DEVICE FOR THE PACKAGING OF A THREE OR MORE COMPONENT CHEMILUMINESCENT SYSTEM

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References Cited
UNITED STATES PATENTS
3,539,794 11/1970 Rauhut et al. ......................... 240/2.25

ABSTRACT

A device for providing chemiluminescent light from a chemical reaction of suitable compounds in the presence of a fluorescent compound. The device comprises a self-contained light transmitting package having a number of reactants stored in separate compartments, all sealed with a single sealing clip. The reactants in the separate compartments are combined in a single compartment by removal of the single sealing clip.

4 Claims, 3 Drawing Figures
DEVICE FOR THE PACKAGING OF A THREE OR MORE COMPONENT CHEMILUMINESCENT SYSTEM

This invention relates to systems and devices for providing chemiluminescent light incorporating chemical components which react chemically and provide excitation for a fluorescent compound. The invention more particularly relates to systems and devices whereby the reactive components are maintained in a non-reactive condition until light is desired, the systems incorporating means to bring said components into a reactive condition and means to display the resultant light.

Under certain circumstances, it is desirable to have a source of visible light which is not electrically activated. Light can be provided by chemical systems, wherein the luminosity is solely the result of chemical reaction without provision of any electrical energy. Such light is known as chemiluminescent light.

Chemiluminescent light may be useful where there is no source of electricity. For example, in emergencies where sources of electrical power have failed, a chemiluminescent system could provide light. Since the system requires no externally generated source of energy, devices can be made small and highly portable. Moreover, chemiluminescent light is cold light and can be used where the heat of conventional illumination is not desired. It is also useful where electrical means could cause a fire hazard, such as in the presence of inflammable agents. Chemiluminescent light is also effective under water since there are no electrical connections to short out. Thus it may be seen that chemiluminescent light can have many useful applications.

A principal object of the present invention is to provide systems and devices incorporating chemiluminescent components for the provision of chemiluminescent light.

A further object of this invention is to provide means for containing chemically reactive chemiluminescent components in a non-reactive condition and means to combine said components when desired to provide chemiluminescent light.

A further object is to provide a self-contained, highly portable chemiluminescent lighting device having all chemical components therein and in which the light is displayed.

Another object is to provide a chemiluminescent lighting device which is inexpensive to make, easy to activate and highly effective.

Another object of the invention is to provide an illuminated package having a chemiluminescent lighting system.

A further object is to provide a chemiluminescent lighting package capable of containing different reaction materials, in separated compartments which may be readily and easily combined by removal of a single seal and the products of the reaction without leakage.

These and other objects of the invention will become apparent as the description thereof proceeds.

A chemical lighting system may require the separate packaging of three or more non-compatible components. This present device has as many compartments for component storage as required, but still uses only one clip for removal during mixing and reacting. An added advantage is the ability to fill each component from the same side of the package.

The device consists of a package separated into two sections by a removable clip placed horizontally across the package. One section of this device is partitioned vertically (at 90° to the clip) into the number of components required in the chemiluminescent system be either heat sealing or some other permanent or semipermanent manner. (One of the components can also be stored in the second section below the clip partitioning.) By removing the single clip all components can be mixed to form the chemiluminescent system.

Three or more non-compatible components may be packaged separately within one device. Only one removable clip is required to allow the mixing of all the components. All the components can be filled into the device from one end thus facilitating its production.

Chemical lighting systems e.g., thixotropic systems may require the addition to a regular chemiluminescent system of a 'non-storage-compatible' thickening agent. This device permits the packing of the agent in a separate compartment.

The chemiluminescent system of this invention thus comprises the device as described accomodating the admixture of at a number of chemiluminescent reagents and providing for the admixture in the device of at the chemiluminescent reagents comprising either (a) one component containing a chemiluminescent compound, another component containing a hydroperoxide compound, a component containing a fluorescence fluid or solid, and the like. Any other necessary ingredients for the production of chemiluminescent light, or for lifetime control, or for intensity improvement, or for storage stabilization may be included in one of the components with which they are compatible, or included as additional components. In particular with the preferred oxalic-type chemiluminescent compounds of this invention, a fluororescent compound must be included in the system.

The preferred chemiluminescent light is obtained in this invention by the reaction of a hydroperoxide with a chemiluminescent composition which, in combination, comprises a chemiluminescent compound selected from the group consisting of (1) an oxalic-type anhydride of the type disclosed and claimed in the co-pending application, 485,920 now U.S. Pat. No. 3,399,137, which is hereby incorporated by reference, (2) an oxalic-type amide of the type disclosed and claimed in co-pending applications, Ser. No. 520,052 now U.S. Pat. No. 3,442,815 and 547,782 now abandoned, which are hereby incorporated by reference, (3) an oxalic-type O-acylhydroxylamine of the type disclosed and claimed in co-pending application, Ser. No. 547,761 now abandoned, and (4) an oxalic-type ester disclosed and claimed in application, Ser. No. 491,896 now abandoned, in the presence of a fluororescent compound, and a solvent. Other suitable chemiluminescent compounds are 3-aminophthalaldehyde, 3,4,5-triphenylimidaole, 10,10'-dialkyl-9,9'-biacridinium salts, and 9-cholorocarboxyl-10-methylacridinium chloride. The latter is disclosed and claimed in co-pending application, Ser. No. 427,459 now U.S. Pat. No. 3,352,791. All of the foregoing provide chemiluminescence when reacted with a hydroperoxide compound in the presence of a base. Other chemiluminescent materials are described by K. D. Gunderman, Angew. Chemie, Int. Ed., 4, 566/1965.

The preferred chemiluminescent compound of this invention is an oxalic-type ester selected from the
group consisting of (a) an ester of an oxalic-type acid and an alcohol characterized by acid ionization constant in water greater than $1.3 \times 10^{-10}$, and (b) a vinyl ester of an oxalic-type ester. Similarly, in a preferred embodiment thereof, the alcohol would be aromatic alcohol substituted by a substituent characterized by a positive Hammett sigma value. The preferred species of oxalic-type esters include bis(substituted-phenyl)oxalate such as bis(2-nitrophenoxy)oxalate, bis(2,4-dinitrophenyl)oxalate, bis(2,6-dichloro-4-nitrophenoxy)oxalate, bis(3-trifluoromethyl-4-nitrophenoxy)oxalate, bis(2-methyl-4,6-dinitrophenyl)oxalate, bis(1,2-dimethyl-4,6-dinitrophenyl)oxalate, bis(2,4-dichlorophenoxy)oxalate, bis(2,5-dinitrophenyl)oxalate, bis(2-formyl-4-nitrophenoxy)oxalate, bis(pentachlorophenoxy)oxalate, bis(1,2-dihydro-2-oxo-1-pyrindyl)-glyoxal, bis-N-phthalimidyl oxalate.

Other bisphenyl esters of oxalic acid which have been found to be effective are disclosed in copending, commonly assigned applications Ser. Nos. 124,142, now U.S. Pat. No. 3,749,679 and 886,406, now abandoned, filed Mar. 15, 1971, and Dec. 18, 1969, respectively. These applications disclose bisphenyl esters of oxalic acid as discussed above, and which are further substituted with alkoxypbenyl and carbalkoxy groups. These applications are incorporated herein by reference. The peroxide employed in the components of this invention may be any hydroperoxide compound. Typical hydroperoxides include t-butylhydroperoxide, peroxybenzoic acid, and hydrogen peroxide. Hydrogen peroxide is the preferred hydroperoxide and may be employed as a solution of hydrogen peroxide in a solvent or as an anhydrous hydrogen peroxide compound such as perhydrate of urea (urea peroxide), perhydrate of pyrophosphate (sodium pyrophosphate peroxide), perhydrate of histidine (histidine peroxide), sodium perhydrate of pyrophosphate (sodium pyrophosphate peroxide), perhydrate of histidine (histidine peroxide), sodium perborate, sodium peroxide, and the like. Whenever hydrogen peroxide is contemplated to be employed, any suitable compound may be substituted which will produce hydrogen peroxide.

The peroxide concentration may range from about 15 molar down to about $10^{-5}$, preferably about 2 molar down to about $10^{-2}$ molar. The ester of this invention may be added as a solid or in admixture with a suitable solid peroxide reagent or in a suitable diluent, or alternatively dissolved directly in a solution containing the peroxide reagent.

Typical diluents, which additionally may be used in conjunction with the necessary diluent of this invention, are those which do not readily react with a peroxide such as hydrogen peroxide, and which do not react with an ester of oxalic acid.

Where a solvent is employed with the hydroperoxide-containing component of this invention said solvent can be any fluid which is unreactive toward the hydroperoxide and which accommodates a solubility of at least 0.01 M hydrogen peroxide. Typical solvents for the hydroperoxide component include water; alcohols, such as ethanol or octanol; ethers, such as diethyl ether, diisopropyl ether, tetrahydrofuran, dioxane, dibutyl-diethyleneglycol, perfluoropropyl ether, and 1,2-di-methoxyethane; and esters, such as ethyl acetate, ethyl benzoate, dimethyl phthalate, dioctylphthalate, propyl formate. Solvent combinations can, of course, be used such as concentrations of the above with aromatic anisole, tetralin, and polychlorobiphenyls, providing said solvent combination accommodates hydroperoxide solubility. However, when oxalic-type chemiluminescent materials are used, strong electron donor solvents such as dimethyl formamide, dimethyl sulfoxide, and hexamethylphosphoramide should not, in general, be used as a major solvent component.

Where a solvent is employed with a component containing the chemiluminescent material any fluid can be used providing said fluid solubilizes at least 0.01 M concentration of the chemiluminescent material and is unreactive toward the chemiluminescent material. Typical solvents include ethers, esters, aromatic hydrocarbons, chlorinated aliphatic and aromatic hydrocarbons, such as those cited in the preceding paragraph. For oxalic-type chemiluminescent compounds, hydroxyl solvents such as water or alcohols and basic solvents such as pyridine should not be employed since such solvents used in general, react with and destroy oxalic-type chemiluminescent compounds. Solvent combinations may, of course, be used but such combinations when used with oxalic-type chemiluminescent compounds should not include strong electron donor solvents.

When a component comprising a solid chemiluminescent compound and a solid hydroperoxide is used, the solvent or solvent composition comprising the second component may vary broadly. Said solvent, however, should preferably dissolve at least 0.02 M concentration of both, the hydroperoxide and the chemiluminescent compound, and for oxalic-type chemiluminescent compounds, strong electron donor solvents should be avoided as major solvent components.

The fluorescent compounds contemplated herein are numerous; and they may be defined broadly as those which do not readily react on contact with the peroxide employed in this invention, such as hydrogen peroxide, likewise, they do not readily react on contact with the chemiluminescent compound.

A fluorescent compound is required for light emission when the prepared oxalic-type chemiluminescent compound of the invention is employed. For other types of chemiluminescent compounds a fluoroscer is not required but may be used to shift the wavelength of emitted light toward the red region of the spectrum so as to change the color of emitted light. Fluorescent compounds for use with oxalic-type chemiluminescent compounds should be soluble in the reactive solvent at least to the extent of 0.0001 moles per liter.

Typical suitable fluorescent compounds for use in the present invention are those which have a spectral emission falling between 330 millimicrons and 1000 millimicrons and which are at least partially soluble in any of the above diluents, if such diluent is employed. Among these are the conjugated polycyclic aromatic compounds having at least 3 fused rings, such as anthracene, substituted anthracene, benzanthracene, phenanthrene, substituted phenanthrene, naphthalene, substituted naphthalene, pentacene, substituted pentacene, and the like. Typical substituents for all of these are phenyl, lower alkyl, chlorine, bromine, cyano, alkyl (C₁-C₄₀), and other like substituents which do not interfere with the light-generating reaction contemplated herein.

Numerous other fluorescent compounds having the properties given hereinabove are well known in the art. Many of these are fully described in "Fluorescence and
ers are described in “The Colour Index,” Second Edition, Volume 2, The American Association of Textile Chemists and Colorists, 1956, pp. 2907–2923. While only typical fluorescent compounds are listed herein-
above, the person skilled in the art is fully aware of the fact that this invention is not so restricted and that nu
merous other fluorescent compounds having similar
properties are contemplated for use herein.
A fluorescent oxalic-type ester, such as the oxal
c acid ester of 2-naphthol-3,6,8-trisulfonic acid, does not
require a separate fluorescent compound to obtain light. Other typical fluorescent oxalic acid esters in
clude esters of oxalic acid (1) 2-carboxyphenol, (2)
2-carboxy-6-hydroxyphenol, (3) 1,4-dihydroxy-9,10-
diphenylanthrancene, and (4) 2-naphthol. Thus, a react
 tant including a fluorescent oxalic-type ester would thereby include at least one fluorescent compound.

It has been found that the molar (moles per liter of diluent) concentrations of the major components of the novel composition herein described may vary considerably. It is only necessary that components be in suffi
cient concentration to obtain chemiluminescence. The ester of oxalic acid molar concentration normally is in
the range of at least about $10^{-5}$ to 5 molar, preferably in the range of at least about $10^{-3}$ to about 1 molar; the fluorescent compound is present in the range from about $10^{-3}$ to 5, preferably $10^{-2}$ to $10^{-1}$ molar; and the diluent must be present in a sufficient amount to form at least a partial solution of the reactants involved in the chemiluminescent reaction. If the ester is liquid, it may serve as either the sole diluent or a partial diluent.

The wavelength of the light emitted by chemilumi
nescence of the compositions of this invention, i.e., the color of the light emitted, may be varied by the addition of any one or more energy transfer agents (fluorescers) such as the known fluorescent compounds discussed at length above.

The wavelength of the light emitted by the composi
tion of this invention will vary, depending upon the par
icular fluorescent component employed in the reac
tion.

Additionally, it has been found that the superior in
tensity of chemiluminescence is obtained when the final mixture producing the luminescence is maintained at a temperature of between about −40°C and 75°C, preferably between about 20°C and 50°C. However, temperature is not critical and the luminescence of Ap
licants’ process is not limited to these ranges.

The lifetime and the intensity of the chemilumi
nescence light obtained with the preferred oxalic-type chemiluminescent compounds of this invention can be regulated by the use of certain regulators such as:

(1) By the addition of base to the chemiluminescent composition. Both the strength and the concentration of the base are critical for purposes of regulation.

(2) By the variation of hydroperoxide. Both the type and the concentration of hydroperoxide are critical for the purposes of regulation.

(3) By the addition of water.

(4) By the addition of a catalyst which changes the rate of reaction of hydroperoxide with the oxalic-type ester. Catalysts which accomplish that objective in
clude those described in M. L. Bender, “Chem. Revs.,” Vol. 60, p. 53 (1960). Also, catalysts which alter the

rate of reaction or the rate of chemiluminescence in
clude those accelerators of copending application, Ser.
No. 577,595, now abandoned, and decelerators of co
pending application, Ser. No. 577,615, now aban
doned.

While acids are not in general accelerators for oxalic
type chemiluminescent reactions it should be noted specifically that acids are accelerators for the oxalic
amide chemiluminescent compounds of copending ap
plication, Ser. No. 547,782, now abandoned.

More specifically, the advantages obtained by the in
corporation of a catalyst of Ser. No. 577,595 may be
obtained in conjunction with the objects of this present
invention by employing, according to the copending
application, an ionized salt having a cation selected from (a) an organic quaternary cation selected from the group consisting of ammonium, arsenic, and phos
phorous, and (b) alkali metal having an atomic weight above 22, the salt of said cation preferably being soluble in an organic solvent and preferably being charac
terized by a property of forming cation-aggregates when reacted with the oxalic-type ester and a hydro
peroxide. One of the advantages is the fact that an exces
sive amount of the chemiluminescent agent may be
employed whereby a higher quantum yield may be ob
tained when the ionized salt is employed, in contrast to systems not employing the accelerator whereby such systems would be limited to a much lower maximum concentration of chemiluminescent agent which would continue to increase rather than decrease the total quantum yield of chemiluminescent light.

Similarly, within the scope of the present invention is
the concurrent employment of one or more decelerators
either alone in the composition of this invention, or in conjunction with one or more of the accelerators
discussed in the preceding paragraphs. By employing
one of the accelerators of the preceding paragraph, it
would be possible to employ a greater total concentra
tion of the chemiluminescent agent while concurrently
would be possible to employ a decelerator which would prolong the period during which the light of high inten
sity is obtained from the chemiluminescent reaction. Such decelerators set forth in the copending applica
tion, Ser. No. 577,615, include for example a com
pound such as oxalic acid.

When oxalate-type chemiluminescent compounds are
used in a solution component it may be desirable to
include a stabilizing agent such as those described in copending application, Ser. No. 614,397, now aban
doned.

The chemical compounds, components and their re
actions for providing chemiluminescent light are de
scribed in copending, commonly assigned applications, Ser. Nos. 442,802 now U.S. Pat. No. 3,329,621;
442,818 now U.S. Pat. No. 3,425,949, and those previ
ously mentioned, and as such they do not form a part of the present invention.

If a gelled chemiluminescent composition is desired, a thixotropic or gelling agent may be included in one
of the compartments. Suitable agents are disclosed in U.S. Pat. No. 3,671,450, and are for example, alkali
and alkaline earth metal soaps; bentonite; polyaryl
ureas; silicas; saccharides; indanthrene blue (less desir
able because of intense color); natural plant hydrocol
loids such as guar, tragacanth, algin and the pectins;
starches and starch derivatives; cellulose and synthetic
cellulose derivatives such as nitrocellulose; polyvinyl
compounds such as polyvinyl alcohol, polyvinyl pyrrolidone, polycrylates, and ethylene oxide polymers; hevea latex.

In this invention, the reactive components are stored in a multiple compartment container device having a plurality of compartments, wherein the separate components may be brought into contact to produce the reaction which provides chemiluminescent light to be displayed in said container. When either the chemiluminescent compounds, hydroperoxide, or both are fluid, they must be in separate compartments. The diluent and fluorescent compounds can be in either of these two compartments. If the chemiluminescent compounds, hydroperoxide and fluorescent compounds are dry powdered solids, they may be kept together in one compartment with the diluent in the other compartment. The reactive components are brought together to provide chemiluminescent light.

The invention may be better understood by reference to the drawings in which

FIG. 1 shows a plan view of a flexible package as one embodiment of the chemiluminescent light device.

FIG. 2 shows a view of the same device as in FIG. 1, showing reactants in the various package compartments.

Referring to FIGS. 1 and 2 an elongated rectangular package A is composed of two sheets 5 and 6 of flexible material, sealed around the outside edges. The seal may be by any suitable means, such as heat sealing, or suitable adhesive. One portion B of package A is divided into a plurality of compartments, as shown compartments 1, 2, 3 and 4 by means of heat sealing (or otherwise sealing) sheets 5 and 6 at points 7, 8, and 9, from package edge 10 towards the opposite end 11. Thus the compartment 1, 2, 3 and 4 are open at their ends facing edge 11 and open into a larger single compartment 12 in portion C of package. A clamp D is provided which reaches across all open ends of compartments 1, 2, 3 and 4 to close them from single compartment 12. Thus each of the four compartments may contain separate, non-compatible ingredients for chemiluminescent light production which are kept separate from each other. When clip D is removed the ingredients from compartments will be admixed in compartment 12 and reach chemically to provide chemiluminescent light as shown in FIG. 3. Thus at least one of the sheets 5 and 6 should be light transmitting i.e., transparent or translucent. The other sheet may be reflective, opaque or likewise transparent as desired.

The sheets may be plastic, flexible aluminum foil, or the like, so long as they are impervious to the fluids contained and inert to the reactants. Polyethylene, polypropylene, teflon or the like have been found suitable as plastics.

Clip D may be of any suitable design, many of which are known in the art. One type is shown in FIG. 2 which consists of a rod 13 around which package A is wrapped and an outer clamp 14 conforming to the rod shape. This type of clip may be easily removed by pulling on package edges 10 and 11.

The package A is moreover easily prepared. For this, it may be held with edge 11 upward and unsealed, while the other three edges are sealed. Ingredients are placed in compartments 1, 2, 3, 4 at the open ends and clip D is attached which effectively closes the open ends of compartments 1, 2, 3, and 4. Edge 11 is then heat sealed to form compartment 12, and finish the light package.

The color of the light emission will depend on the type of fluorescent compound and its spectral response. However, the visible color could be varied by using a colored plastic for sheets 5 or 6.

It will be obvious that the construction of the invention may be varied so long as the basic requirements are maintained.

The invention provides a device for providing visible light whenever and wherever desired, independent of conventional electrical lighting methods and without the hazards and limitations of electrical lighting. The chemiluminescent lighting systems can be especially useful in emergency situations where other forms of lighting have failed. The systems do not have the fire of ignitable lighting devices such as candles, gas, or oil lights.

It will be readily apparent that the chemiluminescent device is not confined to emergency lighting, however. It can be used at any time where a cold, safe illuminating means is desired. It is also useful to provide illumination where electrical illumination is unavailable. The device is highly portable and can be hand held for signalling. In addition, by use of a magnetic attachment, the package could be made to adhere to metal surfaces.

While certain specific embodiments and preferred modes of practice of the invention have been described, it will be understood that this is solely for illustration, and that various changes and modifications of the invention may be made without departing from the spirit of the disclosure or the scope of the appended claims.

1 claim:

1. A chemiluminescent light comprising a package having two coextensive, flexible sheets, sealed together at the outer edges, a plurality of parallel elongated compartments formed by sealing said sheets together along spaced apart lines parallel to one edge of the package, said lines extending a portion of the distance along said one edge, a removable clamp means adapted to clamp across said package transverse to said elongated compartments and close said compartments from the remainder of the package, to form a larger single compartment in the remaining area of the package, at least one of said sheets being light transmitting, and a reactive chemiluminescent ingredient in each elongated compartment.

2. The chemiluminescent light of claim 1 wherein said sheet material is a plastic of the group polyethylene, polypropylene or teflon.

3. The chemiluminescent light of claim 1 wherein one of said flexible sheets is reflective.

4. The chemiluminescent light of claim 1 having four elongated compartments and wherein each elongated compartment contains one of the ingredients, a bis-(aryl)xalate ester, an organic fluorescent compound, hydrogen peroxide, a rate regulator catalyst, one of said compartments also containing an organic solvent for said ingredients.