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- (71) Applicant: SOLVAY SA [BE/BE]; Rue de Ransbeek,  
310, B-1120 Bruxelles (BE).
- (72) Inventor: KIM, Hag-Soo; 706, 266-1, Daean-Ri, Onyang-  
Eup Ulju-Kun, Ulsan 689-903 (KR).
- (74) Agents: MROSS, Stefan et al.; Rue de Ransbeek, 310, B-  
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(54) Title: METHOD FOR PURIFYING FLUORINATED ORGANIC CARBONATES

(57) Abstract: Provided are methods for purifying fluorinated organic carbonates, especially fluoroethylene carbonates produced from the reaction of ethylene carbonate and elemental fluorine. The methods according to the present invention comprise a step of treating the fluorinated organic carbonated with an organosilicon compound having at least one -Si-N- bond, and a step of distillation of the resulting mixture in at least one distillation column.

Method for purifying fluorinated organic carbonates**TECHNICAL FIELD**

This application claims priority to European application No. 12187802.9 filed on 09 October 2012, the whole content of this application being incorporated herein by reference for all purposes.

5 The present invention relates to methods for purifying fluorinated organic carbonates, especially fluorinated linear carbonates or fluorinated cyclic carbonates, during their production process.

**BACKGROUND OF THE INVENTION**

10 Fluorinated linear and cyclic carbonates, e.g. monofluoroethylene carbonate, fluoromethyl methyl carbonate, difluoroethylene carbonate and difluorinated dimethyl carbonate, but also the respective tri- and tetrafluorinated carbonates, are especially suitable as solvents or solvent additives for lithium ion batteries.

15 Generally, fluorinated organic carbonates can be prepared by the reaction of aliphatic linear or cyclic carbonates which are not substituted by F, or which have at least one substitutable H atom with elemental fluorine.

WO 2011/036283 describes a process for the manufacture of difluoroethylene carbonate, trifluoroethylene carbonate and/or tetrafluoroethylene carbonate or a mixture of two or more thereof by reaction of 20 an ethylene carbonate which is nonfluorinated or has a lower degree of fluorination, in the liquid phase with elemental fluorine (F<sub>2</sub>) to form tetrafluoroethylene carbonate.

WO 2009/118369 describes that mixtures with depleted hydrogen fluoride content are prepared from a mixture comprising organic carbonate, preferably 25 fluorinated organic carbonate, and hydrogen fluoride by stripping HF from the mixture by passing inert gas through the mixture. Noble gases or their mixtures with nitrogen or carbon dioxide or its mixtures with nitrogen are also suitable as inert gas for stripping; air also might be suitable, but it is not preferred. Nitrogen is especially suitable as stripping gas.

30 WO 2009/118368 provides a process for the preparation of fluorosubstituted organic carbonate which is depleted in HF wherein an HF-contaminated fluorinated organic carbonate comprising at least one CH-CF

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group in the molecule, is contacted with an inorganic reactant comprising SiO<sub>2</sub>, forming a mixture of a solid and HF-depleted fluorinated organic carbonate, and the resulting HF-depleted fluorosubstituted organic carbonate is separated from the solid. It is preferred to use solids with high surface area, especially  
5 amorphous solid silica or silica containing compounds. Silica gel is very preferred. The high surface of such gels (which can be applied in the form of shaped bodies, e.g. in the form of beads) provides for a fast HF-removing reaction.

WO 2011/020830 provides a process for the purification of a reaction  
10 mixture comprising fluoroethylene carbonate, ethylene carbonate, higher fluorinated ethylene carbonate or carbonates and hydrogen fluoride and optionally trace impurities (for example, trifluoroethylene carbonate) which is distilled in at least two distillation steps wherein the reaction mixture which is fed to the first distillation step contains not more than 5 % by weight of HF.  
15 Preferably, the reaction mixture which is fed to the first distillation column contains no more than 1 % by weight of HF. The purified fluoroethylene carbonate obtained is so pure, especially in view of the HF content, that no recrystallization is needed.

#### **DESCRIPTION OF THE INVENTION**

20 It is a purpose of the present invention to provide a method for further purifying fluorinated organic carbonates to obtain substantially decreased amount of impurities, such as hydrogen fluoride, during their production process. This purpose and other purposes which are apparent from the description and the claims are achieved by the methods of the present invention.

25 The present invention therefore provides a method for purifying fluorinated organic carbonate comprising a step of treating the fluorinated organic carbonate with an organosilicon compound having at least one -Si-N- bond, and a step of distillation of the resulting mixture in at least one distillation column.

30 Indeed, it has been surprisingly found that the certain chemical compounds, i.e. organosilicon compounds having at least one -Si-N- bond, can function as an effective scavenger for the impurities, such as hydrogen fluoride, water, etc. during the production process of fluorinated organic carbonate.

In the present invention, the impurities during the production process of  
35 fluorinated organic carbonates are understood to include not only hydrogen

fluoride and water, but also any other by-products and/or 2<sup>nd</sup> reaction products which can be harm for the final fluorinated organic carbonate products.

In the present invention, the singular form is intended to include the plural form; and the plural form is intended to include the singular form. Thus, for  
5 instance, the term “fluorinated organic carbonate” is not limited to a single carbonate compound, but includes a composition comprising two or more fluorinated organic carbonates including isomeric forms.

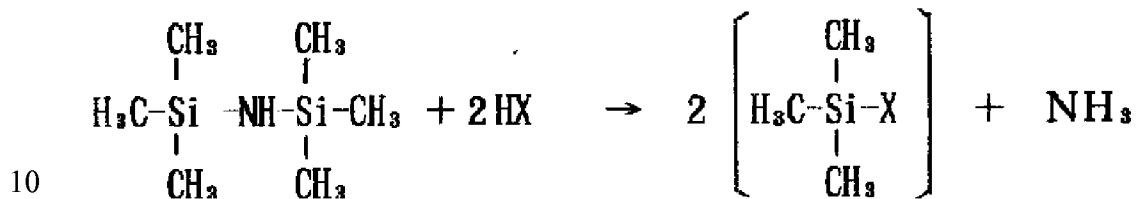
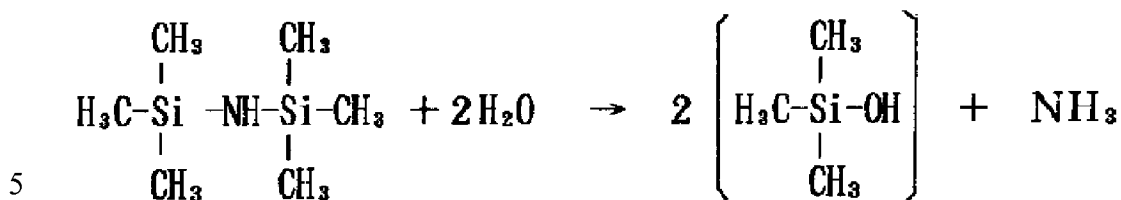
The organosilicon compounds having at least one -Si-N- bond according to the present invention can react effectively with water, hydrogen halide, such as  
10 hydrogen fluoride, and/or other impurities, rapidly forming the reaction product which can be removed or separated from the target product in at least one subsequent step.

In the present invention, the organosilicon compounds having at least one -  
Si-N- bond is understood to denote in particular the compounds capable of  
15 reacting at least hydrogen fluoride and/or water by the cleavage of Si-N bond to decompose the target impurities. The resulting decomposition product can be removed or separated from the targeted final product in one or more subsequent process. The organosilicon compounds can possess one or more Si-N bond in the form of, e.g., Si-N-Si bond, in addition to single or plural Si-N bonds per  
20 molecule.

In one embodiment according to the present invention, the organosilicon compound having at least one -Si-N- bond is preferably selected from the group consisting of organosilazane compounds, organodisilazane compounds and organotrisilazane compounds. More preferably, the organosilicon compound  
25 having at least one -Si-N- bond is selected from the group consisting of (N,N-diethylamino)trimethylsilane, N,O-bis(trimethylsilyl)acetamide, N,N'-bis(trimethylsilyl)-1,4-butanediamine, 1,1,1,3,3,3-hexamethyldisilazane, 1,1,3,3,5,5-hexamethylcyclotrisilazane, and any combination thereof. 1,1,1,3,3,3-hexamethyldisilazane is especially preferred, at least as this  
30 compound possesses two -Si-N- bonds, each can be a reacting spot with the target impurities, such as hydrogen halide and water. These compounds can be used singly or in combination of two or more compounds. These compounds are described for example in Japanese patent no. 3,348,344 B2. The expected reaction schemes are as follows :

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The amount of the organosilicon compound to be added during the production process of the fluorinated organic carbonate can be determined with relation with the approximate amount of impurity or impurities to be removed.

Alternatively, the end point for the addition of the organosilicon compounds can be determined by physical or chemical methodologies, for example, by gas chromatography, NIR or MIR. Generally, the addition of the organosilicon compound such that its amount relative to the crude product to be treated is 0.01 to 5 % by weight. Preferably, the amount of the organosilicon compound to be added can be determined by the analysis of remaining impurities, such as HF and/or H<sub>2</sub>O, and the amount can be 0.5~2.5 equivalent mol of the analyzed amount of remaining impurities, preferably 0.8~2 equivalent mol, more preferably 1.0~1.5 equivalent mol, most preferably around 1.2 equivalent mol, but the present invention shall not be limited thereto.

The organosilicon compound can be added in any one or more step before the separation step to collect the fluorinated organic carbonate as a final product. In a specific embodiment of the present invention, the organosilicon compound can be added to the crude reaction mixture, and/or to the pre-purified product. The compound can be contacted with the crude reaction mixture or the pre-purified product in a batch reactor. Preferably, it is added directly to the reactor where the fluorination of organic carbonate occurs.

The method of the present invention is especially suitable for the purification of fluorinated organic carbonates produced by the reaction of aliphatic linear or branched organic carbonates as starting compounds with elemental fluorine. The fluorination reaction can be performed batch wise or continuously, resulting a crude reaction mixture. Such processes are, for

example, described in WO 2011/036281 which describes a continuously performed fluorination process, or in US patent application publication 2006-0036102. In one alternative, the aliphatic linear or branched organic carbonates used as starting material may not be substituted by F atoms, and after the reaction with elemental fluorine, the respective reaction product comprises fluorinated organic carbonates which are substituted by at least 1 F atom up to being perfluorinated. In another alternative, the aliphatic linear or branched organic carbonates used as starting material are substituted by at least 1 F atom and comprise at least 1 H atom, and after reaction with elemental fluorine, the respective reaction product comprises fluorinated organic carbonates which are substituted by at least 2 F atoms up to being perfluorinated.

Now, fluorosubstituted organic carbonates which can be purified according to the present method are described.

According to one alternative, fluorosubstituted aliphatic linear or branched organic carbonates can be purified according to the method of the invention. Especially, fluorosubstituted organic carbonates of formula (I),  $(R^1O)(R^2O)C(O)$  can be purified. In formula (I),  $R^1$  and  $R^2$  may be the same or different.  $R^1$  and  $R^2$  are linear alkyl or branched alkyl with the proviso that at least one of  $R^1$  and  $R^2$  is substituted by at least 1 F atom. The term "linear alkyl" preferably denotes a C1 to C5 alkyl group or a C1 to C5 alkyl group substituted by at least 1 F atom. The term "branched alkyl" preferably denotes a C3 to C5 alkyl group or a C3 to C5 alkyl group substituted by at least 1 F atom. A condition is that At least one of  $R^1$  and  $R^2$ , as mentioned above, must be substituted by at least 1 F atom. This condition will not be repeated below.

Preferably,  $R^1$  is methyl, fluoromethyl, difluoromethyl, trifluoromethyl, ethyl, fluoroethyl, difluoroethyl, trifluoroethyl, tetrafluoroethyl or pentafluoroethyl, n-propyl, isopropyl, n-propyl, substituted by 1 to 7 F atoms, or i-propyl substituted by 1 to 7 F atoms. Preferably,  $R^2$  is methyl, fluoromethyl, difluoromethyl, trifluoromethyl, ethyl, fluoroethyl, difluoroethyl, trifluoroethyl, tetrafluoroethyl or pentafluoroethyl, n-propyl, isopropyl, n-propyl, substituted by 1 to 7 F atoms, or i-propyl substituted by 1 to 7 F atoms.

According to another alternative, fluorosubstituted aliphatic cyclic organic carbonates of formula (II),  $(OR^3O)C(O)$  are purified according to the method of the present invention.  $R^3$  is preferably an aliphatic alkylene group having 2 to 10 C atoms and is substituted by at least 1 F atom. More preferably,  $R^3$  is a C2 to C8 group substituted by at least 1 F atom. Especially preferably,  $R^3$  is a C2

group substituted by 1, 2, 3 or 4 F atoms; a linear or branched C3 group substituted by at least 1 F; a methylpropylene group substituted by at least 1 F atom; a dimethyl ethylene group substituted by at least 1 F atom; an ethyl ethylene group substituted by at least 1 F atom; a diethyl ethylene group substituted by at least 1 F atom; or a methyl ethyl ethylene group substituted by at least 1 F atom. Preferably, in this alternative embodiment, R<sup>3</sup> is monofluoroethylene, difluoroethylene, trifluoroethylene, tetrafluoroethylene, monofluoromethyl ethylene, difluoromethyl ethylene, methyl monofluoroethylene, methyl difluoroethylene, monofluoromethyl monofluoroethylene, monofluoromethyl difluoroethylene, difluoromethyl monofluoroethylene, difluoromethyl difluoroethylene, trifluoromethyl difluoroethylene, difluoromethyl trifluoroethylene, or trifluoromethyl trifluoroethylene. In this embodiment, R<sup>3</sup> is preferably monofluoroethylene, difluoroethylene, trifluoroethylene, tetrafluoroethylene, and most preferably, monofluoroethylene or difluoroethylene. "Difluoroethylene" may be a CF<sub>2</sub>-CH<sub>2</sub> group or a CFH-CFH group in cis or trans configuration.

The fluorinated organic carbonate can be provided as described, for example, in JP-A 2000-309583, US 2006-0036102, US-A 7,268,238 or WO 2011036281.

In a specific embodiment of the present invention, the fluorinated organic carbonate is selected from the group consisting of monofluoroethylene carbonate, fluoromethyl methyl carbonate, difluoroethylene carbonate, and bis-(fluoromethyl) carbonate. Preferably, the fluorinated organic carbonate is monofluoroethylene carbonate. Alternatively, the fluorinated organic carbonate is selected from the group consisting of ethyl-1-fluoroethyl carbonate, methyl-1-fluoroethyl carbonate, propyl-1-fluoroethyl carbonate, and allyl-1-fluoroethyl carbonate.

The method of the invention can be applied to purify a fluorinated product which was not yet subjected to a purifying treatment. Alternatively, it can be applied to purify a pre-purified fluorination product which had already been subjected to a pre-purification treatment to provide pre-purified fluorinated carbonate but which is considered as being not pure enough to be applied as solvent or additive for lithium ion batteries; or it can be applied to a purified product which while it is considered as being pure enough for use as a solvent or an additive for lithium ion batteries, still forms some HF when stored or contacted with moisture.

Accordingly, in another embodiment of the present invention, the fluorinated organic carbonate to be treated with the organosilicon compound is a crude reaction mixture obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution by fluorine, or a pre-purified product obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution by fluorine and at least one step of removing hydrogen fluoride. Preferably, the fluorinated organic carbonate to be treated is a crude reaction mixture obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution and at least one step of removing hydrogen fluoride. Particularly, the addition of the organosilicon compound can be conducted to the reactor where such reaction including at least one step of reacting elemental fluorine with an organic carbonate occurs.

The post-removal of HF from the pre-purified reaction mixture containing the fluorinated organic carbonate obtained in a fluorination process with elemental fluorine and the following or simultaneous treatment with the organosilicon compound is preferred. It is expected to the expert that in the fluorination step, for each consumed mol of  $F_2$ , one mol of HF is produced. Several processes are suitable for the post purification to remove HF. The preferred methods comprise at least one step of stripping with at least one gas, at least one step of distillation, or steps of both. A process for stripping HF from a raw product is described in WO 2009/118369. An inert gas, e.g.  $N_2$ , is passed through the crude product to remove entrained HF. The distillation to remove is an alternative, e.g. by a multiple distillation as described in WO 2011/020830. Both methods can be combined with each other or with an adsorptive treatment to remove HF, e.g. by contacting with silica.

A combination of stripping and distillation is especially preferred as post steps of purification.

The organosilane compound can be alternatively added after the pre-purification step mentioned above to remove a major part of present HF, and it is preferably added before a final separation step such as distillation.

In a further embodiment of the present invention, the method for purification is conducted in several steps to provide purified carbonate, comprising the following steps:



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- a) at least one step of reacting fluorine with an organic carbonate with a lower degree of fluorination to obtain a crude reaction mixture comprising fluorinated organic carbonate containing hydrogen fluoride ;
- b) at least one step of treating the crude reaction mixture comprising fluorinated organic carbonate with the organosilicon compound having at least one -Si-N-bond to remove hydrogen fluoride ;
- 5 c) at least one step of further removing the hydrogen fluoride contained in the pre-purified fluorinated organic carbonate in which a content of the hydrogen fluoride is decreased ; and
- 10 d) at least one step of recovering a purified fluorinated organic carbonate by distillation of the purified fluorinated organic carbonate.

The content of hydrogen fluoride in the reaction products after the treatment according to the present invention is preferably equal to or lower than 5% by weight, more preferably equal to or lower than 2 % by weight of the reaction mixture. Especially preferably, it is equal to or lower than 1 % by weight. Still more preferably, it is equal to or lower than 0.5 % by weight. Yet more preferably, it is equal to or lower than 0.1 % by weight.

More details for post-purification of the fluorinated organic carbonates are described below.

20 In a most simple manner, stripping could be performed in a vessel containing reaction mixture by blowing inert gas through the reaction mixture. This can be done batch wise or continuously.

It is preferred to perform stripping in a way which provides for a sufficient contact area between reaction mixture and gas. For example, reaction mixture could be sprayed into a stream of inert gas, or stripping gas and liquid to be treated can be contacted in a bubble tray column. A very preferred method is performed in a stripping column. In a stripping column, internals or packings are installed with a high specific area per m<sup>3</sup> of the equipment to provide a high contact surface between gas and liquid. Suitable packings are, for example, Raschig rings. The stripping column is usually a cylindrical tube positioned vertically. The inert gas is introduced at the bottom of the stripping column below the packings; the reaction mixture is fed at the top. Inert gas comprising hydrogen fluoride leaves the column via a separate line at the top.

30 The efficiency of the removal of hydrogen fluoride from the HF-containing carbonate is higher at higher temperatures. If the contact is performed in a vessel, heat can be supplied in a known manner, for example, by heating the

walls of the vessel. Optionally, the inert gas and/or the liquid to be treated can be heated.

If the reaction is performed in a stripping column with internals or packings, it is preferred to heat inert gas, liquid to be treated or both to improve the efficiency of the stripping process.

Thus, the inert gas, especially nitrogen, advantageously is heated before introducing it into the reaction mixture. The temperature to which it is heated is preferably equal to or higher than 60°C ; more preferably, it is equal to or higher than 75°C. Very preferably, it is equal to or higher than 100°C. The temperature can still be higher, for example, equal to or higher than 120°C. Preferably, it is equal to or lower than 150°C. Depending on the heat resistance and the resistance of corrosion of the vessel, column, pipes, fittings etc used, the temperature can be higher than 150°C.

The reaction mixture preferably is also heated before a continuous stripping process is performed. If a vessel is used to perform a batch wise process, the reaction mixture can be heated before and/or during the stripping process. Preferably, it is heated to a temperature equal to or greater than 60°C. Preferably, it is heated to a temperature equal to or lower than 120°C.

It is advantageous to perform the stripping step at ambient pressure. If desired, a slight vacuum can be applied. For example, the pressure can be reduced to 0.5 bar or even 0.2 bar. The temperature should not be so high that organic compounds would be carried out of it with the flow of inert gas.

In a batch wise process, stripping is performed until the desired maximal amount of hydrogen fluoride is present.

In a continuous process in a stripping column, the height of the column is selected such that, for a given hydrogen fluoride concentration, flow rate of inert gas and reaction mixture, the desired residual hydrogen fluoride concentration is reached.

The post-purification can also be performed by several distillations as described in WO 2011/020830. In this method, a pre-purified reaction mixture is distilled in at least two distillation steps wherein the reaction mixture which is fed to the first distillation step. Excess hydrogen fluoride can be removed for example by the treatment with the organosilicon compound according to the present invention and/or stripping before the distillation steps.

The wording "at least two distillation steps" denotes passing the mixture at least twice through a distillation column. According to one embodiment, this is

one distillation column through which the mixture to be separated is passed at least twice. This embodiment can be performed in a batch wise distillation.

According to another embodiment, the at least two distillation steps are performed in at least two distillation columns. This embodiment is especially  
5 suitable for performing a continuous distillation process.

In the first distillation step, a mixture of substances with a lower boiling point (for example, hydrogen fluoride and carbonates with higher degree of fluorination) is drawn off from the top; the higher boiling constituents are drawn off from the bottom and are fed into the second distillation step. Often, the  
10 pressure at the top of the column of the first distillation step is equal to or lower than 100 mbar (abs). Preferably, the pressure at the top of the column of the first distillation step is equal to or lower than 75 mbar (abs.). Preferably, it is equal to or higher than 10 mbar (abs.). A pressure at the top of the column of the first distillation step in the range between 10 and 50 mbar (abs.) is especially  
15 preferred.

The mixture of substances with a lower boiling point drawn off from the top of the column of the first distillation step can be separated from each other if desired. For example, hydrogen fluoride can be removed by washing the mixture with water or, which is highly preferred, by stripping the mixture with  
20 an inert gas. The remaining fluorinated carbonates can be separated by distillation. Alternatively, the mixture from the top of the column of the first distillation step can be separated into the different compounds simply by distillation without any other treatment like washing or stripping. Carbonates with higher degree of fluorination are valuable side products because they can be  
25 applied as additive for lithium ion battery solvents. If desired, they may be dumped or burned. Any recovered hydrogen fluoride also is a valuable product *per se*.

In the second column, the bottom product of the first column is distilled. Preferably, the pressure at the top of the column of the second distillation step is  
30 equal to or lower than 50 mbar (abs.). More preferably, the pressure at the top of the second column is equal to or lower than 30 mbar (abs.). Preferably, the pressure at the top of the column of the second distillation step is equal to or higher than 5 mbar (abs). At the top of the column of the second distillation step, highly pure fluorinated carbonate, for example, monofluoroethylene  
35 carbonate, is obtained. The content of hydrogen fluoride in the purified organic carbonate is equal to or lower than 30 ppm by weight, preferably equal to or

lower than 20 ppm by weight. Even lower hydrogen fluoride content can be achieved, e.g. equal to or lower than 10 ppm.

Should the disclosure of any patents, patent applications, and publications which are incorporated herein by reference conflict with the description of the present application to the extent that it may render a term unclear, the present description shall take precedence.

While preferred embodiments of this invention have been shown and described, modifications thereof can be made by one skilled in the art without departing from the spirit or teaching of this invention. The embodiments and examples described herein are exemplary only and are not limiting. Many variations and modifications of systems and methods are possible and are within the scope of the invention. Accordingly, the scope of protection is not limited to the embodiments described herein, but is only limited by the claims that follow, the scope of which shall include all equivalents of the subject matter of the claims.

### **Examples**

*Example 1-1. Preparation of a crude reaction mixture comprising monofluoroethylene carbonate ("F1EC")*

The manufacture of F1EC is performed by the reaction of ethylene carbonate ("EC") with F2/N2 in a reactor. Simultaneously, 1,1,1,3,3,3-hexamethyldisilazane compound is added to the reactor to reduce the content of free HF. The crude reaction mixture comprises not only EC and F1EC, but also HF and further fluorinated ethylene carbonates, such as difluoroethylene carbonate ("F2EC").

*Example 1-2. Purification of the reaction mixture*

The reaction mixture from Example 1-1 in which the content of HF is reduced by the treatment with 1,1,1,3,3,3-hexamethyldisilazane compound is subjected to stripping by contacting the same with nitrogen gas in a stripping column to further reduce the content of HF.

*Example 1-3. Distillation*

After the purification, the resulting reaction mixture is distilled to provide final product, which is F1EC. The amount of free HF in the final product is significantly decreased, providing a highly purified F1EC.

CLAIMS

1. A method for purifying fluorinated organic carbonate comprising a step of treating the fluorinated organic carbonate with an organosilicon compound having at least one -Si-N- bond, and a step of distillation of the resulting mixture in at least one distillation column.
2. The method according to claim 1, wherein the organosilicon compound having at least one -Si-N- bond is selected from the group consisting of organosilazane compounds, organodisilazane compounds and organotrisilazane compounds.
3. The method according to claim 1 or 2, wherein the organosilicon compound having at least one -Si-N- bond is selected from the group consisting of (N,N-diethylamino)trimethylsilane, N,O-bis(trimethylsilyl)acetamide, N,N'-bis(trimethylsilyl)-1,4-butanediamine, 1,1,1,3,3,3-hexamethyldisilazane, 1,1,3,3,5,5-hexamethylcyclotrisilazane, and any combination thereof.
4. The method according to any one of claims 1 to 3, wherein the organosilicon compound having at least one -Si-N- bond is 1,1,1,3,3,3-hexamethyldisilazane.
5. The method according to any one of claims 1 to 4, wherein the fluorinated organic carbonate is a crude reaction mixture obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution by fluorine, or a pre-purified product obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution by fluorine and at least one step of removing hydrogen fluoride.
6. The method according to claim 5, wherein the fluorinated organic carbonate is a crude fluorinated organic carbonate obtained from a reaction including at least one step of reacting elemental fluorine with an organic carbonate with a lower degree of substitution and at least one step of removing hydrogen fluoride.

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7. The method according to claim 5 or 6, wherein the step of removing hydrogen fluoride is conducted by stripping with at least one gas, distillation, or both of them.

5 8. The method according to any one of claims 1 to 7, wherein the fluorinated organic carbonate is selected from the group consisting of monofluoroethylene carbonate, fluoromethyl methyl carbonate, difluoroethylene carbonate, and bis-(fluoromethyl) carbonate.

10 9. The method according to any one of claims 1 to 8, wherein the amount of the organosilicon compound used for the treatment is, relative to the fluorinated organic carbonate to be treated, 0.01 to 5 % by weight.

10. The method according to any one of claims 1 to 9, comprising the following steps:

15 a) at least one step of reacting fluorine with an organic carbonate with a lower degree of fluorination to obtain a crude reaction mixture comprising fluorinated organic carbonate containing hydrogen fluoride ;

b) at least one step of treating the crude reaction mixture comprising fluorinated organic carbonate with the organosilicon compound having at least one -Si-N-bond to remove hydrogen fluoride ;

20 c) at least one step of further removing the hydrogen fluoride contained in the pre-purified fluorinated organic carbonate in which a content of the hydrogen fluoride is decreased ; and

d) at least one step of recovering a purified fluorinated organic carbonate by distillation of the purified fluorinated organic carbonate.

25 11. The method according to claim 10, wherein the steps (a) and (b) are conducted simultaneously in the same reactor.

# INTERNATIONAL SEARCH REPORT

International application No <b>PCT/EP2013/070974</b>
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<b>A. CLASSIFICATION OF SUBJECT MATTER</b> INV. C07D317/42 ADD.		
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b>		
Minimum documentation searched (classification system followed by classification symbols) C07D		
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 practicable, search terms used) EPO-Internal, WPI Data		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO 2011/020830 A1 (SOLVAY FLUOR GMBH [DE]) 24 February 2011 (2011-02-24) cited in the application the whole document -----	1-11
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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2013/070974

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