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(54) Title: POLYOLEFINS AS FRAGRANCE DELIVERY VEHICLES

(57) Abstract: A fragrance release composition comprising: (a) a polyolefin comprising polymerized units of ethylene and at least one C_4-C_{12} alkene; and (b) a fragrance.

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POLYOLEFINS AS FRAGRANCE DELIVERY VEHICLES

This invention relates to a method for producing a vehicle for delivering fragrances and controlling their release.

Use of polymers as vehicles for fragrance delivery is known. For example, CN102504385A discloses polymer resins used for this purpose, including ethylene-octene copolymer. However, this reference does not disclose the composition described herein.

The problem solved by this invention is the need for improved vehicles for delivery of fragrances.

STATEMENT OF INVENTION

The present invention provides a fragrance release composition; said composition comprising: (a) a polyolefin comprising 40 to 85 wt% polymerized units of ethylene and 15 to 60 wt% polymerized units at least one C_4-C_{12} alkene; and (b) a fragrance.

DETAILED DESCRIPTION

Percentages are weight percentages (wt%) and temperatures are in °C, unless specified otherwise. Operations were performed at room temperature (20-25°C), unless specified otherwise. An "alkene" is an unsaturated aliphatic hydrocarbon. Preferably, alkenes have only one double bond. Alkenes may be linear or branched, preferably linear.

A "fragrance" includes any hydrophobic component which provides a pleasant scent. Examples include scents that are floral, ambery, woody, leather, chypre, fougère, musk, vanilla, fruit, and/or citrus. Fragrance oils are obtained by extraction of natural substances or synthetically produced. Fragrances produced may be simple (one essence) or complex (a mélange of essences). Often, the fragrance oils are accompanied by auxiliary materials, such as fixatives, extenders, stabilizers and solvents.

Preferably, the polyolefin comprises polymerized units of ethylene and at least one C_6-C_{10} alkene, preferably a C_8 alkene, preferably 1-octene. The polyolefin comprises 40 to 85 wt% polymerized units of ethylene and 15 to 60 wt% polymerized units of the alkene; preferably at least 20 wt% alkene, preferably at least 25 wt% alkene, preferably at least 30 wt% alkene; preferably no more than 55 wt% alkene, preferably no more than 50 wt% alkene; preferably at least 45wt% ethylene, preferably at least 50 wt% ethylene, preferably at least 55 wt% ethylene, preferably at least 60 wt% ethylene, preferably at least 65 wt% ethylene, preferably at least 70 wt% ethylene; preferably no more than 80 wt% ethylene, preferably no more than 75 wt% ethylene, preferably no more than 70 wt% ethylene.

Preferably, the polyolefin has a density (g/cm^3) from 0.80 to 0.92, a Melt Index (g/10 min as measured at 2.16 kg @ 190 °C) from 0.3 to 35 and a DSC Melting Peak (°C, Rate 10 °C/min) from 30 to 100. Preferably, density is at least 0.85; preferably no more than 0.91, preferably no more than 0.90, preferably no more than 0.89, preferably no more than 0.88. Preferably, Melt Index is at least 0.5, preferably at least 0.7; preferably no more than 30, preferably no more than 15, preferably no more than 10, preferable no more than 5, preferably no more than 3, preferably no more than 2. Preferably, the DSC Melting Peak is no more than 90, preferably no more than 85, preferably no more than 75; preferably at least 35.

Preferably, the weight-average molecular weight (M_w) of the polyolefin is from 35,000 to 200,000; preferably at least 40,000, preferably at least 70,000, preferably at least 80,000, preferably at least 90,000, preferably at least 100,000; preferably no more than 170,000, preferably no more than 150,000, preferably no more than 130,000.

The polyolefin may be a random copolymer or a block copolymer. Preferably, the polyolefin is a random copolymer.

Preferably, the fragrance may be delivered to the polyolefin beads using glycol ethers and/or surfactants. Surfactants and glycol ethers may be used to solubilize the fragrance compositions to enhance the delivery to the polyolefin bead. Preferably, glycol ethers useful have the following composition:

- $R—(OC_nH_{2n}O_z)OX$

wherein R is a substituted or unsubstituted C_1-C_{12} aliphatic group, a substituted or unsubstituted C_6-C_{12} aryl group, a group of the formula —C(=O)C_6H_5, or a group of the formula —C(=O)CH_3, n is 2 to 4, z is 1 to 4, and X is —H, —CH_3, —C(=O)CH_3, or —C(=O)C_6H_5. Preferably, R is a substituted or unsubstituted C_1-C_{10} aliphatic group, preferably an unsubstituted C_2-C_{10} alkyl group, more specifically an unsubstituted C_2-C_6 alkyl group. In a preferred embodiment, n is 2 to 4, z is 1 to 3, and X is —H. Representative examples of the glycol ether compounds include tripropylene glycol methyl ether, dipropylene glycol n-butyl ether, tripropylene glycol n-butyl ether, dipropylene glycol n-propyl ether, dipropylene glycol phenyl ether, dipropylene glycol methyl ether acetate, propylene glycol n-propyl ether, diethylene glycol monobutyl ether, diethylene glycol n-butyl ether, diethylene glycol monohexyl ether, diethylene glycol hexyl ether, or a combination thereof. Other examples of the second compound may include dipropylene glycol methyl ether, propylene glycol methyl ether, propylene glycol methyl ether acetate, dipropylene glycol methyl ether acetate, or propylene glycol diacetate

The surfactants may be a nonionic, cationic, or anionic material, and it may be a blend of surfactants. Non-limiting examples of surfactants known in the art that may suitably be used include those described in U.S. Pre-Grant publication 2002/0045559. Combinations of surfactants and glycol ethers may be used to enhance the delivery of the fragrance compositions to the polyolefin bead.

Preferably, the amount of fragrance in the fragrance release composition is from 9 to 50 wt% based on the total weight of the composition, preferably at least 10 wt%, preferably at least 12 wt%, preferably at least 15 wt%, preferably at least 20 wt%; preferably no more than 40 wt%, preferably no more than 30 wt%.

Examples

Fragrances:

Fragrance 1 (F1): Orange Oil is a product of Sigma Aldrich Corporation, St Louis, Missouri, and U.S.A. CAS # 8008-57-9

Fragrance 2 (F2): Tropical Breeze is a product of Givaudan Flavor Corporation, East Hanover, NJ, U.S.A.

Table 1: Properties Of Polymers

Material Description	No.	% alkene in ethylene-alkene copolymer, wt%	Approx. M_w*	Dens., g/cm^3 ASTM D792	Melt Index (dg/ min, 190C)	DSC Melting Peak, °C Rate 10°C / min(4)	Tg, °C DSC inflec-tion point(4)
Polyolefin Elastomers	P1	45	115K	0.857	1	38	-58
Ethylene 1-Octene Grades	P2	38	115K	0.87	1	60	-52
	P7	28	115K	0.885	1	77	-46
	P3	38	45K	0.87	30	65	-54
	P8	18	45K	0.902	30	96	-36
	P9	28	45K	0.885	30	80	-47
	P10	42	140K	0.863	0.5	47	-55
ELITE™ Enhanced Polyethylene Resins	P11	NA		0.964	0.85	134	
ATTANE™ Ultra Low Density PE Resin	P12	NA		0.906	8.0	124	
AFFINITY™ Polyolefin Plastomers and Polyolefin Elastomers	P4	35	100K	0.875	3	68	
(ethylene-octene)	P5	38	10K	0.87	1000 *	68	
	P13	18	50K	0.902	7.5	98	
	P14	12	115K	0.909	1	106	
	P6	38	15K	0.874	500 *	70	
PRIMACOR™ (20.5% Acrylic Acid)	P15	NA	7.5K	0.958	300	77	
(9.7% Acrylic Acid)	P16	NA	65K	0.938	10	98	
(20% Acrylic Acid)	P17	NA	5.5K	0.955	1300	75	
DOW LDPE	P18	NA	35K	0.923	55	110	
AMPLIFY™ Ethylene-EA copolymer	P19	NA	110K	0.932	1.3	99	
Aldrich PE Maleic Anhydride	P20	NA					
Aldrich Ethylene-VA	P21	NA					
ELVAX™, Ethylene-VA (32% VA) 150	P22	NA	32K	0.957	43	63	
ELVAX™, Ethylene-VA (28% VA) 210W	P23	NA	6K	0.951	400	65	
ELVAX™, Ethylene-VA (28% VA) 220W	P24	NA	15K	0.951	150	70	
ELVAX™, Ethylene-VA (28% VA)240W	P25	NA	32K	0.951	43	74	
ELVAX™, Ethylene-VA (18% VA)420	P26	NA	15K	0.937	150	73	
ELVAX™, Ethylene-VA (18% VA)450	P27	NA	52K	0.941	8	86	

VA=vinyl acetate; EA=ethyl acrylate; PE=polyethylene

* MI estimates based on viscosity

ENGAGE™ Polyolefin Elastomers;"ELITE™ Enhanced Polyethylene Resins"; "ATTANE™ Ultra Low Density Polymers"; "AMPLIFY™ functional polymers " and AFFINITY™ Polyolefin Plastomers are products of the Dow Chemical Company, Midland, Michigan, U.S.A. "Polyolefin Elastomers-ethylene 1-octene grades" where obtained from Aldrich products from Sigma-Aldrich Corporation". "PRIMACOR™ copolymers" are products of SK Global Chemical Co. LTD., Seoul, Korea; "ELVAX™ copolymer resins are products from DuPont Company, Wilmington, Delaware, U.S.A.

Melt Index measured at (2.16 kg @ 190°C) ASTM D1238

K=1,000, i.e., "115K"=115,000 g/mole

Table 2: Bead Integrity In Presence Of Fragrance Oil

Each vial has 1 gram of beads. Fragrances; 0.1 mls= 100 µl pipetted equivalent to Dosage wt% of 9.1; 0.5 mls= 500 µl pipetted equivalent to Dosage wt% 33.3; 1.0 mls= 1000 µl pipetted equivalent to Dosage wt% 50 in Table 2 below.

	Performance Index	F1	F1	F1	F2	F2	F2	Bead integrity after fragrance
Dosage wt%		9.1	33.3	50	9.1	33.3	50	
P1	**4.43**	5	5	5	5	4	2	5
P2	**4.43**	5	5	5	5	4	2	5
P7	3.57	5	5	4	3	2	1	5
P3	3.86	5	5	5	5	3	1	3
P8	3.29	5	3	1	5	3	1	5
P9	**4**	5	5	4	5	3	1	5
P10	3.43	5	3	3	5	4	1	3
P11	3	5	3	1	5	1	1	5
P12	3	5	4	2	5	3	1	1
P4	**4**	5	5	5	5	4	1	3
P5	**4.14**	5	5	4	5	4	3	3
P13	3.29	5	3	1	5	3	1	5

	Performance Index	F1	F1	F1	F2	F2	F2	Bead integrity after fragrance
P14	2.71	5	3	3	4	3	1	0
P6	3.14	5	5	5	3	3	1	0
P15	3.14	5	4	2	4	3	1	3
P16	2.86	5	4	1	4	2	1	3
P17	3.43	5	5	2	5	3	1	3
P18	3	5	3	1	3	3	1	5
P19	3.14	5	4	2	4	3	1	3
P20	3	5	3	1	5	3	1	3
P21	3.86	5	5	5	3	3	3	3
P22	3.71	5	4	4	5	4	1	3
P23	3.86	5	4	4	5	3	3	3
P24	3.86	5	4	4	5	3	1	5
P25	3.86	5	4	4	5	3	1	5
P26	3.86	5	5	4	4	3	1	5
P27	3.71	5	5	4	5	1	1	5

Performance Rating Assessment

Fragrance adsorbed	Assessment Rating
M= most >75% no fluid fragrance observed	**5**
P= partial 25-75% slight/minor fragrance fluid observed	**3**
N= none < 25% most fragrance fluid observed and not adsorbed	**1**

Integrity	
OK = beads appear unchanged in shape or size	**5**
SW= Swollen beads	**3**
D= Dissolved	**1**
A= Agglomerated or sticking together	**0**

Performance Index is average of Numerical Assessment Ratings for Fragrance Adsorbed and Integrity.

The assessment ratings were based on qualitative determinations by skilled laboratory personnel.

The substrates chosen to measure fragrance release and fragrance longevity were chosen based on Performance Index ratings >4.

Table 3 Formulations Of Polyolefin Beads With Fragrance 1

Composition	Polyolefin	Fragrance
1	1 gram P1	1 gram F1
2	1 gram P2	1 gram F1
3	1 gram P3	1 gram F1
4	1 gram P4	1 gram F1
5	1 gram P5	1 gram F1
6	1 gram P6	1 gram F1

Methodology for preparation of polyolefin beads for GC/MS and Headspace analysis.

1 gram of each type of Polyolefin were placed into a 1 ounce vial and weighed. An equivalent weight of fragrance (1 gram), in this case Orange oil (limonene) was added to each vial, so typically the fragrance oil was adsorbed, and the POE beads adsorbed between 46-53% fragrance oil based on weight.

Two vials of each polyolefin was used for the GC/MS and 2 vials were used for the headspace analysis, resulting in 4 vials, each containing 1 gram for every incremental time analysis to be run. The average values in the table were for the 4 vials per time increment, and represent the average amount of fragrance adsorbed per vial and include the standard deviation. So for the initial evaluation there were 4 sets of 1 gram beads/fragrance, at 7 days there were 4 sets of 5 beads/fragrance, at 14 days there were 4 sets of 1 gram beads/fragrance, etc. There were a total of 6 Polyolefin beads examined and were weighed out for: measurements occurring at Initial, 7 days, 14 days, 21 Days, and 28 days.

Methodology preparation and analysis for fragrance using GC-MS (Table 4).

A 1% standard mix of the fragrance was prepared in toluene.

The standard mix was diluted in toluene to make the following concentrations:

10,000 and1000, 500, 100, 10, and 1ppm.

Each standard were injected into a microvial in a TDU (thermal desorption tube), directly into the TDU.

A calibration curve was made for the fragrance.

Sample size: 5 grams.

Analysis by headspace GC-MS:

33°C for 0.5 min of heating prior to introduction into GC-MS (to understand VOCs that might contribute to odor.

Column: DB-Wax (30m x 0.25mm x 0.50µm).

Column: Rtx-5MS (30m x 0.25mm x 0.25µm). Units are ppm, vol/vol.

Table 4

Headspace GC-MS Analysis Average of two runs

Parts per million of fragrance detected in the headspace

	T = 0	T = 7 days	T = 14 days	T = 21 days	T = 28 days
Comparative 6	1335	1581	1227.5	1049.5	521
Example 4	1289	199.5	1200	1018	**692**
Comparative 5	1411	1385	1339.5	670	248.5
Example 1	1253.5	1194	1180	882	**776**
Example 3	1317	1287	1175.5	972	**779.5**
Example 2	1162.5	1287	1194	1022.5	**787**

T=Time

Lowest density (least crystalline) and lowest melt peak as well as most elastic Polyolefin P1 bead demonstrated best performance regarding fragrance release.

Examples 1-4 show higher fragrance release at 28 days (parts per million fragrance measured in headspace between 692 – 779.5 ppm) compared to the Comparative 5 and Comparative 6 (>600 ppm).

Example 1 (polymer P1) showed the best fragrance longevity using Headspace GC and Extraction performed in Hexanes and averaged over 5 beads/per measurement in duplicate.

Methodology for Evaluation of Fragrance for BULK Analysis (Table 5).

- The bulk levels of the fragrance was also tested at T = 0 and T = 56 Days by HS-GC-MS.
- Bulk samples were prepared by dissolving each bead 20-fold in Hexanes.
- The headspace and bulk levels of the fragrance was quantitated using a calibration curve of fragrance in Hexanes (1 – 1000 ppm).

BULK Data (beads dissolved and extracting fragrance from bead)

(Parts per million in substrate), Concentration, wt/wt

Table 5

Bead type		T = 0	T = 7 Days	T = 14 Days	T = 21 Days	T = 28 Days	T = 48 Days	T = 56 Days
P6	Comparative 6	55.4	50.2	33.7	23.7	17.9	1.3	0.1
P4	Example 4	56.9	46.7	34.7	25.4	13.2	6.5	0.7
P5	Comparative 5	50.8	42.9	32.6	22.6	10.7	3.3	0.3
P1	Example 1	51.3	47.5	41.6	33.2	18.4	13.2	4.8
P3	Example 3	51.2	49.6	35.0	31.0	18.0	2.0	0.4
P2	Example 2	52.5	46.8	38.1	26.6	20.4	4.8	2.4

Example 1, P1 had slower fragrance release comparatively, therefore longer fragrance longevity with bead integrity.

Information in Table 5 demonstrates fragrance retained in beads….and then the release from the beads. Example 1 based on P1 allows fragrance (Limonene) to last up to 56 days.

Examples 1 - 4 show higher fragrance retained at 56 days (parts per million fragrance measured in bead) compared to the Comparative 5 and Comparative 6.

CLAIMS

1. A composition comprising: (a) a polyolefin comprising 40 to 85 wt% polymerized units of ethylene and 15 to 60 wt% polymerized units of at least one C_4-C_{12} alkene; and (b) a fragrance.

2. The composition of claim 1 in which the polyolefin comprises 15 to 60 wt% polymerized units of a C_6-C_{10} alkene.

3. The composition of claim 2 in which the polyolefin comprises 40 to 80 wt% polymerized units of ethylene and 20 to 60 wt% polymerized units of at least one C_6-C_{10} alkene.

4. The composition of claim 3 in which the polyolefin has M_w from 35,000 to 170,000.

5. The composition of claim 4 in which the polyolefin comprises 20 to 60 wt% polymerized units of 1-octene.

6. The composition of claim 1 in which the polyolefin has a melt index from 0.3 to 35.

7. The composition of claim 6 in which the polyolefin has a density from 0.8 to 0.91 g/cm^3.

8. The composition of claim 7 in which the polyolefin comprises 40 to 80 wt% polymerized units of ethylene and 20 to 60 wt% polymerized units of at least one C_6-C_{10} alkene.

9. The composition of claim 8 in which the polyolefin comprises 20 to 60 wt% polymerized units of 1-octene.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2018/051423

A. CLASSIFICATION OF SUBJECT MATTER
INV. A61K8/81 A61Q13/00
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)
A61K A61Q

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, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

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Date of the actual completion of the international search	Date of mailing of the international search report
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Name and mailing address of the ISA/	Authorized officer
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Form PCT/ISA/210 (second sheet) (April 2005)

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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INTERNATIONAL SEARCH REPORT

Information on patent family members

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