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<p>(54) Title: STERILIZING COMPOSITIONS OF PENTAFLUOROPROPANE AND ETHYLENE OXIDE</p> <p>(57) Abstract</p> <p>Azeotrope-like and nonazeotropic compositions of 1,1,1,3,3-penta fluoropropane and ethylene oxide are provided. The compositions of the invention are useful as sterilizing agents.</p>		

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STERILIZING COMPOSITIONS OF PENTAFLUOROPROPANE AND ETHYLENE OXIDE

Field of the Invention

The invention relates to compositions of 1,1,1,3,3-pentafluoropropane and
5 ethylene oxide. The compositions of the invention are useful as sterilizing agents.

Background of the Invention

Sterilization with a germicidal agent, such as ethylene oxide gas or an ethylene
oxide-containing gas mixture, plays an important role in sterilizing heat sensitive
materials. Rapid growth in the use of sterile, disposable, medical devices is just one
10 consequence of gaseous sterilization with such agents.

In the basic gaseous sterilization process, a sterilization chamber containing
articles to be sterilized is evacuated and the articles are preconditioned at an optimal
relative humidity, generally between about 20 and 70 percent. The sterilizing gas is
admitted at an appropriate pressure and temperature while contact is maintained
15 between the sterilizing atmosphere and the articles to be sterilized. After an
appropriate time period, the chamber is evacuated to remove the sterilant gas.
Although there are many variations on this basic process, the major factors that must
be controlled to effect sterilization are exposure time, temperature, ethylene oxide
pressure or partial pressure, and relative humidity.

20 Two blends that have been used in sterilizing processes are
dichlorodifluoromethane ("CFC-12")/ethylene oxide and carbon dioxide/ethylene
oxide. A disadvantage of using fully halogenated chlorofluorocarbons, such as CFC-
12, is that they have substantial potential for stratospheric ozone depletion and global
warming. Additionally, the CFC-12 and carbon dioxide blends are non-azeotropic and,
25 thus, suffer the disadvantage of segregating during vaporization leading to potentially
flammable or explosive situations.

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A candidate diluent preferably should be miscible with ethylene oxide in the liquid phase and should not be so highly volatile that it will segregate from the ethylene oxide to any great extent during vaporization. Azeotrope-like mixtures are useful in this context because they do not fractionate by normal evaporation or distillation processes. Therefore, a need exists for an environmentally acceptable diluent that is compatible with the objects being sterilized, is chemically stable, is minimally segregating, and provides sufficient vapor pressure to deliver the liquid mixture to the sterilization chamber.

Description of the Invention and Preferred Embodiments

The invention provides sterilizing gas compositions and methods for using the compositions. In one embodiment, the invention provides a method for sterilizing articles comprising, consisting essentially of, or consisting of the step of exposing the articles to a mixture of ethylene oxide and 1,1,1,3,3-pentafluoropropane ("HFC-245fa"). In another embodiment of the invention, mixtures of ethylene oxide and HFC-245fa are provided, which mixtures comprise, consist essentially of, or consist of ethylene oxide and HFC-245fa.

For purposes of this invention, by "mixtures of ethylene oxide and HFC-245fa" is meant both nonazeotropic and azeotrope-like compositions of ethylene oxide and HFC-245fa. Thus, in yet another embodiment of the invention, the invention provides azeotrope-like and nonazeotropic compositions comprising from about 1 to about 99 weight percent ethylene oxide and from about 99 to about 1 weight percent HFC-245fa.

In a preferred embodiment, the invention provides azeotrope-like compositions comprising, consisting essentially of, and consisting of effective amounts of ethylene oxide and HFC-245fa. By "effective amounts" is meant the amount of each component that, in combination with the other component, results in the formation of an azeotrope-like composition. In a more preferred embodiment, the invention comprises azeotrope-like compositions comprising from about 4 to about 50, preferably from about 10 to about 40, weight percent ethylene oxide and from about

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96 to about 50, preferably from about 90 to about 60, weight percent HFC-245fa having a vapor pressure of about 7.4 psia to about 8.6 psia at about 0° C.

In yet another preferred embodiment, nonazeotropic sterilant gas compositions comprising from about 96 to about 50 weight percent ethylene oxide and from about 4
5 to about 50 weight percent HFC-245fa are provided.

For purposes of this invention, azeotrope-like compositions are compositions that behave like azeotropic mixtures. From fundamental principles, the thermodynamic state of a fluid is defined by pressure and temperature as well as liquid composition and vapor composition. An azeotropic mixture is a system of two or more components in
10 which the liquid composition and vapor composition are equal at the state pressure and temperature. In practice, this means that the components of an azeotropic mixture are constant boiling and cannot be separated during a phase change.

Azeotrope-like compositions behave like azeotropic mixtures, *i.e.*, or are constant boiling or essentially constant boiling. In other words, for azeotrope-like
15 compositions, the composition of the vapor formed during boiling or evaporation is identical, or substantially identical, to the original liquid composition. Thus, with boiling or evaporation, the liquid composition changes, if at all, only to a minimal or negligible extent. This is to be contrasted with non-azeotrope-like compositions in which, during boiling or evaporation, the liquid composition changes to a substantial
20 degree. All azeotrope-like compositions of the invention within the indicated ranges as well as certain compositions outside these ranges are azeotrope-like.

The azeotrope-like compositions of the invention may include additional components that do not form new azeotropic or azeotrope-like systems, or additional components that are not in the first distillation cut. The first distillation cut is the first
25 cut taken after the distillation column displays steady state operation under total reflux conditions. One way to determine whether the addition of a component forms a new azeotropic or azeotrope-like system so as to be outside of this invention is to distill a sample of the composition with the component under conditions that would be expected to separate a nonazeotropic mixture into its separate components. If the

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mixture containing the additional component is nonazeotropic or nonazeotrope-like, the additional component will fractionate from the azeotropic or azeotrope-like components. If the mixture is azeotrope-like, some finite amount of a first distillation cut will be obtained that contains all of the mixture components that is constant boiling or behaves as a single substance.

It follows from this that another characteristic of azeotrope-like compositions is that there is a range of compositions containing the same components in varying proportions that are azeotrope-like, or constant boiling. All such compositions are intended to be covered by the terms "azeotrope-like" and "constant boiling". As an example, it is well known that at differing pressures, the composition of a given azeotrope will vary at least slightly as does the boiling point of the composition. Thus, an azeotrope of A and B represents a unique type of relationship, but with a variable composition depending on temperature and/or pressure. It follows that, for azeotrope-like compositions, there is a range of compositions containing the same components in varying proportions that are azeotrope-like. All such compositions are intended to be covered by the term azeotrope-like as used herein.

The components of the composition of the invention are known materials that are commercially available or may be prepared by known methods. Preferably, the components are of sufficiently high purity so as to avoid the introduction of adverse influences on the properties of the system.

It is a further discovery of this invention that HFC-245fa is not as good a flame suppressant as other ethylene oxide diluents. Therefore, in a most preferred embodiment, the invention provides azeotrope-like and nonazeotropic compositions comprising, consisting essentially of, and consisting of ethylene oxide and HFC-245fa and a flame suppressant effective amount of at least one diluent. A flame suppressant effective amount of diluent is an amount sufficient to increase the flame suppressing effect of the HFC-245fa. Generally, an amount of from about 60 to about 90, preferably from about 66 to about 84, weight percent of diluent is used, based on the sum of the weight percentages of the ethylene oxide and HFC-245fa components.

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Suitable diluents are any diluent other than HFC-245fa that is more volatile than the ethylene oxide/HFC-245fa blend. Illustrative diluents include, without limitation, 1,1,2,2,2-pentafluoroethane ("HFC-125"), chlorodifluoromethane ("HCFC-22"), 1,1,1,2-tetrafluoroethane ("HFC-134a"), 1,1,2,2-tetrafluoroethane ("HFC-134"), 1,1,1,3,3,3-hexafluoropropane (HFC-236fa), 1,1,1,2,3,3,3-heptafluoropropane (HFC-227ea), nitrogen, carbon dioxide, and sulfur hexafluoride. Other suitable diluents will readily occur to those ordinarily skilled in the art. The preferred additional diluent is HFC-125.

The compositions containing the azeotrope-like composition and diluent should be greater than about 6 weight percent, preferably from about 8 to about 13 weight percent, more preferably from about 9 to about 11 weight percent, most preferably about 10 weight percent, ethylene oxide. Preferred compositions containing these mole percents of ethylene oxide are shown on Table 1. The values on Table 1 are understood to be prefaced by the term "about".

15

Table 1

Weight % EO	Weight % 245fa	Weight % 125
6	9	85
7	11	82
8	13	79
9	14	77
10	14	76
11	16	73
13	20	67

In a process embodiment of the invention, the compositions of the invention are used as sterilizing gases in any manner well known in the art such as by exposing the article to be sterilized to the sterilizing gas under conditions and for a period of time necessary to achieve the desired degree of sterility. Typically, the process is effected

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by placing the article to be sterilized in a chamber, evacuating the chamber, humidifying the chamber, and exposing the article to the sterilizing gas for an appropriate period of time.

The sterilant compositions of the invention may be used to sterilize a great many articles. Examples of medical equipment and materials that may be sterilized include, without limitation, diagnostic endoscopes, syringes, test tubes, incubators, pacemakers, tubing, gloves, catheters, sheeting, needles, scalpels, dilators, pumps, and intraocular lenses. Additionally, the compositions may be used as a fumigant for items outside of the medical field including, without limitation, foodstuffs, furs, bedding, and paper goods.

The sterilant compositions are effective against all forms of life, particularly insects, bacteria, viruses, molds, fungi, and other microorganisms. The compositions of the invention may be prepared using any effective mixing technique known to those ordinarily skilled in the art. For example, each compound may be pumped gravimetrically through a manifold into a sterilant container and the container rolled to intermix the compounds into a homogeneous composition. Alternatively, the compounds may be pumped into a mixing tank, recirculated in the tank until a fully homogeneous composition is formed, and then pumped from the mixing tank into a sterilant container.

The compositions of the invention may be packaged in any storage containers of suitable design such as United States Department of Transportation ("DOT") Specifications 4BA 240, 4BA 300, 4BW 240, or other DOT specification cylinders or trailers of suitable working pressure. The compositions also may be packaged in American Society of Mechanical Engineers storage vessel.

The storage cylinders may be delivered to the use site holding the sterilant compositions at a pressure generally within the range of from about 70 to about 190 pounds per square inch absolute at 70° F. The cylinders are then connected through a

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series of valves, control valves, vaporizers, and appropriate conduits to carry out the sterilization

The invention will be clarified further by a consideration of the following, non-limiting examples.

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Example 1

Two cylinders connected to a differential pressure gauge ranging from -4 to +4 psi were filled with 3.5g of HFC-245fa. The initial gauge reading was -0.1 psi. Ethylene oxide was added in small increments to the sample cylinder and the vapor pressure differences between the two cylinders was measured and recorded. The ethylene oxide vapor pressure at 0° C is 1.24 psi higher than that of HFC-245fa and, thus, a pressure rise when the ethylene oxide was added was predicted.

However, experimentally, the vapor pressure was lower than that of HFC-245fa when the ethylene oxide was added meaning that the boiling point of the mixture was higher than that of either component. The result demonstrated that HFC-245fa and ethylene oxide form an azeotrope. The following Table 2 shows the results of the incremental addition of ethylene oxide to the HFC-245fa.

Table 2

Weight % EO in HFC-245fa	Pressure Difference From HFC-245fa reference Cylinder
0	-0.1
4.35	-0.46
9.04	-0.46
19.6	-0.28
22.6	-0.38
30.4	+0.05
34.1	+0.29
46.8	+0.53

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What is claimed is:

1. A method for sterilizing articles comprising the step of exposing the articles to a sterilizing gas composition comprising sterilizing effective mixtures of ethylene oxide and 1,1,1,3,3-pentafluoropropane.
- 5 2. The method of claim 1 wherein the mixture of ethylene oxide and 1,1,1,3,3-pentafluoropropane is an azeotrope-like composition comprising effective amounts of ethylene oxide and 1,1,1,3,3-pentafluoropropane.
3. The method of claim 1 wherein the mixture of ethylene oxide and 1,1,1,3,3-pentafluoropropane is an azeotrope-like composition consisting essentially of
10 effective amounts of ethylene oxide and 1,1,1,3,3-pentafluoropropane.
4. The method of claim 2 wherein the azeotrope-like composition comprises from about 4 to about 50 weight percent ethylene oxide and from about 96 to about 50 weight percent 1,1,1,3,3-pentafluoropropane.
5. The method of claim 4 wherein a flame suppressant effective amount of a diluent is
15 used.
6. The method of claim 5 wherein the flame suppressant effective amount of diluent is an amount of from about 60 wt% to about 90 wt%.
7. The method of claim 5 wherein the diluent is selected from the group consisting of 1,1,2,2,2-pentafluoroethane, chlorodifluoromethane, 1,1,1,2,3,3,3-
20 heptafluoropropane, 1,1,1,3,3,3-hexafluoropropane, nitrogen, and carbon dioxide.
8. The method of claim 7 wherein the diluent is 1,1,2,2,2-pentafluoroethane.
9. The method of claim 1 wherein the mixture of ethylene oxide and 1,1,1,3,3-pentafluoropropane is a nonazeotropic composition comprising ethylene oxide and 1,1,1,3,3-pentafluoropropane.

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10. The method of claim 9 wherein the ethylene oxide is present in an amount of from about 99 to about 51 weight percent and the 1,1,1,3,3-pentafluoropropane is present in an amount of from about 1 to about 49 weight percent.
- 5 11. The method of claim 10 wherein a flame suppressant effective amount of a diluent is used.
12. The method of claim 11 wherein the flame suppressant effective amount of diluent is an amount of from about 60 wt% to about 90 wt%.
13. The method of claim 10 wherein the diluent is selected from the group consisting of 1,1,2,2,2-pentafluoroethane, chlorodifluoromethane, 1,1,1,2,3,3,3-
10 heptafluoropropane, 1,1,1,3,3,3-hexafluoropropane, nitrogen, and carbon dioxide.
14. The method of claim 13 wherein the diluent is 1,1,2,2,2-pentafluoroethane.
15. A sterilizing gas composition comprising ethylene oxide and 1,1,1,3,3-pentafluoropropane.
16. The sterilant gas composition of claim 15 further comprising a flame suppressant
15 effective amount of a diluent.
17. The sterilant gas composition of claim 16 wherein the flame suppressant effective amount of diluent is an amount of from about 60 wt% to about 90 wt%.
18. The sterilant gas composition of claim 17 wherein the diluent is selected from the group consisting of 1,1,2,2,2-pentafluoroethane, chlorodifluoromethane,
20 1,1,1,2,3,3,3-heptafluoropropane, 1,1,1,3,3,3-hexafluoropropane, nitrogen, and carbon dioxide.
19. The sterilant gas composition of claim 18 wherein the diluent is 1,1,2,2,2-pentafluoroethane.
20. An azeotrope-like composition comprising effective amounts of ethylene oxide and
25 1,1,1,3,3-pentafluoropropane.

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21. The azeotrope-like composition of claim 20 consisting essentially of effective amounts of ethylene oxide and 1,1,1,3,3-pentafluoropropane.
22. The azeotrope-like composition of claim 20 comprising from about 4 to about 50 weight percent ethylene oxide and from about 96 to about 50 weight percent
5 1,1,1,3,3-pentafluoropropane having a vapor pressure of about 7.6 psia to about 8.6 psia at about 0° C.
23. The azeotrope-like composition of claim 20 wherein the amount of ethylene oxide is from about 10 to about 40 weight percent and the amount of 1,1,1,3,3-pentafluoropropane is from about 90 to about 60 weight percent.

INTERNATIONAL SEARCH REPORT

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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)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
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A	US 5 254 309 A (FELIX VINCI M ET AL) 19 October 1993 see claims; examples ---	1-23
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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
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