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[54] **LIQUID SCINTILLATION, COUNTING AND COMPOSITIONS**

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Related U.S. Application Data

[63] Continuation-in-part of Ser. Nos. 613,109, Feb. 1, 1967, abandoned, and Ser. No. 29,267, April 16, 1970, abandoned.

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[51] **Int. Cl.** **F21k 2/02**

[58] **Field of Search** ..... 252/301.2 SC, 301.2 P; 252/408, 364; 250/483, 484

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3,307,628	3/1967	Sena	166/274
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[57] **ABSTRACT**

The emissions of radioactive isotopes in both aqueous and organic samples can be measured by liquid scintillation counting in micellar systems. The micellar systems are made up of scintillation solvent, scintillation solute and a mixture of surfactants, preferably at least one of which is relatively oil-soluble water-insoluble and another which is relatively water-soluble oil-insoluble.

10 Claims, No Drawings

LIQUID SCINTILLATION, COUNTING AND COMPOSITIONS

CROSS REFERENCE TO RELATED APPLICATIONS

This Application is a continuation in part of our earlier, now abandoned, U.S. Pat. Applications Ser. No. 613,109 and Ser. No. 29,267, filed Feb. 1, 1967, and Apr. 16, 1970, respectively.

SUMMARY OF THE INVENTION

This invention includes micellar systems and processes for liquid scintillation counting of emissions from radioactive materials using micellar systems. The micellar systems contain a plurality of surfactants having substantially different characteristics. Sometimes the differing surfactants may be of the same surfactant type, but more often and preferably, the surfactants are both substantially different in character and of differing surfactant types. Preferably, the surfactants include a relatively water-soluble oil-insoluble specie and a relatively oil-soluble water-insoluble specie and the most preferred surfactants are a combination of anionic type and non-ionic type surfactants.

BACKGROUND FOR THE INVENTION

Many commercial surfactants used to prepare emulsions and micellar systems over the years are mixtures made from natural products, such as fats (soaps) or refinery distillation cuts (petroleum sulfonates). Other commercial surfactants are mixtures obtained by separating, by distillation or other means, a fraction of polymerized, condensed, etc. mixture of components. Blends of these commercial surfactants have been used to formulate emulsions via the Hydrophile-Lipophile Balance. This approach is discussed in C. G. Sumner's "Clayton's The Theory of Emulsions and Their Technical Treatment," 5th Ed., Chemical Publishing Co., Inc., N.Y. (1954), p. 178. The alkyl phenoxy polyethoxyethanols, the alkanolamides, etc. are examples of this type of surfactant.

Sena, U.S. Pat. No. 3,307,628, teaches the use of combinations of surfactants which are preferentially water and oil soluble in micellar systems used in secondary oil recovery.

Soft radiation is measured by incorporating a scintillation solvent in solution with an emitting substance. (Principles of Radio Isotope Radiology by G. D. Chase and J. L. Rabonowitz, Burgess Publishing Co., Minneapolis, Minn., (1964), pp. 202–205) Various aspects of the technology of liquid scintillation counting are reviewed in the book, "Liquid Scintillation Counting," Bell and Hayes, Ed., Permagon Press, N.Y., N.Y. (1958). Advances in this area were later reviewed by E. Rapkin, International Journal of Applied Radiation and Isotopes, Vol. 15, (1964), pp. 69–87.

R. C. Green, et al., Anal. Chem., Vol. 40, (1968), pp. 2035–2037, suggest that mixtures of alkyl phenoxy polyethoxyethanol surfactants improve on the Benson process of U.S. Pat. No. 3,573,218. Benson teaches emulsion counting in a solubilized system utilizing the alkyl phenoxy polyethoxyethanols in liquid scintillation counting. However, the procedure is beset by a host of problems (R. H. Benson, Anal. Chem., Vol. 38 (1966), pp. 1353–1356, R. C. Green, et al., loc cit. and E. D. Bransome in the book, "The Current Status of Liquid Scintillation Counting", published by Grune and Stratton, N.Y. and London, (1970), Chap. 19.)

W. C. Tosch, Anal. Chem. 37, 959, June 1965, found transparent emulsions (solubilized systems) to be useful in uniformly dispersing water solutions containing gamma-emitting tracers. Tosch speculated that systems of the type described might prove useful in liquid scintillation work with lower energy sources.

However, transparent systems used by Tosch are not suitable for counting emissions from low energy sources such as tritium, even when the usual primary and secondary scintillation solutes and scintillation solvents are incorporated in the systems. In tests, chemical quenching and chemiluminescence were quite pronounced—sometimes to the extent that the emissions desired to be counted could not be detected.

Hansen, et al., U.S. Pat. No. 3,506,828, teach that quaternary ammonium compounds can be used for liquid scintillation counting, but that the use of secondary solvents reduces the efficiency of the systems. Still later, however, R. V. Webb, U.S. Pat. No. 3,677,964, combined a quaternary amonium base, methanol and another alcohol to form a suggested equilibrium mixture of a quaternary hydroxide and methoxide which was used as a solvent composition for liquid scintillation counting. The art is apparently evolving down a number of paths and meaningful insights into the art of liquid scintillation counting are often lacking.

DESCRIPTION OF THE INVENTION

We have now discovered that thermodynamically stable micellar systems made up of surfactants having substantially differing characteristics and counting media prepared therefrom provide high counting efficiencies and show little or no chemiluminescence, quenching or phosphorescence when made up of a plurality of surfactants of substantially differing properties. Surprisingly, the micellar systems are operative in the presence of dyes when the dyes form a part of the micelles. These systems differ from the prior art solubilizers in that the prior art emulsion systems, on vapor phase and nuclear magnetic resonance study, contain substantial bulk fluid as an internal phase.

This invention includes micellar systems and the formulated counting samples as well as processes for their preparation. Useful micellar systems are uniphase or can have an internal micellar phase within an external phase. Where the micellar system has an internal phase, the vapor pressure of this phase is less than that of the bulk fluid. The internal phase thus contains little or no bulk fluid because, we postulate, the fluid, speaking in terms of water, "hydrates" with the micelles and becomes a part of the actual micellar structure. NMR studies also show the absence of any substantial bulk dispersed phase fluid when studied according to the procedure of P. D. Cratin, "Dielectric, Near Infrared, and NMR Studies of Solubilized and Emulsified Water," paper presented at the 147th National Meeting of the A.C.S. (1964).

While a micellar system useful in liquid scintillation counting is oftentimes no more than a mixture of surfactants and cosurfactants, the formulated counting sample may contain, in addition to the base micellar system, one or more of scintillation solvent, primary solute and secondary solute. The scintillation solvent, for example toluene or the xylenes, can be the nonpolar phase of a micellar system with an aqueous dispersed phase. The micellar system and the counting

medium will solubilize a fluid or dissolvable radioactive sample to at least one-half and preferably at least an equal amount of volume of the original micellar system present without forming a transparent thermodynamically unstable dispersion which contains dispersed free (bulk) sample material. Thermodynamically unstable dispersions are those which will not repeatedly reform a stable dispersion after breaking of the dispersion through temperature cycling.

The surfactants, cosurfactant and any base solvents, i.e., those needed to solvate isotopic samples, surfactant, etc. used in this invention are substantially inert from the standpoint of chemical reactivity with the other components of the counting samples and from the standpoint of energy transfer which results in chemiluminescence, fluorescence or phosphorescence. Further, all counting medium components are liquid in the sample containing counting samples under counting conditions. Base solvents which can be used in formulating the micellar systems can be both polar and non-polar. Water is the preferred polar base solvent, while lower molecular weight paraffins, olefinic hydrocarbons without substantial conjugation, uncondensed aromatics, etc. are preferred non-polar solvents. The use of non-polar base solvents is preferred where a solvent is utilized.

Micellar systems used in formulating the counting media of this invention are similar to surfactant mixtures used in formulating the "soluble oils" and "microemulsions" of the dry-cleaning, cutting oil, etc. art. Methods useful in compounding the micellar systems are those presently used in formulating emulsions and include, for example, the approach called the Hydrophil-Lipophil Balance method of formulating emulsions.

A variety of surfactants can be used in combination in the micellar systems and counting samples of this invention. However, while the formulation techniques common to the "emulsion" and "soluble oil" art are used in formulating the micellar systems and counting media of this invention, the problems faced in the preparation of emulsions also arise. For example, cationic and anionic surfactants are not always useful in combination. Some empirical experimentation is required to formulate optimum micellar systems and counting samples from given combinations of surfactants and cosurfactants.

One skilled in this art will, like an architect appraising a new construction, know the tests necessary to determine the utility of new materials, for example, scintillation solvents, surfactants, etc.

At least two surfactants which are substantially different in character are used to prepare the micellar solutions and counting media of this invention. Substantially different means that: (a) one surfactant will be preferentially oil-soluble while the other is preferentially water-soluble, (b) the surfactants are of different chemical families, e.g., the polar moiety of the molecule of one surfactant is a carboxyl moiety while the other is a sulfonate moiety, (c) the surfactants are of different surfactant types, e.g., one is an anionic surfactant and the other a non-ionic surfactant, or (d) one surfactant acts as a dispersant while the other acts as a surface tension reducing agent. The definition of "substantially different" is not intended to include the mixtures of close homologs or analogs of a particular surfactant which are found in a commercial surfactant, such as "Triton X-100".

Many surfactants find use in our compositions and processes, but should not have substantial conjugation in the surfactant molecule as, for example, is found in certain petroleum sulfonates. Among the useful surfactants are the fatty acid soaps and alcohols, the polyethoxyethanols, the alkyl succinates, the ethylene oxide-propylene oxide condensates, etc. More specifically included within these groupings are sodium linoleate, linolic acid, stearic acid, potassium oleate, glycerolactopalmitate, lauryl alcohol, dodecyl alcohol, octadecyl alcohol, glycerol monostearate, morpholine oleate, sorbitan trioleate, polyoxyethylene sorbitol hexasterate, propylene glycol monostearate, sorbitan sesquioleate, polyoxyethylene sorbitol-4,5-oleate, diethylene glycol monooleate, polyoxyethylene dioleate, polyoxypropylene mannitol dioleate, polyoxyethylene oxypropylene oleate, tetraethyleneglycol-monolaurate, polyoxyethylene lauryl ether, polyoxyethylene cetyl ether, isooctylphenol-polyethoxyethanol and dihexyl sulfosuccinate.

Surfactants having amines with one or more hydrogens, sulfonates, sulfonic acids having substantial conjugation, ketones and quaternary compounds (under basic or essentially neutral conditions) often severely quench the emissions being analyzed. This phenomena is not understood, but definitely exists as is shown in the specific examples.

Cosurfactants (secondary solvents) used in making up the micellar systems and counting samples include the lower molecular weight alcohols, ethers, and esters. Again, amines, ketones, etc. can cause problems.

The usual primary and secondary scintillation solutes can be used in making up the micellar systems. The primary solute absorbs energy given off by nuclear radiation and emits a longer wave length radiation. A secondary solute absorbs the a longer wave length radiation given off by the primary solute and emits visible light detectable by a photomultiplier tube. PPO, 5-diphenyl oxazole is the preferred scintillation solute while POPOP, 2,2-phenylene bis (5-phenyloxazole) is the preferred secondary scintillation solute. Other scintillation solutes are useful. These include p-terphenyl, dimethyl-POPOP, diphenylhexatriene, α-NPO, α-NOPON, etc. Scintillation solvents used in this invention are also well known in the liquid scintillation counting art and include toluene, xylene, p-cymene, dioxane, etc.

The following examples more specifically illustrate this invention.

EXAMPLE I

A system is made up containing, by weight, 25.1% tall oil fatty acids obtained from the Arizona Chemical Company, 24.2% n-hexanol, 15.8% isopropanol, 22.1% isooctylphenolpolyethoxyethanol (Energetic W-100 from Armour and Company) and 12.8% aqueous solution of NaOH (25%). Two milliliters of this micellar system are added to ten milliliters of toluene containing 7 g/l PPO and 0.36 g/l POPOP to prepare a counting sample.

This counting sample will tolerate moderate amounts of inorganic acids and limited amounts of alkali. On addition of the sample the system clarifies immediately in most cases. If the amount of aqueous solution is near maximum for the amount of micellar system used, the clarification is slower and additional micellar system should be added. If a satisfactory counting system is not obtained on addition of further micellar system, the

sample is diluted where necessary or less aqueous solution is utilized in making up a new counting solution. Strong acids may be neutralized with sodium hydroxide and bases with additional tall oil fatty acids or with the formulation of Example II. Preferably the pH of the sample containing counting sample to be assayed is about 7. If it is necessary to count samples containing dissolved 14_{CO}, small amounts (0.5) ml of redistilled ethanolamine is added to the counting sample.

EXAMPLE II

This counting sample is designed as an acid solubilizer for alkaline tissue digests or any strongly alkaline sample. It contains, by weight, 28.7% tall oil fatty acids, 27.8% n-hexanol, 18.2% isopropanol, and 25.3% isooctylphenolpolyethoxyethanol. This micellar system is added to the same proportions of the toluene-fluor used in Example I. Materials to be assayed, such as 50 mg of wet tissue, are put into an oven at 80° C for 1 hour or until clear and so digested in two milliliters of 2 N NaOH, all of which are added to the cocktail to be counted. This system will not make a satisfactory counting system unless it is neutralized. Again, optimum counting pH is about 7 as tested with pH paper.

If the amount of organic material is large, a clear solution may not form. The counting sample should then be supplemented with small amounts of the counting sample of Example I, in the ratio of 2 ml of the counting sample of Example I to 10 ml of toluene.

EXAMPLE III

About 50 mg hemoglobin is weighed into a liquid scintillation vial. One ml of 2 N NaOH is added to the hemoglobin and the sample is heated in an oven at about 80° C for an hour and cooled. Na_2O_2, about 50 mg, is added to discharge the color. Counting sample of Example II is added, including a slight excess to neutralize the alkali from the Na_2O_2 and counted. Severe quenching may be encountered in the so-treated systems although counting of C-14 and H-3 is possible.

The alkaline digestion technique of this Example followed by a neutralization and solubilization with the counting sample of Example II is applicable to the counting of a variety of insoluble substances, for example aged urine that contains relatively large amounts of precipitated protein. This technique may also be utilized to solubilize tendons, cellulosic materials, dried and decomposed tissue, synthetic polymers, etc. Two ml of the counting sample of Example II will normally solubilize about 25 mg of pure, dried protein.

EXAMPLE IV

A general performance counting sample is prepared, by weight, from 37.9% sodium dihexylsulfosuccinate, 4.8% isopropanol, 4.7% water and 52.6% isooctylphenol polyethoxyethanol. This micellar system forms a counting sample having a slightly higher tolerance for salts than the counting sample of Example I and, also, a lower natural radiation background. The volume of counting sample required is only about two times that of the volume of aqueous sample to be counted. This counting sample works well in the counting of uranyl salt solutions. The micellar system is added to the toluene-fluor of Example I in a ratio of 1 : 2 : 10.

EXAMPLE V

Table I shows typical results in several counting situations with tritium labeled compounds. All counting vials contained 12 ml of the toluene-flour mixture and 3 ml of the micellar system of Example I or IV. Except as noted, the toluene-fluor solution contained 7 g/l PPO and 0.36 g/l of POPOP. Efficiencies were determined by the internal standard method using a Beckman liquid scintillation spectrometer employing an RCA photomultiplier tube.

TABLE I

Counting Efficiency of Several Tritium Labeled Compounds in Counting Sample Made with Solubilizers

Substance Solubilized	Amount	Efficiency
WATER, spiked with tritiated water	0.1 ml	42%
	0.2 ml	39
	1.0 ml	37
	2.0 ml	32
TOLUENE H-3 (alone)	—	55
Toluene H-3 with water	1.0 ml (water)	49
H-3 labeled URINE	0.5 ml	41
	1.0 ml	37
ASCORBIC ACID H-3 (aqueous solution)	1.0 ml	45
BLOOD PLASMA labeled with H-3	1.0 ml	30
WATER, spiked with tritiated water (o-xylene PBD as fluor)	0.2 ml	44
	0.7 ml	39
	1.0 ml	35

EXAMPLE VI

Carbon 14 can be counted at about 80-90% efficiency using the counting sample of Example IV. A counting solution spiked with C-14 labeled toluene gave an efficiency of 89% when anhydrous or when containing 1 ml of added water. 87% efficiency was obtained in a counting sample formulated with the micellar system of Example I and containing 1 ml of aqueous fuctrose C-14 (at 1%) solution. 87% efficiency was obtained with the cocktail of Example I containing 1 ml of aqueous glycine C-14 in 1% solution.

EXAMPLE VII

Fe-55 was counted at about 30% efficiency when 1 ml of an Fe-55 labeled brine containing approximately 22,000 parts per million totaled dissolved solids was dispersed in a cocktail formulated with the micellar system of Example IV.

EXAMPLE VIII

Cl-36 was counted at about 100% efficiency when 1 ml of a Cl-36 labeled isotonic saline sample was dispersed in a cocktail formulated utilizing the micellar systems of Example IV.

EXAMPLE IX

Co-60 was counted with 100% efficiency (beta radiation being monitored) when 1 ml of Co-60 labeled aqueous solution of $CoCl_2$ was solubilized in a cocktail formulated with micellar system of Example IV. Efficiencies approaching 100% are expected for radioisotopes with maximum beta radiation energies of a few hundred KEV or greater.

EXAMPLE X

A toluene-fluor of 6 g PPO in 1 liter of toluene was prepared. The toluene-fluor (25 g) was then mixed with 25 g each of sulfonate surfactants A, B and C. Surfactant A is Petronate K (a highly refined petroleum sulfonate manufactured by the Witco Chemical Com-

pany's Sonneborn Division). Sulfonate B is "Shell Sac" (a Shell Chemical Company petroleum sulfonate which was vacuum evaporated to remove the vehicle oil. This material was approximately 100% active.). Surfactant C is "Shell" No. 65,413, Lot No. S-65-148 petroleum sulfonate analyzing approximately 62% active sulfonate. The toluene-fluor, 15 ml, was then added to 1, 2 and 3 ml samples of each of the three surfactant-fluor micellar systems described above. A control of 15 ml toluene-fluor was used in each instance.

The samples were placed in a Beckman Liquid Scintillation System without addition of radioactive sample to obtain "background" counting rates. The samples were then counted repeatedly until an approximately constant counting rate was observed for all samples. Initial counting rates of 24,609.0, 37,294.0 and 49,987.0 were observed for the 1, 2 and 3 ml solutions of surfactant A respectively. After approximately 60 minutes, the counting rates were 2,093.0, 5,096.0 and 7,866.0. The initial counting rates for systems made up with surfactant B were 34,576.0, 40,687.0, and 48,686.0 while the counts at the end of about 60 minutes were 3,239.0, 5,510.0, and 7,259.0 respectively. The initial counting rates of samples containing surfactant C were 32,305.0, 46,595.0, and 34,353.0 and, after approximately 60 minutes were, respectively, 2,512.0, 4,399.0, and 4,471.0.

During this time the controls were running approximately 30 to 40 counts per minute, sometimes with a high initial background count of 100 to 200 counts per minute decaying almost immediately to below 100.

EXAMPLE XI

Water was added until all of the control were turbid and contained a water layer at the bottom.

The counting sample containing 1 ml of surfactant A received 0.5 ml of water. It became turbid and remained so though some isopropyl alcohol was added in attempts to clarify the system. The addition of isopropanol up to a total of 0.07 ml isopropanol increased the turbidity of the system. At this point, the system was a stable, opaque macroemulsion. All other samples received only 0.10 ml of water except the sample containing 3 ml of surfactant A which received 0.12 ml of water. All of these samples were clear both before and after counting.

On addition of water, the three samples of counting sample containing surfactant A had an initial counting rate of 1,666.0, 4,190.0 and 10,320.0 respectively while a counting rate of 1,319.0, 3,353.0 and 7,134.0 was observed after 36 minutes of counting time. The counting sample containing surfactant B had initial counting rates of 2,563.0, 3,730.0, and 6,364.0, respectively. After 36 minutes, the counting rates were 2,034.0, 3,139.0, and 4,746.0 respectively. The samples containing surfactant C had initial counting rate of 1,594.0, 2,814.0, and 9,063.0 respectively, while the counting rates were 1,180.0, 2,373.0, and 4,312.0 respectively after 36 minutes of counting time.

EXAMPLE XII

Tritiated water (0.05 ml) was added to all samples counted in Example XI. 91,000 counts per minute were expected at 100% efficiency after making the correction for natural decay of the material from Jan. 10, 1964.

Phase stability and appearance of sample remained exactly the same as noted in the prior Example. A slight amount of water, however, appeared at the bottom of the vial used in counting the counting sample containing 1 ml of surfactant C. Initial counts for the three samples containing 1, 2 and 3 ml of micellar systems containing surfactants A, B and C are, respectively, 2,937.0, 6,755.0, 4,309.0, 3,627.0, 1,667.0, 1,367.0, 4,190.0, 1,441.0, and 1,049.0. After about 390 minutes, the final counts for these various samples were, respectively, 2,876.0, 6,214.0, 3,440.0, 3,316.0, 1,143.5, 606.5, 4,020.5, 963.5 and 484.5. With respect to the systems containing surfactants A, B and C, the following is noteworthy. Chemiluminescence was noted in all surfactant containing samples. This chemiluminescence decayed rapidly at first, but then persisted for long periods of time. The backgrounds of the systems prepared with these surfactants after approximately 156 minutes of counting range from 800.0 to 5,500.0 counts per minute and with small amounts of added water as described above, after 390 minutes of counting, from 480 to 6,000 counts/minute.

The background rate increased with the amount of surfactant added to the system. When tritiated water was added to the systems, at best, counting efficiencies approached about 3%. In some cases, the counts per minute of the sample were lower after adding the radioactive material than they were before any radioactive tritium was added. Apparently, the small amount of water added to the system can quench the chemiluminescence process.

The two-phase control samples counted with approximately equal efficiency as the best systems prepared containing petroleum sulfonates.

EXAMPLE XIII

A counting sample was prepared containing 2.0 ml of 25.1%, by weight, tall oil fatty acids, 24.2% n-hexanol, 15.8% isopropanol, 22.1% isooctylphenol-polyethoxyethanol and 12.8% of an aqueous solution containing 25% NaOH and 15 ml of the toluene-fluor mixture of Examples I and X and 0.4 ml of water. Initial counting rates were 74.0, 55.0, and 54.0 counts per minute. 0.05 ml of tritiated water was added to the samples and counting rates of 34,207.0, 34,266.0, and 34,311.0 counts per minute were obtained. These rates represent counting at approximately 37% efficiency. One ml of a solution containing equal parts surfactant A and toluenefluor of Example X was added to the first of the three samples just counted. The color of the final mixture was approximately the same intensity of yellow as the original color of the surfactant solution. After one minute, the counting rate was 39,973.0 and after eleven minutes, it was 15,805.0. The counting rate decay with time is noticeable. Another 1.0 ml increment of surfactant A solution was added and counted. After one minute, the counting rate was 54,729.0 but had decayed, after six minutes, to 22,910.0.

In both cases, after adding the solution of surfactant A, the count rates were initially higher than that observed in the absence of the surfactant, but decayed rapidly. Given enough time, the petroleum sulfonate added almost completely destroys the counting ability of counting samples prepared from the micellar systems of this invention.

EXAMPLE XIV

A system was prepared containing 2.0 g of a C_{15} C_{18} mixture (28% 28% 28% 14%) of n-alkane sulfonates, 6.0 ml of water and approximately 2.0 ml of isopropa-

nol. Ten ml of the usual toluene-fluor solution was added. Background radiation for replicates of the surfactant systems are 38.0 and 42.0. Tritiated water (0.05 ml of material previously utilized) was then added. Counting rates of 6,010.0 and 6,066.0 were obtained.

In another sample prepared similarly but with approximately 2.5 ml of isopropanol, counts of 4,468.0, 4,402.0 and 4,362.0 were obtained on addition of 0.05 ml of tritiated water. This sample contained enough water to saturate the counting solution in the vial. These systems count at about 4.9 to 6.6% efficiency.

The background count on the above sample indicates that the sulfonate group alone does not account for the apparent chemiluminescence of the counting sample. In some way, however, it may add to the quenching noted in the low counting rates obtained. This is particularly true since several hours were spent working up the above formulation which counted the best of any of the alkane sulfonate systems counted.

EXAMPLE XV

The procedure of Example X was followed except that 0.05 ml of tritiated toluene was added to each sample. Specific activity of the tritiated toluene was 2.14×10^6 PM/ml on July 10, 1965. Approximately 98,000.0 counts per minute at 100% efficiency would be expected if corrected for decay of material standardized on July 10, 1965. Only counting sample containing 1 and 2 ml of surfactants A, B and C were counted here and were, respectively, 10,386.0, 17,716.0 and 10,325.0, and 3,914.0, 6,916.0, and 7,841.0. After about 440 minutes counting rates were, respectively, 7,677.0, 14,241.0, 7,042.0, 2,177.0, 3,882.5, and 2,005.5.

The control samples were initially between 53,000 and 60,000 and remained so at the end of the counting period.

Now having described our invention what we claim is:

1. In a process of counting in liquid scintillation counting devices using liquid scintillation counting medium containing surfactant; the step comprising forming the medium by admixing

an amount of at least one of scintillation solvent and scintillation solute sufficient to absorb emissions from isotopes and radiate the absorbed energy at device-detectable levels

with

an amount of a plurality of different surfactants sufficient to impart thermodynamic stability to the resulting medium on addition of sample and during which counting reacts to provide essentially no fluorescence, chemiluminescence or phosphorescence, the plurality of surfactants being selected from the group consisting of

a. a combination of preferentially oil-soluble surfactant and preferentially water-soluble surfactant;

b. a combination of anionic surfactant and nonionic surfactant;

c. a combination of cationic and nonionic surfactant; and

d. a combination of surfactant which acts as a dispersant and surfactant which acts as a surface-tension reducing agent.

2. The process of claim 1 wherein the micellar system includes a cosurfactant.

3. The process of claim 1 wherein a scintillation solvent sufficient to dissolve any undissolved scintillation solute is admixed with the micellar system and scintillation solute.

4. The process of claim 1 wherein a secondary scintillation solute is admixed with the micellar system and scintillation solute.

5. A micellar liquid scintillation counting composition comprising scintillation solute in small amounts effective to provide countable scintillation in the composition on addition of radioactive sample to the composition, scintillation solvent in amounts effective to permit interaction between the scintillation solute and emissions from the radioactive sample on addition of radioactive sample to the composition, and a plurality of surfactants selected from the group consisting of

a. a combination of preferentially oil-soluble surfactant and preferentially water-soluble surfactant;

b. a combination of anionic surfactant and nonionic surfactant;

c. a combination of cationic and nonionic surfactant; and

d. a combination of surfactant which acts as a dispersant and surfactant which acts as a surface-tension reducing agent

the surfactants being present in an amount in excess of that amount required to render the mixture thermodynamically stable and to reduce the vapor pressure of the dispersed phase liquid, the surfactants, scintillation solute, and scintillation solvent being substantially inert with regard to chemical reactivity or energy transfer which results in chemiluminescence, fluorescence, or phosphorescence.

6. The medium of claim 5 wherein the medium contains cosurfactant.

7. The medium of claim 5 comprising a higher molecular weight fatty acid, relatively water-insoluble alcohol, relatively water-soluble lower molecular weight alcohol and n-isoalkyl phenol polyethoxyethanol.

8. The medium of claim 5 comprising about 25% higher molecular weight fatty acid, about 25% n-hexanol, about 15% isopropanol, about 22% isooctylphenol-polyethoxyethanol, and aqueous NaOH to volume at a pH of about 7.

9. the medium of claim 5 comprising about 29% higher molecular weight fatty acid, about 28% n-hexanol, about 18% isopropanol, and about 25% isooctylphenol-polyethoxyethanol.

10. The medium of claim 5 comprising about 38% sodium dihexyl-sulfosuccinate, about 5% isopropanol, about 52–53% isooctylphenol-phenoxy polyethoxyethanol, and 4.8% water.

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