

In the particular case of elements of the invention as coatings of magnetic metal or alloy parts (or anyway associated to these parts) for use in therapeutic treatments as cited above, the joining of the element with the metal or alloy may be easily achieved by placing a droplet of the starting sol onto the surface of said metal or alloy part (covering it totally or partially), or by dipping the metal or alloy part into a bath of the sol (in this case obtaining complete coating of the part). After gelation, care must be paid to the fact that magnetism is lost when the metal or alloy is subjected to temperatures higher than its Curie point; to overcome the problem, it is possible not to fully consolidate the silica matrix, by treating it only a temperatures in the range of 400 to 500 °C, generally in an oxidizing atmosphere (to burn off residues of organic compounds remaining from the process), possibly using magnetic compounds endowed with high Curie point values, such as Alnico magnets, having Curie point of about 850 °C (Alnico magnets typically comprise, by weight, 8–12% aluminum, 15–26% nickel, 5–24% cobalt, up to 6% copper, up to 1% titanium, the balance being iron); or preferably, the metal or alloy part is magnetized after completion of the production of the element of the invention, comprising its full consolidation, by exposure to an external magnetic field.

CLAIMS

1. Nuclear fuel element formed as a monolithic quartz body in which are comprised one or more fissile or fertile radioactive elements.
2. Nuclear fuel element according to claim 1 in the form of a solid solution of said radioactive elements in a quartz matrix.
3. Nuclear fuel element according to claim 1 in the form of a body comprising particles of at least one oxide of said radioactive elements in a matrix essentially consisting of pure silica.
4. Sol-gel process for the production of a nuclear fuel element according to claim 2, comprising:
   - preparing an aqueous or hydroalcoholic dispersion containing silica, at least one alkoxysilane and a hydrolizable chemical compound of at least a radioactive element, said dispersion having a pH not higher than 4 and in which the molar ratio silica:alkoxysilane is between 6 and 50, preferably between 6.5 and 25, more preferably between 7 and 15;
   - modifying the dispersion pH to obtain a sol;
   - pouring the sol thus obtained into a mould having a shape corresponding to that of the desired nuclear fuel element;
   - allowing the sol to gel to hence obtain a hydrogel;
   - drying the hydrogel to obtain a dried gel;
   - sintering the dried gel to obtain the vitreous silica article.
5. Sol-gel process for the production of a nuclear fuel element according to claim 3, comprising:
   - preparing an aqueous or hydroalcoholic dispersion of silica having a pH not higher than 4;
   - adding to said dispersion at least one alkoxysilane in a quantity such as to obtain a silica:alkoxysilane molar ratio between 6 and 50, preferably between 6.5 and 25, more preferably between 7 and 15;
   - modifying the dispersion pH to obtain a sol while adding particles of at least one oxide of a radioactive element, said particles having dimensions below about 1 μm;
   - pouring the sol thus obtained into a mould;

- allowing the sol to gel to hence obtain a hydrogel;
- drying the hydrogel to obtain a dried gel;
- sintering the dried gel to obtain the vitreous silica article.

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2010/064054

A. CLASSIFICATION OF SUBJECT MATTER
INV. G21C3/62 G21C3/64
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
G21C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 6 190 582 B1 (MARLOWE MICKEY O [US]) 20 February 2001 (2001-02-20)	1-3
A	column 4, line 23 - column 6, line 54; claims 1-17 -----	4,5
X	FR 1 280 619 A (SOCONY MOBIL OIL CO) 8 January 1962 (1962-01-08)	1-3
A	page 2, paragraph 3 - page 29, paragraph 3; claims 1-45 -----	4,5
A	US 3 088 892 A (CAIN JR FRANCIS M ET AL) 7 May 1963 (1963-05-07) column 3, line 31 - column 3, line 40 -----	1-5
A	GB 942 852 A (BABCOCK & WILCOX LTD) 27 November 1963 (1963-11-27) the whole document ----- -/--	1-5

☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

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Date of the actual completion of the international search 7 March 2011	Date of mailing of the international search report 16/03/2011
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Lohberger, Severin

INTERNATIONAL SEARCH REPORT

International application No
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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 852 692 A (BELGE PRODUITS CHIMIQUES SA) 26 October 1960 (1960-10-26) examples 1,3 -----	1-5
A	US 3 325 419 A (HURLEY FORREST R ET AL) 13 June 1967 (1967-06-13) example 2 -----	1-5

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
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