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(54) Title: REFRIGERANT COMPOSITION COMPRISING DIFLUOROMETHANE (HFC32) AND 2,3,3,3-TETRAFLUOROPROPENE (HFO1234yf)

(57) Abstract: The present invention provides a refrigerant with a comprehensively lowered environmental impact, including a low global warming potential resulting in an excellent effect on direct environmental impact, and a good energy efficiency when charged into an apparatus resulting in an excellent effect on indirect environmental impact, and low LCCP. Specifically, the present invention relates to a refrigerant composition containing 10 to 26 mass% of difluoromethane (HFC32) and 74 to 90 mass% of 2,3,3,3-tetrafluoropropene (HFO1234yf).



WO 2010/002016 A1

-1-

DESCRIPTION

Title of Invention: REFRIGERANT COMPOSITION COMPRISING
DIFLUOROMETHANE (HFC32) AND 2,3,3,3-TETRAFLUOROPROPENE
(HFO1234yf)

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Technical Field

The present invention relates to a mixed refrigerant composition containing difluoromethane (HFC32) and 2,3,3,3-tetrafluoropropene (HFO1234yf) for use in refrigeration and air-conditioning systems.

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Background Art

With global warming becoming an increasingly serious issue worldwide, the development of environmentally friendly refrigeration and air-conditioning systems has become increasingly important. Refrigerants have a great influence not only on global warming, but also on the performance of refrigeration and air-conditioning systems. Therefore, the selection of a refrigerant has an important role in reducing carbon dioxide emissions that contribute to global warming.

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Recently, various partially-fluorinated propenes having a double bond in the molecule, with a lower global warming potential (GWP) than known chlorofluorocarbons (CFCs), hydrochlorofluorocarbons (HCFCs), and hydrofluorocarbons (HFCs), have been proposed.

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2,3,3,3-Tetrafluoropropene (HFO1234yf) is one such propene (see, for example, Patent Literatures 1 and 2). Compared to HCFC22 conventionally used for stationary air conditioners, or to R407C and R410A developed as substitutes for HCFC22 that are uninvolved in the depletion of the ozone layer, 2,3,3,3-tetrafluoropropene has a high boiling point, and therefore fails to retain refrigerating capacity when used in a single component.

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In selecting a refrigerant, a low global warming potential (GWP) is obviously important; however, the energy use efficiency of the device using the refrigerant is equally, or

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-2-

more important. The former is evaluated as a direct environmental impact, and the latter is as an indirect environmental impact. LCCP (Life Cycle Climate Performance) has been proposed as an objective index for these evaluations (see, for example, Non-Patent Literature 1). LCCP is now widely recognized for use in the comprehensive evaluation of refrigerants. Evaluations including an LCCP evaluation allow for the clear detection of environmental influence, and the appropriate selection of a refrigerant with a low environmental impact.

Citation List

Patent Literature

PTL 1: WO2005/105947

PTL 2: WO2006/094303

Non Patent Literature

NPL 1: "LIFE CYCLE CLIMATE PERFORMANCE OF SOME APPLICATIONS IN JAPAN", HARUO ONISHI, 15th Annual Earth Technologies Forum and Mobile Air Conditioning Summit, April 13-15, 2004 Conference Proceedings

Summary of Invention

Technical Problem

When a refrigerant having a high boiling point is used at a low operating pressure, the vapor compression refrigeration cycle has an insufficient capacity. Therefore, it is necessary to increase the size of the device etc. to ensure the desired cooling or heating capacity, which normally results in a deteriorated indirect impact due to pressure loss. An object of the present invention is to provide a refrigerant, the refrigerant with a low GWP resulting in an excellent effect on direct environmental impact, and with a good energy efficiency when charged into an apparatus resulting in an excellent effect on indirect environmental impact; such excellent effects resulting in low LCCP, allowing production of a refrigerant with

-3-

a comprehensively lowered environmental impact.

Solution to Problem

The present inventors carried out extensive research
5 in view of the above problems, and found that the problems can be
solved by using a refrigerant composition containing 10 to 26
mass% of difluoromethane (HFC32) and 74 to 90 mass% of 2,3,3,3-
tetrafluoropropene (HFO1234yf), and preferably 14 to 22 mass% of
HFC32 and 78 to 86 mass% of HFO1234yf, in an apparatus in which
10 the refrigerant is circulated through a compressor to form a
refrigeration cycle. The present invention has been accomplished
based on the above finding.

Specifically, the present invention provides the
following refrigerant composition.

15 Item 1. A refrigerant composition comprising 10 to 26
mass% of difluoromethane (HFC32) and 74 to 90 mass% of 2,3,3,3-
tetrafluoropropene (HFO1234yf).

Item 2. The refrigerant composition according to Item
1, comprising 14 to 22 mass% of difluoromethane (HFC32) and 78 to
20 86 mass% of 2,3,3,3-tetrafluoropropene (HFO1234yf).

Item 3. The refrigerant composition according to Item 1
or 2, which further comprises a polymerization inhibitor.

Item 4. The refrigerant composition according to any
one of Items 1 to 3, which further comprises a drying agent.

25 Item 5. The refrigerant composition according to any
one of Items 1 to 4, which further comprises a stabilizer.

Item 6. A method of operating a refrigerator comprising
circulating the refrigerant composition according to any one of
Items 1 to 5 through a compressor.

30 Item 7. A method of producing the refrigerant
composition according to Item 1, comprising mixing 10 to 26 mass%
of difluoromethane (HFC32) with 74 to 90 mass% of 2,3,3,3-
tetrafluoropropene (HFO1234yf).

Item 8. A refrigerator comprising the refrigerant
35 composition according to any one of Items 1 to 5.

Advantageous Effects of Invention

The following effects can be achieved by the refrigerant composition of the invention.

- 5 (1) The refrigerant composition of the invention has a low global warming potential (GWP) compared to conventionally available refrigerants such as R407C and R410A.
- (2) The refrigerant composition of the invention has no ozone depletion potential (ODP); therefore, it does not contribute
10 to ozone layer depletion, even when not completely collected after use.
- (3) The refrigerant composition of the invention is excellent in LCCP evaluation, and its contribution to global warming is as low as, or lower than, that of the conventionally
15 available refrigerants such as R407C or R410A, when used as a refrigerant for a heat pump apparatus.

Brief Description of Drawing

FIG. 1 is a graph showing LCCP in the refrigerant
20 compositions obtained in Examples (see Test Examples 1 to 3) and Comparative Examples.

Description of Embodiments

The present inventor conducted extensive research on
25 the relationship between LCCP (Life Cycle Climate Performance) and the mixing ratios of HFC32 and HFO1234yf. The LCCP was evaluated according to the method described in Test Example 1.

The evaluation results reveal that the refrigerant composition containing 10 to 26 mass% of HFC32 and 74 to 90 mass%
30 of HFO1234yf (the refrigerant composition containing HFC32/HFO1234yf at a ratio of 10/90 to 26/74 mass%) has a lower environmental impact and achieves excellent effects in LCCP evaluation, while retaining a refrigerating capacity. Further, the refrigerant composition containing 12 to 25 mass% of HFC32
35 and 75 to 88 mass% of HFO1234yf (the refrigerant composition

-5-

containing HFC32/HFO1234yf at a ratio of 12/88 to 25/75 mass%) achieves more excellent effects, and the refrigerant composition containing 14 to 22 mass% of HFC32 and 78 to 86 mass% of HFO1234yf (the refrigerant composition containing HFC32/HFO1234yf at a ratio of 14/86 to 22/78 mass%) achieves most excellent effects.

The GWP (Integration Time Horizon; ITH=100yr) of HFC32 is 675, and that of HFO1234yf is 4. When the content of HFC32 in the mixed refrigerant becomes 21.8 mass% or less, the GWP (ITH=100yr) of the mixed refrigerant becomes 150 or less.

The composition of the invention has high stability. If necessary, stabilizers may be added to meet the requirement of high stability under severe conditions.

Examples of such stabilizers include (i) aliphatic nitro compounds such as nitromethane and nitroethane; aromatic nitro compounds such as nitrobenzene and nitrostyrene; (ii) ethers such as 1,4-dioxane; amines such as 2,2,3,3,3-pentafluoropropylamine and diphenylamine; butylhydroxyxylene, benzotriazole, etc. The stabilizers can be used singly or in a combination of two or more.

The amount of stabilizer may vary depending on the type of stabilizer, as long as it does not impair the performance of the composition. In general, the amount of stabilizer is preferably about 0.01 to about 5 parts by weight, and more preferably about 0.05 to about 2 parts by weight, per 100 parts by weight of the mixture of HFC32 and HFO1234yf.

The composition of the present invention may further contain a polymerization inhibitor. Examples thereof include 4-methoxy-1-naphthol, hydroquinone, hydroquinone methyl ether, dimethyl-t-butyl phenol, 2,6-di-tert-butyl-p-cresol, benzotriazole, etc.

In general, the amount of polymerization inhibitor is preferably about 0.01 to about 5 parts by weight, and more preferably about 0.05 to about 2 parts by weight, per 100 parts by weight of the mixture of HFC32 and HFO1234yf.

-6-

The composition of the present invention may further contain a drying agent.

A refrigeration cycle can be formed by circulating the refrigerant composition of the present invention through a compressor. It is also possible to produce an apparatus for forming a refrigeration cycle in which the refrigerant composition of the present invention is circulated through a compressor.

Examples of refrigerating systems capable of using the refrigerant composition of the invention include, but are not limited to, car air conditioners, refrigerating units for automatic vending machines, industrial/household air conditioners, gas heat pumps (GHP)/electrical heat pumps (EHP), etc. Particularly, the present refrigerant composition is applicable for industrial or household air conditioners whose downsizing is desired.

Examples

The present invention will be described in more detail below by way of Examples; however, the invention is not limited thereto.

Test Example 1

HFC32 and HFO1234yf were used as refrigerants, and mixed in the following ratio (HFC32/HFO1234yf): 10.6/89.4 in Example 1, 14.3/85.7 in Example 2, 18.0/82.0 in Example 3, 21.8/78.2 in Example 4, and 25.5/74.5 in Example 5 (figures are expressed in mass%). Using a heat pump apparatus, operation was conducted under the following conditions:

[cooling rated] capacity: 4kW, evaporating temperature of the refrigerant in the evaporator: 10°C, condensing temperature of the refrigerant in the condenser: 45°C;

[cooling intermediate] capacity: 2kW, evaporating temperature: 17°C, condensing temperature: 42°C;

[heating rated] capacity: 5kW, evaporating temperature: 0°C,

-7-

condensing temperature: 42°C;
 [heating intermediate] capacity: 2.5kW, evaporating temperature:
 2°C, condensing temperature: 32°C.

5 Degrees of superheat and subcool were set to 0°C in
 each condition.

The refrigerant pipe used for the operation had a
 length of 7.5 m.

As Comparative Examples, HFC32 and HFO1234yf were used
 as refrigerants and mixed in the following ratio: 5.0/95.0 in
 10 Comparative Example 1, 6.9/93.1 in Comparative Example 2,
 29.2/70.8 in Comparative Example 3, 36.7/63.3 in Comparative
 Example 4, and 44.1/55.9 in Comparative Example 5 (figures are
 expressed in mass%). The heat pump apparatus with the same
 refrigerant pipe length as above was operated under the same
 15 conditions.

The results were used for calculating the annual power
 consumption (kWh) in conformity with JRA 4046:2004. APF (Annual
 Performance Factor) was obtained using the following formula.

$$\text{APF} = (\text{annual total load}) / (\text{annual power consumption})$$

20 The sum of the cooling/heating capacity required for
 year (annual total load) is 8,015 kWh when the rated cooling
 capacity is 4kW.

Based on the above result, the LCCP was evaluated.

$$\text{LCCP} = \text{Direct impact (kg-CO}_2\text{)} + \text{Indirect impact (kg-CO}_2\text{)}$$

25 Direct impact = (leakage during charging at a manufacturing
 plant) + (annual regular leakage) + (annual irregular leakage) +
 (leakage during servicing) + (leakage during disposal)

Indirect impact = (CO₂ emissions during the use of the air
 conditioning system) + (CO₂ emissions during production and
 30 transportation of the refrigerant)

Specifically, the direct impact and indirect impact can
 be obtained by the following formulae.

$$\text{Direct impact} = \text{GWP} \times \text{M} \times (1-\alpha) + \text{GWP}_{\text{AE}} \times \text{M}$$

$$\text{Indirect impact} = \text{N} \times \text{E} \times \beta$$

35 GWP: global warming potential in terms of CO₂ per kg (kg-CO₂/kg),

-8-

integration time horizon (ITH): 100 years

GWP_{AE} : Additional GWP caused by release during production etc.
(including those caused by leakage of by-products etc., and
indirect release)

5 N: operation period of the system (year) $N = 12$

M: amount (kg) charged into the system $M = 1.3$

α : recovery rate during the disposal of the system (recovery
amount/charge amount) $\alpha = 0.6$

E: annual power consumption of the system (kWh/year)

10 β : CO_2 emissions necessary for 1kWh power generation (kg-
 CO_2/kWh) $\beta = 0.378$

The results are shown in Table 1.

Table 1

	Example					Comparative Example				
	1	2	3	4	5	1	2	3	4	5
HFC32 mass%	10.6	14.3	18.0	21.8	25.5	5.0	6.9	29.2	36.7	44.1
HFO1234yf mass%	89.4	85.7	82.0	78.2	74.5	95.0	93.1	70.8	63.3	55.9
GWP (ITH=100yr)	75.1	100.0	124.8	150.3	175.1	37.6	50.3	199.9	250.3	299.9
Annual Power Consumption (kWh)	1618	1613	1610	1609	1610	1661	1643	1611	1613	1612
APF	4.954	4.968	4.980	4.982	4.978	4.827	4.878	4.977	4.969	4.971
Indirect Impact kg- CO_2	7339	7317	7301	7298	7303	7533	7454	7306	7316	7314
Direct Impact kg- CO_2	69.0	81.9	94.8	108.0	121.0	49.4	56.1	133.9	160.0	185.9
LCCP kg- CO_2	7408	7399	7396	7406	7424	7582	7510	7440	7476	7500

15 Test Example 2

Using the same refrigerants as in Test Example 1, the
heat pump apparatus was operated under the same conditions as in
Test Example 1, except for using a refrigerant pipe 5 m in length.
The results are shown in Table 2 (Examples 6 to 10, Comparative
20 Examples 6 to 10).

Table 2

	Example					Comparative Example				
	6	7	8	9	10	6	7	8	9	10
HFC32 mass%	10.6	14.3	18.0	21.8	25.5	5.0	6.9	29.2	36.7	44.1
HFO1234yf mass%	89.4	85.7	82.0	78.2	74.5	95.0	93.1	70.8	63.3	55.9
GWP (ITH=100yr)	75.1	100.0	124.8	150.3	175.1	37.6	50.3	199.9	250.3	299.9
Annual Power Consumption (kWh)	1605	1600	1598	1599	1602	1633	1620	1603	1607	1607
APF	4.994	5.011	5.016	5.013	5.005	4.909	4.947	5.000	4.989	4.987
Indirect Impact kg-CO ₂	7281	7255	7249	7253	7264	7406	7350	7271	7288	7290
Direct Impact kg-CO ₂	69.0	81.9	94.8	108.0	121.0	49.4	56.1	133.9	160.0	185.9
LCCP kg-CO ₂	7350	7337	7344	7361	7385	7456	7406	7405	7449	7476

Test Example 3

- Using the same refrigerants as in Test Example 1,
- 5 the heat pump apparatus was operated under the same conditions as in Test Example 1, except for using a refrigerant pipe 10 m in length. The results are shown in Table 3 (Examples 11 to 15, Comparative Examples 11 to 15).

10 Table 3

	Example					Comparative Example				
	11	12	13	14	15	11	12	13	14	15
HFC32 mass%	10.6	14.3	18.0	21.8	25.5	5.0	6.9	29.2	36.7	44.1
HFO1234yf mass%	89.4	85.7	82.0	78.2	74.5	95.0	93.1	70.8	63.3	55.9
GWP (ITH=100yr)	75.1	100.0	124.8	150.3	175.1	37.6	50.3	199.9	250.3	299.9
Annual Power Consumption (kWh)	1643	1629	1622	1619	1619	1727	1676	1619	1620	1618
APF	4.879	4.921	4.941	4.950	4.978	4.641	4.786	4.952	4.949	4.955
Indirect Impact kg-CO ₂	7453	7389	7359	7346	7345	7834	7601	7343	7346	7338
Direct Impact kg-CO ₂	69.0	81.9	94.8	108.0	121.0	49.4	56.1	133.9	160.0	185.9
LCCP kg-CO ₂	7522	7471	7454	7454	7466	7884	7657	7477	7506	7524

Test Example 4

- As Comparative Examples, the heat pump apparatus with the same refrigerant pipe length was operated under the same
- 15 conditions as in Test Example 1, using R410A (Comparative Example

-10-

16) and R407C (Comparative Example 17) as refrigerants. The results are shown in Table 4.

Table 4

	Comparative Example	
	16	17
Refrigerant	R410A	R407C
GWP (ITP=100yr)	2060	1774
Annual Power Consumption (kWh)	1606	1595
APF	4.992	5.026
Indirect Impact kg-CO ₂	7283	7235
Direct Impact kg-CO ₂	1101	952
LCCP kg-CO ₂	8384	8187

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Fig. 1 shows LCCP in the refrigerant compositions of the present invention, based on the results of Tables 1 to 3. The results reveal that, regarding LCCP, i.e., an index reflecting direct and indirect impacts of carbon dioxide gas, the refrigerant of the present invention has a lower LCCP level than R410A and R407C, which indicates that the refrigerant composition having 10 to 26 mass% (particularly, 14 to 22 mass%) of HFC32 has a low environmental impact.

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15 Industrial Applicability

The mixed refrigerant composition of the present invention is effective as a refrigerant composition for refrigeration and air-conditioning systems.

-11-

CLAIMS

[Claim 1] A refrigerant composition comprising 10 to 26 mass% of difluoromethane (HFC32) and 74 to 90 mass% of 2,3,3,3-tetrafluoropropene (HFO1234yf).

[Claim 2] The refrigerant composition according to claim 1, comprising 14 to 22 mass% of difluoromethane (HFC32) and 78 to 86 mass% of 2,3,3,3-tetrafluoropropene (HFO1234yf).

10

[Claim 3] The refrigerant composition according to claim 1, which further comprises a polymerization inhibitor.

[Claim 4] The refrigerant composition according to claim 1, which further comprises a drying agent.

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[Claim 5] The refrigerant composition according to claim 1, which further comprises a stabilizer.

[Claim 6] A method of operating a refrigerator comprising circulating the refrigerant composition according to claim 1 through a compressor.

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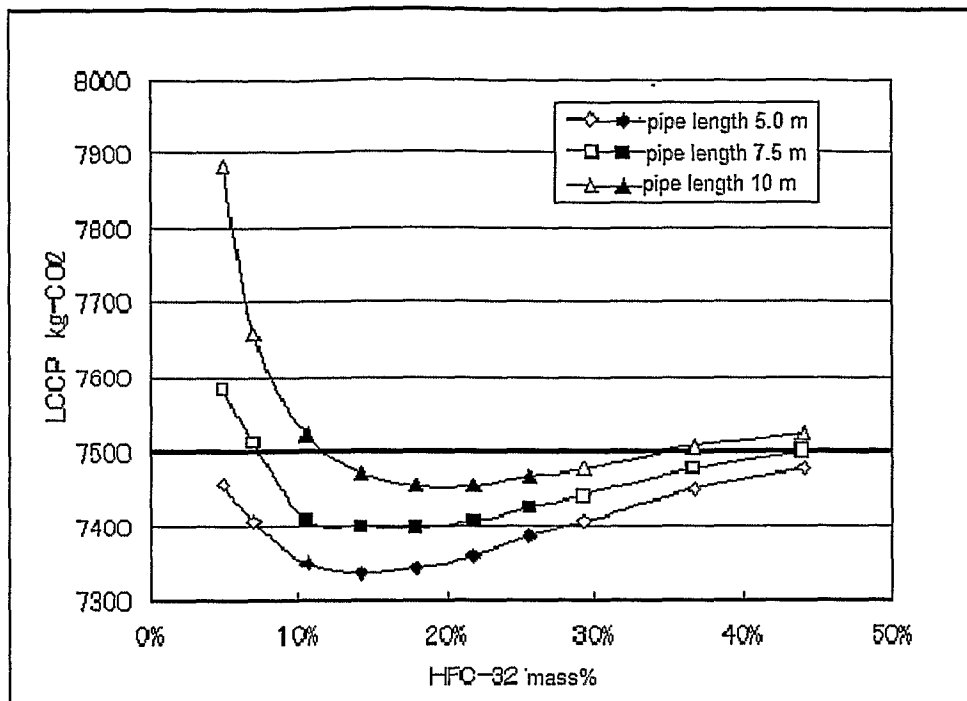
[Claim 7] A method of producing the refrigerant composition according to claim 1, comprising mixing 10 to 26 mass% of difluoromethane (HFC32) with 74 to 90 mass% of 2,3,3,3-tetrafluoropropene (HFO1234yf).

25

[Claim 8] A refrigerator comprising the refrigerant composition according to claim 1.

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Fig. 1



INTERNATIONAL SEARCH REPORT

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A. CLASSIFICATION OF SUBJECT MATTER
INV. C09K5/04

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

B. FIELDS SEARCHEDMinimum documentation searched (classification system followed by classification symbols)
C09K

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, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2006/094303 A (DU PONT [US]; MINOR BARBARA HAVILAND [US]; RAO VELLIYUR NOTT MALLIKARJ) 8 September 2006 (2006-09-08) cited in the application * page 2, lines 6-10 and lines 29-35, page 14, lines 9-14, Table 2, page 15, 22nd entry, Table 3, page 21, first entry, Table 5, page 25, first entry, Table 9, page 61, HFC-1234yf/HFC-32 compositions, claims * ----- -/--	1-8

☒ Further documents are listed in the continuation of Box C.☒ See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search

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International application No

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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