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NEUTRON SCINTILLATION DETECTOR

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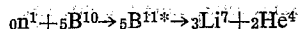
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The present invention is concerned with neutron counters and more particularly with compositions of matter suitable for employment as scintillation mediums in scintillation neutron counters or detectors.

The most widely used generic type of counter for atomic and nuclear particles is that based upon a combination of two electrodes in a gas. The passage of a charged atomic particle through the gas will result in ionization or the production of ion pairs. The positive ions will move to the negative electrode and the corresponding negative ions to the positive electrode, resulting in a pulse in the electrical system connected with the electrodes. Depending upon the method in which they are operated, these counters are usually termed an ionization chamber, proportional counter, or a Geiger counter. Since these counters only respond to charged particles, they cannot be used as neutron counters without modification.

The modification normally used is the introduction of a compound of a substance such as boron or lithium which reacts with neutrons to form charged particles. The isotope B^{10} (which comprises one-fifth of naturally occurring boron) reacts with thermal neutrons to form charged alpha particles according to the following reaction:



The boron is usually introduced into the chamber as a gas, for example BF_3 , but it may also be placed in the chamber as a coating on the inner surface of the chamber in the form of some suitable compound. The B^{10} responds primarily to slow neutrons or thermal neutrons, so that an additional modification must be made in the counter if it is to be used for counting fast neutrons. The usual modification is the surrounding of the counter with an hydrogenous material, coating or layer, so that the neutrons passing through the layer are slowed down to thermal energies. Now, while the reaction of B^{10} with neutrons has a large cross section, counters employing such a method have some very obvious disadvantages, perhaps the chief of which is the low efficiency of the counter. The method of determining the efficiency of BF_3 neutron counters is described in *Electron and Nuclear Counters—Theory and Use*—Korff, Van Nostrand, 1946, page 52. The maximum efficiency that may be expected for BF_3 -filled neutron counters is of the order of 10% and the maximum efficiency for boron-lined counters is of the order of 5%.

It is an object of the present invention to provide a neutron counting device having an efficiency approaching 100%.

It is an additional object of the present invention to provide compositions of matter suitable for use as the scintillator in a scintillation counter for neutrons.

We have discovered that a neutron detector, which is approximately 100% efficient, has a very short response time, and is relatively insensitive to gamma rays, can be constructed by employing novel compositions which are hereinafter described as the "scintillation medium" or "scintillator" in a scintillation counter. The novel com-

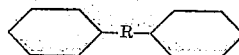
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positions which we have discovered have three components, a radiator, a phosphor, and a solvent. The radiator is the constituent of our solution which reacts with neutrons to produce charged particles. The charged particles thus produced then react with the phosphor and solvent to produce scintillations which are counted by a photomultiplier tube or other similar photosensitive device. The exact mechanism by which scintillations are produced has not been finally determined, but it is believed that the energy from the charged particles is first absorbed by the solvent molecules, and then at least partially transferred to the phosphor molecules; which in turn emit the energy in the form of light waves. In any event the choice of a solvent cannot be made solely upon solubility characteristics.

The phosphors which are employed in our scintillating mediums are aromatic or aralkyl compositions containing a large proportion of double bonds. Aliphatic borate compounds are used as the radiators and alkyl, aryl, and aralkyl liquid organic compositions are useful as the solvent.

While various fluorescent materials and solvents are known, the known materials cannot be combined indiscriminately to obtain a product useful as the detecting medium in a liquid neutron scintillation counter. There are certain optical, physical, chemical and electrical properties which are of importance in determining the usefulness of the final product. The mixture of phosphor, radiator and solvent must be clear and should be colorless and have a high transmission factor for visible as well as for ultraviolet light. The composition should be fairly dense and be comprised of atomic constituents having low atomic numbers. With respect to the chemical properties of the product, the three components preferably form a true solution and this solution should be stable over a considerable range of temperature. For example, the neutron detector medium in a scintillation counter is often cooled to a low temperature of the order of $-20^\circ C.$ during the cooling of the photomultiplier tube in order to reduce spurious and background counts and a scintillation medium in which one of the components will precipitate out at low temperatures is undesirable. While the compositions are usually used in the liquid form it is contemplated that they may be used as solid solutions. The response time of the scintillation medium should be fast, of the order of a microsecond or less. The magnitude of the response is important and discrimination between alphas and gammas is of the utmost importance. The efficiency of alpha detection must be considered and the spectrum of the light emitted is also important, since it should be of a suitable spectral range to couple with the phototube or other light-sensitive counting device. The final characteristics which determine the suitability of a mixture of phosphor, radiator and solvent for a neutron detector medium cannot be determined theoretically from a knowledge of the individual characteristics of the components forming the mixture, but depend greatly upon the interaction of the components of the mixture not only with respect to the substance used but also with respect to the proportions.

The phosphors which are employed in the compositions of the present invention are in general complex aromatic organic compositions having a large proportion of double bonds to the molecule. Preferable phosphors have the general formula



in which R represents a phenyl ring, a heterocyclic ring containing at least one double bond or a conjugated aliphatic system having not more than eight carbon atoms. Where R represents a phenyl ring the ring may be joined

to the outer phenyl rings by single bonds or may be "fused." Examples of phosphors in which R represents a phenyl ring include terphenyl, anthracene, and phenanthrene. R represents a conjugated aliphatic linkage in such compounds as diphenylbutadiene, diphenylhexatriene, and diphenyloctatetraene. An example in which R represents a heterocyclic ring is 2,5-diphenyloxazole. The phosphors which are preferable in particular compositions include 2,5-diphenyloxazole, terphenyl and diphenylhexatriene. Phosphors which may be used in particular combinations include the following:

Fluorene	Naphthalene
Carbazole (diphenyleneimine)	α -Naphthol
Phenanthrene	β -Naphthol
Anthracene	α -Naphthylamine
Diphenyl	Phenol
m-Diphenylbenzene (terphenyl)	Trans-stilbene
Diphenylbenzidine	Triphenylamine
Diphenylbutadiene	Dihydrocollidine
Diphenylhexatriene	Carotene
Diphenyloctatetraene	Diphenylacetylene
Fluoranthene	Melaniline

The solvents which are employed are organic compositions which are normally liquid at room temperature. The preferred solvents are aralkyl compositions having a single phenyl ring with an attached aliphatic group or groups, such as phenylcyclohexane and xylene. The commercial mixture of xylene isomers or the pure isomers may be used. Other organic solvents may be used in particular combinations. Such solvents include:

Aniline	Hexane
Benzene	Mesitylene
Benzyl alcohol	Napthalene monobromide
Benzyl ether	Pyridine
α -Chloronapthalene	Quinoline
Cyclohexane	Styrene
1,4-dioxane	Toluene
n-Heptane	

The radiators which are employed in our composition are aliphatic borates, in which the aliphatic groups are straight chain groups having not more than four carbon atoms. Trimethyl borate has been found to be the preferable boron composition because of the high boron per molecule atomic ratio. Triethyl borate, tripropyl borate, tributyl borate or aliphatic borates having mixed aliphatic groups may be used. While the boron content of the aliphatic borate may be natural boron (B^{10} , 18.8%, B^{11} , 81.2%), the boron enriched in B^{10} is preferred.

The solvent, for example xylene or phenylcyclohexane, is normally combined with the radiator, for example trimethyl borate, in a proportion of about one to one by volume. This ratio may be varied from about 25% solvent-75% radiator, to 75% solvent-25% radiator. This variation, however, may result in reduced pulse height and decreased temperature solubility range if the radiator is increased in volume at the expense of the solvent, or in reduced efficiency if the solvent is increased at the expense of the radiator. The scintillating material is usually added to the mixture of solvent and radiator in proportions of about 2 to 8 grams of scintillating material per liter of mixed radiator and solvent. Mixtures of two scintillating materials may be used and often a minor amount of a second scintillating material may be added to the composition to shift the spectrum of the emitted light to one more suitable for the particular photomultiplier tube used. For example, when the photosensitive device to be used is the 5819 photomultiplier tube and the main scintillator is 2,5-diphenyloxazole or terphenyl, a few milligrams per liter of diphenylhexatriene may be added to the mixture to shift the spectral response. Between 2 and 20 milligrams per liter of diphenylhexatriene is usually sufficient to shift the response to an im-

proved spectral region for detection by a 5819 tube. A particularly suitable liquid scintillator composition which may be used at a temperature as low as -10°C . is as follows.

(a) Equal volumes of trimethyl borate and phenylcyclohexane containing 4 grams per liter of para-diphenylbenzene (terphenyl) and 8 milligrams per liter of diphenylhexatriene. A second composition which gives larger pulse heights than does the first one and whose solubility characteristics are such that it can be used at a temperature as low as -20°C . is as follows.

(b) Equal volumes of trimethyl borate and phenylcyclohexane containing 4 grams per liter of 2,5-diphenyloxazole and 16 milligrams per liter of diphenylhexatriene. The spectral response of these two compositions is particularly suitable when they are used with a 5819 photomultiplier tube as the detecting device, and the diphenylhexatriene may be eliminated from the compositions if they are to be used with a scintillation detecting device which responds to a lower spectral range. The scintillation medium is placed in a suitable cell made of material or materials having low atomic numbers such as quartz or polystyrene. The cell is light-tight except for a transparent window which communicates with the photosensitive device. The light-tight coating may be made by placing the cell in an aluminum container, or coating the surface with aluminum foil or magnesia. The window of the cell is placed in conjunction with a photosensitive device such as a phototube or photomultiplier tube. Photomultiplier tubes such as the 1P21 or 5819 are particularly suitable photosensitive devices. These are high-vacuum phototubes containing a plurality of dynodes. The photocurrent produced at the cathode is multiplied many times by secondary emission occurring at successive dynodes. The equivalent noise input of such tubes is low at room temperature, but can be still further decreased by cooling such tubes to below room temperature, for example -10°C . The window of the tube is usually connected with the window of the cell by a light pipe made of an acrylic resin such as "Lucite." The pulses from the photomultiplier tube are counted with conventional electronic counting devices. Measurements of counting rates obtained with the above-described solutions (a) and (b) in counter cells approximately 2 inches in diameter and 1 inch long coupled with the 5819 photomultiplier tube connected to a suitable counter circuit have indicated that the efficiency of counting of the neutrons in the scattered thermal neutron flux of a heavy water reactor was approximately 100%.

The mean life for capture of neutrons in the energy region in which the $1/v$ law for boron is operative is approximately 0.42 microsecond if enriched boron is used in solutions (a) and (b) above, and approximately 2.2 microseconds if normal boron is used in the above compositions.

The gamma discrimination for counters containing solutions (a) and (b) above and operated with a counting mechanism biased to give the optimum counter response necessary for detection of boron disintegration pulses was found to be good. Both experiment and calculation indicated that the response of the meter used as a slow neutron counter to gamma rays (produced by the Compton effect) was approximately 2% per gram per square centimeter of the total response to neutrons and gamma rays.

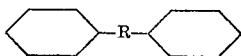
It will be understood that the compositions contemplated by this invention are not to be limited to those of the specific compositions described, but may be considered to include all those within the scope of the appended claims.

What is claimed is:

1. A neutron detection composition capable of producing scintillations, comprising a mixture of 25-75 weight per cent aralkyl organic solvent of the group consisting of xylene and phenylcyclohexane and an aliphatic borate in

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which the aliphatic groups are straight chain groups having not more than 4 carbon atoms, said mixture containing 2-8 grams per liter of an organic phosphor having the formula



in which R is a member of the group consisting of a phenyl ring, a heterocyclic ring containing at least one double bond, and a conjugated aliphatic chain having not more than 8 carbon atoms.

2. The composition of claim 1 in which the aliphatic borate compound is trimethyl borate.

3. The composition of claim 1 in which the aliphatic borate compound is triethyl borate.

4. The composition of claim 1 in which the phosphor is terphenyl.

5. The composition of claim 1 in which the phosphor is 2,5-diphenyloxazole.

6. The composition of claim 1 in which the solvent is phenylcyclohexane.

7. The composition of claim 1 in which the solvent is xylene.

8. A neutron detection composition capable of producing scintillations, comprising a mixture of 25-75 weight per cent phenylcyclohexane and trimethyl borate, contain-

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ing 2-8 grams per liter of said mixture of 2,5-diphenyloxazole and 2-16 milligrams per liter diphenylhexatriene.

9. The composition of claim 8 in which the boron of the trimethyl borate is enriched in the boron isotope B¹⁰.

10. A neutron detection composition capable of producing scintillations, comprising a mixture of 25-75 weight per cent xylene and trimethyl borate, containing 2-8 grams per liter terphenyl and 2-16 milligrams per liter diphenylhexatriene.

11. A neutron detection composition capable of producing scintillations, comprising a mixture of 50 weight per cent phenylcyclohexane and 50 weight per cent trimethyl borate, said mixture containing 4 grams per liter of 2,5-diphenyloxazole and 16 milligrams per liter of diphenylhexatriene.

12. A neutron detection composition capable of producing scintillations, comprising a mixture of approximately 50 weight per cent 2,5-diphenyloxazole and 50 weight per cent trimethyl borate, containing 4 grams per liter of said mixture of terphenyl and 8 milligrams per liter of said mixture of diphenylhexatriene.

References Cited in the file of this patent

Review of Scientific Instruments, July 1951, vol. 22, No 7, page 543.