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CHEMILUMINESCENT AEROSOLS

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U.S. Cl. 252—188.3 10 Claims

ABSTRACT OF THE DISCLOSURE

Described and claimed are chemiluminescent aerosols containing tetrakis(disubstitutedamino)ethylenes in suitable propellants, e.g., tetrakis(dimethylamino)ethylene in a fluorinated hydrocarbon.

Related applications

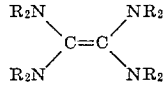
This application is a continuation-in-part of my copending application Ser. No. 389,808, filed Aug. 14, 1964, and now abandoned. Ser. No. 389,808 is a division of my application Ser. No. 174,404, filed Feb. 20, 1962 (now U.S. Patent 3,239,519), itself a continuation-in-part of my copending application Ser. No. 91,589, filed Feb. 27, 1961, and now abandoned which in turn was a continuation-in-part of my copending application Ser. No. 836,062, filed Aug. 26, 1959, now abandoned in favor of the continuation-in-part application thereof, Ser. No. 91,590, filed Feb. 27, 1961 (now U.S. Patent 3,239,518).

FIELD OF THE INVENTION

This invention relates to, and has as its principal object provision of, chemiluminescent aerosols based on tetrakis(disubstitutedamino)ethylenes, i.e., pertertiaryaminoethylenes.

DESCRIPTION OF THE INVENTION

Pruett et al., J. Am. Chem. Soc. 72, 3646 (1950) prepared the first member of the series of the tetrakis(disubstitutedamino)ethylenes, i.e., tetrakis(dimethylamino) ethylene (TMAE). U.S. Patent 3,239,519 describes and claims other related compounds to complete the series of compounds



in which the R's, which can be alike or different, are straight or branched chain alkyl or cyloalkyl hydrocarbon radicals of from 1–10 carbons each, preferably of no more than five carbons each, which can be joined pairwise on one nitrogen to form 3–5 membered monoaza heterocycles and on two nitrogens to form 4–7 membered diaza heterocycles. See also U.S. Patent 3,239,534 for a preparation of all of these compounds.

In accordance with the present invention, it has been found that useful chemiluminescent areosols can be readily made from the above-identified series of compounds by combining them in finely divided form with appropriate propellants. The areosol or aerosol-generating mixtures per se consist essentially of at least one nonquenching propellant, e.g., 10–98% by weight, and an effective chemiluminescing amount, e.g., 90–2%, of at least one of the compounds named above, but inert materials which do not interfere with the function of the propellant or quench the luminescent material can also be present.

The term "aerosols" is believed to be well known to the packaging art. The first and probably most scientific usage thereof occurred in early colloidal chemistry—see, for instance, Whitlaw-Gray and Patterson, "Smoke," Arnold and Company, Limited, London (1932), as defining a colloidal system consisting of very finely subdivided liquid or solid particles dispersed in and surrounded by a gas. More recently, Sinclair in "Handbook on Aerosols" (1950), Washington, D.C., on page 64, stated that such particles should be smaller in diameter than 50 microns and usually less than 10 microns. In the early 1940's, this term was used to describe insecticides packed in a self-pressurized pack and the Chemical Specialties Manufacturers Association, Incorporated, of New York evolved the following definition of an aerosol product:

"A self-contained sprayable product in which the propellant force is supplied by a liquefied gas. Includes space, residual, surface coating, foam and various other types of product but does not include gas-pressurized products such as whipping cream. The term aerosol as used here is not confined to the scientific definition."

The best modern definition is believed to be as follows:

"A much more satisfactory term for the subject described in this book is 'pressurized pack' which may be defined as a self-contained pack which contains the product and the propellant necessary for the expulsion of the former. This definition includes packs which utilize compressed gases as propellants and, by discarding the term 'aerosol', acknowledgement is made of the fact that this word has a scientific meaning."

These foregoing two quotations are taken from "Pressurized Packaging (Aerosols)" by Herzka and Pickthall, Academic Press, New York (1958).

Recognizing that some confusion does exist and that the last quoted definition is the most recent, the term 'aerosols' is used here as covering generically all pressurized packs including those where the propellant is a pressurized gas, either supplied in situ from another state form thereof, or where the pressurized gas is charged as such directly. In any event, however, and regardless of the specific definition used, aerosols are regarded in this application primarily as composition-of-matter, i.e., mixtures, containing the propellant and an effective chemiluminescent amount, i.e., down to about 2% by weight, of the chemiluminescent material and any inert substance desired. Both the propellant and any inert substance should, of course, be nonquenching to the chemiluminescence: cf. my U.S. Patent 3,264,221.

The possible variables in the formulation of these chemiluminescent aerosols as to propellants, dispenser components, filling techniques, laboratory evaluation, formulations, and the like, are the same as those known already in the general aerosol art, all as discussed in detail in the above definitive text by Herzka and Pickthall. The only variation in the present compositions and techniques over those discussed in this publication is the necessary presence of one or more of the chemiluminescent tetrakis (disubstitutedamino)ethylenes which serve to render the thus formulated aerosols chemiluminescent. By reference to this publication all disclosures and discussions therein are made common to this specification.

In the foregoing and elsewhere, reference is made to "chemiluminescent aerosols." It is recognized that this is not always correct since some of the compositions in "aerosol" form do not chemiluminesce per se. However, in all instances when the aerosol containing the chemiluminescent tetrakis(disubstitutedamino)ethylene is deposited on a necessarily ultimate surface, and contact is established with oxygen, air, a peroxide, etc., chemiluminescence will occur. In most cases, chemiluminescence appears upon mere contact of the chemiluminescent material with the oxidizing agent. Compare, with respect to oxidizing agents, my above-mentioned U.S. Patent 3,264,221.

The propellent gases used in the present invention are conventional aerosol propellants but must contain no oxidizing oxygen, be essentially insoluble in the composition

to be propelled from the container, and be inert, or non-quenching, toward the above chemiluminescent pertertiaryaminoethylenes. Representative operable propellent gases are chlorodifluoromethane (Du Pont Freon®–22), dichlorofluoromethane (Freon®–21), dichlorodifluoromethane (Freon®–12), 1,2 - dichlorotetrafluoromethane (Freon®–114), octafluorocyclobutane (Freon® C318), chloropentafluoroethane (Freon® 115) and mixtures such as "F–12" and trichlorofluoromethane (Freon® 11), "F–12" and "F–22," "F–12" and "F–114," "F–22" and "F–11," and "F–22" and "F–114" (cf. Wahlin U.S. Patent 3,062,751). Inert compressed gases like nitrogen and helium may be used but they are less advantageous than the above-mentioned fluorine-containing compounds and their mixtures.

It may be stated that aerosols in which the chemiluminescent component is tetrakis(dimethylamino)ethylene are essentially nonoxyluminescent in vapor form when the propellant(s) is one of the polyfluorocarbon types but are oxyluminescent when using other propellants such as nitrogen (seen Example 12). Aerosols in which the oxyluminescent pertertiaryaminoethylene component is of the tetraalkyl-$\Delta^{2,2'}$-bi(imidazolidine) structure, even with the polyfluorcarbon propellants, give oxyluminescent clouds which continue their oxyluminescence when deposited on a surface (see, for instance, Examples 13, 14, 15, 16, 18, 19, 22, 24, 25, and 26).

Other useful low-boiling materials can be used as the propellant in the aerosols of the present invention. As is true of all such, the primary criteria involved in adjudging whether or not a material is useful as a propellant hinge primarily on its boiling point and vapor pressure, and partial pressure in the charged systems in the aerosol containers physically speaking. Chemically speaking, the only overriding criterion is that the material for the propellant must be unreactive with the chemiluminescent peraminoethylene serving as the active ingredient in the overall aerosol formulation. Accordingly, in addition to the foregoing described polyfluorohydrocarbons, other compounds can be used as propellants such as, for instance, the low-boiling hydrocarbon ethers, e.g., dimethyl ether, and to a lesser extent, methyl ethyl ether; the low-boiling saturated aliphatic hydrocarbons such as, for instance, propane, isobutane, butane, and the like.

If desired, and as possibly necessary for viscosity control, inert solvents or diluents can also be present in the aerosol charge. Such diluents must, of course, be unreactive chemically with most importantly, the chemiluminescent pertertiaryaminoethylene and also with the propellant(s). Suitable specific examples of such possibly useful diluents are the liquid hydrocarbons such as cyclohexane, methyl and ethyl cyclohexanes, and the like, the higher aliphatic hydrocarbons such as decane, Nujol [1], purified kerosene, mineral oils, and the like, as well as the lower-boiling hydrocarbons such as, for instance, cyclopentane, cyclobutane, and for that matter, cyclopropane, and the like. Also useful are the higher boiling species of the previously described propellants such as, for instance, the hydrocarbon ethers, e.g., diethyl, dipropyl, dibutyl ether, and the like. Silica or other inert powders can also be present to give powder aerosols.

The following examples in which the parts given are by weight are submitted to further illustrate the present invention but not to limit it. All tetrakis(disubstituted-amino)ethylenes disclosed except TMAE itself are within the scope of my U.S. Patent 3,239,519.

EXAMPLES 1–9

Aerosols containing tetrakis(dimethylamino)ethylene (TMAE) were prepared by charging TMAE into standard aerosol containers under nitrogen. Aerosol valves were then attached to the containers and the propellants were

[1] Nujol is medicinal grade liquid petrolatum.

injected into the containers through the valves. The following table lists the compositions in parts by weight.

TMAE	Dichlorodifluoromethane/trichlorofluoromethane, 20/80	Dichlorodifluoromethane/trichlorofluoromethane, 30/70	Dichlorodifluoromethane/trichlorofluoromethane, 50/50	Dichlorodifluoromethane
2	98			
10	90			
2		98		
10		90		
20			80	
20				80
30				70
50				50
65				35

Objects in contact with air wet with the spray from these aerosols were rendered visible in a darkened room by the blue-green chemiluminescence of the TMAE deposited thereon. Aerosols containing increasing amounts of TMAE gave brighter sprays because of increased amounts of deposition of the chemiluminescent ingredient. The aerosol comprising 20 parts of TMAE and 80 parts of dichlorodifluoromethane contained in a metal can showed no deterioration after three years' storage, although the composition was unstable to storage in transparent glass containers.

EXAMPLE 10

An aerosol was prepared comprising 15 parts of TMAE, 32 parts of perfluorocyclobutane, and 18 parts of dichlorodifluoromethane. The aerosol gave a cloud which chemiluminesced on settling on a surface contacting the air.

EXAMPLE 11

An aerosol was prepared comprising 15 parts of TMAE and 20 parts of isobutane. The aerosol gave a cloud which chemiluminesced for a considerable time after settling on a surface in contact with air.

EXAMPLE 12

TMAE was sprayed from a paint sprayer under nitrogen pressure to give a cloud which chemiluminesced on settling on a surface. When the TMAE was shaken with air to obtain a strong luminescence in the liquid before spraying, the cloud was then luminous.

EXAMPLE 13

By the procedure of Example 1, an aerosol was prepared which comprised 10 parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 22 parts of isobutane. When sprayed in air in the dark, the aerosol formed a luminous cloud which was bright and of good duration.

EXAMPLE 14

By the procedure of Example 1, an aerosol was prepared which comprised five parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine), five parts of diethyl ether, and 22 parts of isobutane. When sprayed in air in the dark, the aerosol formed a luminous cloud of good brightness and duration.

EXAMPLE 15

By the procedure of Example 1, an aerosol was prepared which comprised 4.3 parts of 1,1',3,3'-tetramethyl-$\Delta^{2,2'}$-bi(imidazolidine), five parts of cyclohexane, and 22 parts of isobutane. When sprayed in air in the dark, the aerosol formed a luminous cloud of good brightness and duration.

EXAMPLE 16

By the procedure of Example 1, an aerosol was prepared which comprised 10 parts of diethyldimethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 22 parts of isobutane. When sprayed in air in the dark, the aerosol formed a luminous cloud of good brightness and duration.

EXAMPLE 17

Example 13 was repeated but employing 20 parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 17 parts of isobutane. When sprayed in air in the dark, the aerosol formed a luminous cloud which was bright and of good duration.

EXAMPLE 18

By the procedure of Example 1, an aerosol was prepared which comprised 13 parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 44 parts of 1,2-dichlorotetrafluoroethane. When sprayed in air in the dark, the aerosol formed a luminous cloud which was bright and of good duration.

EXAMPLE 19

By the procedure of Example 1, an aerosol was prepared which comprised 15 parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 40 parts of dichlorodifluoromethane. When sprayed in air in the dark, the aerosol formed a luminous cloud which was bright and of good duration.

EXAMPLE 20

By the general procedure of Example 1, the following sprays, powder aerosols and short grease aerosols were prepared using the indicated ingredients in the proportions given. In all cases the resultant aerosol-type products resulted in significant and notably long-range existing chemiluminescence when sprayed in air, particularly when contacting the ultimate substrate.

SPRAYS

(a)

	Percent
TMAE	54.3
Isobutane	45.7

(b)

TMAE	20
Dichlorodifluoromethane	80

POWDER AEROSOLS

	Percent
Silica pigment (commercially available pyrogenic particulate silica of 0.020–0.007 micron particle size range; 99.0–99.7% SiO_2)	3.4
TMAE	5.8
Dichlorodifluoromethane - 1,2-dichlorotetrafluoroethane (40/60 by weight)	90.8

	Percent	
Short grease aerosols	a	b
Silica pigment	1.60	3.6
Decane	9.75	20.0
TMAE	11.35	23.4
Dichlorodifluoromethane	77.30	
Isobutane		53.0

EXAMPLE 21

By the procedure of Example 1, an aerosol was prepared which comprised 45 parts of TMAE and 5 parts of dichlorodifluoromethane. Objects in contact with air wet with the spray from the aerosol were rendered visible in a darkened room by the chemiluminescence of the TMAE deposited thereon. The chemiluminescence was bright and of long duration.

EXAMPLE 22

By the procedure of Example 1, an aerosol was prepared which compared 37.5 parts of TMAE, 12.5 parts of 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine) and 50 parts of dichlorodifluoromethane. When sprayed in air in the dark the aerosol formed a bright luminous cloud. Material deposited on a surface luminesced brightly with good duration.

EXAMPLE 23

By the procedure of Example 1, an aerosol was prepared from 21.5 parts of TMAE, 22.2 parts of Nujol, 0.83 part of decyl alcohol and 50 parts of dichlorodifluoromethane. Material deposited from the aerosol on a surface contacting air chemiluminesced brightly and with good duration.

EXAMPLE 24

Part A.—Preparation of 1,1',3,3'-tetra-n-butyl-$\Delta^{2,2'}$-bi(imidazolidine)

In a glass reactor fitted with a packed distillation column, condensing means, and a distillation takeoff head, a mixture of 23.8 parts of α,α-dimethoxytrimethylamine and 34.5 parts of N,N'-di-n-butylethylenediamine was heated in an oil bath at 114° C. Over a four-hour period the oil bath temperature was gradually increased to 190° C. during which time the dimethylamine and methanol formed in the reaction were removed by distillation. Continued distillation gave 26.5 parts (73% of theory) of 1,1',3,3'-tetra-n-butyl-$\Delta^{2,2'}$-bi(imidazolidine) as a liquid boiling at 125–128° C. under a pressure corresponding to 0.20 mm. of mercury. The product luminesced strongly on exposure to air.

Analysis.—Calc'd for $C_{22}H_{44}N_4$ (percent): C, 72.5; H, 12.2; N, 15.4. Found (percent): C, 72.5; H, 12.4; N, 14.8.

Part B—Aerosol

By the procedure of Example 1, an aerosol was prepared from 20 parts of 1,1',3,3'-tetra-n-butyl-$\Delta^{2,2'}$-bi(imidazolidine), 20 parts of decane containing 4% decyl alcohol by weight and 40 parts of a 30/70 mixture of dichlorodifluoromethane/trichlorofluoromethane by weight. When sprayed in air in the dark, the aerosol formed an orange-colored luminous cloud.

EXAMPLE 25

Part A.—Preparation of 1,1',3,3'-tetra-n-hexyl-$\Delta^{2,2'}$-bi(imidazolidine)

A mixture of 23.8 parts of α,α-dimethoxytrimethylamine and 45.7 parts of di-n-hexylethylenediamine was heated in a reactor as described in Example 24, Part A, with the bath slowly rising over a period of 4 hours from 107 to 200° C. and the methanol and dimethylamine formed during the heating being removed by distillation. Continued distillation gave 37.0 parts (77% of theory) of 1,1',3,3'-tetra-n-hexyl-$\Delta^{2,2'}$-bi(imidazolidine) as a liquid boiling at 185° C. under a pressure corresponding to 0.20 mm. of mercury. The product luminesced strongly on exposure to air.

Analysis.—Calc'd for $C_{30}H_{60}N_4$ (percent): C, 75.6; H, 12.7; N, 11.8. Found (percent). C, 75.7, 75.8; H, 12.4, 12.8; N, 12.1.

Part B.—Aerosol

By the procedure of Example 1, an aerosol was prepared from 17.2 parts of 1,1',3,3'-tetra-n-hexyl-$\Delta^{2,2'}$-bi(imidazolidine), 17.2 parts of decane containing 4% decyl alcohol by weight and 34.4 parts of a 30/70 mixture of dichlorodifluoromethane/trichlorofluoromethane by weight. When sprayed in air in the dark, the aerosol formed an orange-colored luminous cloud.

EXAMPLE 26

By the procedure of Example 1, an aerosol was prepared from 20 parts of tetrakis(N-pyrrolidinyl)ethylene, 20 parts of decane containing 4% decyl alcohol by weight and 40 parts of a 30/70 mixture of dichlorodifluoromethane/trichlorofluoromethane by weight. The aerosol formed a luminous cloud when sprayed in air in the dark.

EXAMPLE 27

Part A.—1-dimethylamino-1,2,2-tris(N-pyrrolidinyl) ethylene

To 126 parts of dimethyl sulfate heated at 65–70° C.

was added with stirring 73 parts (an equimolar proportion) of dimethylformamide over a period of about 30 minutes. The reaction subsequently became exothermic and the reaction temperature was maintained at 65–70° C., as needed, by external cooling of the reactor. After the exothermic reaction had subsided, the reaction mixture was maintained at 65–70° C. for an additional 15 minutes. There was then added dropwise with continued stirring to the reaction mixture at 60–65° C. 71 parts (an equimolar proportion) of pyrrolidine over a period of 1.5 hours. The reaction mixture was maintained at that temperature for an additional 30 minutes and then stripped by distillation under reduced pressure (30 mm. of mercury) to a bath temperature of 90° C. to remove most of the methanol formed in the reaction. The residue was then diluted with about 110 parts of benzene and 54 parts (an equimolar proportion) of sodium methoxide was then added with stirring and moderate external cooling to maintain the reaction temperature below 35° C. The reaction mixture was stirred for five hours, filtered, the solid filter cake washed with benzene, and the resultant filtrate subjected to distillation. There was thus collected 40 ml. of the benzene/methanol azeotrope equivalent to 0.4 molar proportion of methanol boiling at 58° C. On continued distillation there was obtained 87 parts of a crude product boiling over the range 38–79° C./20.0–1.6 mm. of mercury. This liquid was heated in an oil bath for eight hours while gradually raising the bath temperature to 210° C. at atmospheric pressure while collecting all volatile material. There was thus obtained 17 ml. of largely methanol, and on distillation of the resultant liquid residue there was obtained 18.3 parts (20% of theory) of 1-dimethylamino-1,2,2-tris-(N-pyrrolidinyl)ethylene as a light yellow liquid boiling at 104–112° C. (almost all at 109–112° C.)/0.15 mm. of mercury. The dimethylaminotris(N-pyrrolidinyl)ethylene was identified by its IR and n-m-r spectra.

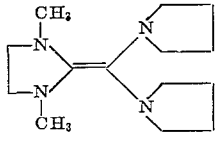
Analysis.—Calc'd for $C_{16}H_{30}N_4$ (percent): C, 69.0; H, 10.9; N, 20.1. Found (percent): C, 69.0; H, 11.2; N, 20.8.

Part B.—Aerosol

By the procedure of Example 1, an aerosol was prepared from 6.1 parts of 1,1,2-tris(N-pyrolidinyl)dimethylaminoethylene, 6.1 parts of decane containing 4% decyl alcohol by weight and 12.2 parts of a 30/70 mixture of dichlorodifluoromethane/trichlorofluoromethane by weight. Material deposited from the aerosol on a surface chemiluminesced in air with moderate duration.

EXAMPLE 28

Part A.—Preparation of bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine



A mixture of 28.5 parts of pyrrolidine, 17.6 parts (0.5 molar proportion) of N,N'-dimethylethylenediamine, and 29.2 parts (an equimolar proportion) of dimethylformamide/dimethyl acetal was heated in an oil bath up to a bath temperature of 100° C. while collecting all distillate. Over a period of four hours there was thus collected 18 ml. of dimethylamine (theory, 26.6 ml.) by condensation in an attached solid carbon dioxide-cooled trap. The bath temperature was gradually raised to 190° C. over a period of four hours, during which time there was collected 29 ml. (theory, 32.4 ml.) of crude methanol. Distillation of the liquid residue afforded 13.4 parts (27% of theory) of crude bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine as a liquid boiling over the range 86–138° C./1.35–0.1 mm. of mercury, largely at 136–138° C., and 15.5 parts (31% of theory) of pure bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine boiling at 138° C./1.35 mm. of mercury. The pure bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine was also characterized by its IR and n-m-r spectra.

Analysis.—Calcd. for $C_{14}H_{26}N_4$ (percent): C, 67.2; H, 10.5; N, 22.4. Found (percent): C, 66.8; H, 10.7; N, 22.3.

Part B.—Aerosol

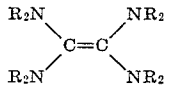
By the procedure of Example 1, an aerosol was prepared from 11.5 parts of bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine, 11.5 parts of decane containing 4% decyl alcohol by weight and 23 parts of a 30/70 mixture of dichlorodifluoromethane/trichlorofluoromethane by weight. Material deposited from the aerosol on a surface contacting air chemiluminesced with short duration.

Since obvious modifications and equivalents in the application will be evident to those skilled in the chemical arts, I propose to be bound solely by the appended claims.

I claim:

1. An aerosol composition consisting essentially of a mixture of:

(1) an effective chemiluminescent amount of at least one chemiluminescent tetrakis(disubstitutedamino) ethylene of the formula



wherein the R's may be alike or different and are selected from the group consisting of monovalent alkyl and cycloalkyl of up to 10 carbons; in the case of two R's joined to the same N, divalent alkylene forming with the N a 3- to 5-membered monoaza heterocycle; and, in the case of two R's joined to different N's, divalent alkylene forming with the two N's a 4- to 7-membered diaza heterocycle; and

(2) at least one compatible inert nonquenching aerosol propellant;

said composition being maintained under pressure in a container.

2. A composition of claim **1** containing, as a chemiluminescent material, tetrakis(dimethylamino)ethylene.

3. A composition of claim **1** containing, as a chemiluminescent material, 1,1',3,3'-tetramethyl-$\Delta^{2,2'}$-bi(imidazolidine).

4. A composition of claim **1** containing, as a chemiluminescent material, 1,1',3,3'-tetraethyl-$\Delta^{2,2'}$-bi(imidazolidine).

5. A composition of claim **1** containing, as a chemiluminescent material, 1,1',3,3'-tetra-n-butyl-$\Delta^{2,2'}$-bi(imidazolidine).

6. A composition of claim **1** containing, as a chemiluminescent material, tetrakis(N-pyrrolidinyl)ethylene.

7. A composition of claim **1** containing, as a chemiluminescent material, 1,1,2-tris(N-pyrrolidinyl)dimethylaminoethylene.

8. A composition of claim **1** containing, as a chemiluminescent material, bis(N-pyrrolidinyl)methylene-1,3-dimethyl-2-imidazolidine.

9. A composition of claim **1** containing, additionally, silica.

10. A composition of claim **1** containing, as an aerosol propellant, a fluorinated hydrocarbon.

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LEON D. ROSDOL, Primary Examiner

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U.S. Cl. X.R.

252—301.2, 305, 308, 408