

July 16, 1957

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2,799,780

NEUTRON SCINTILLATION COUNTER

Filed Sept. 3, 1953

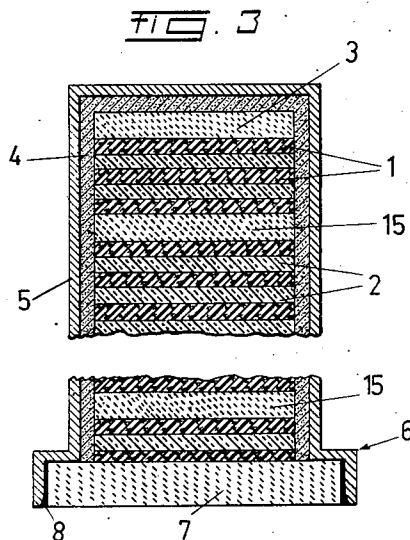
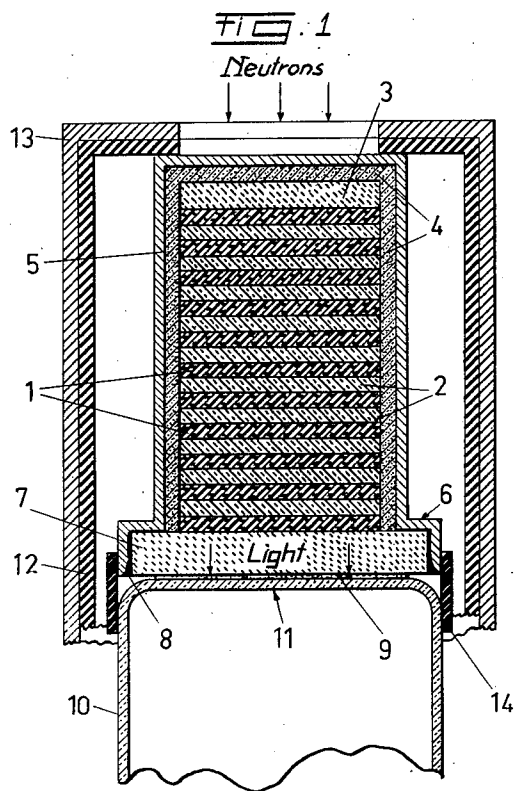
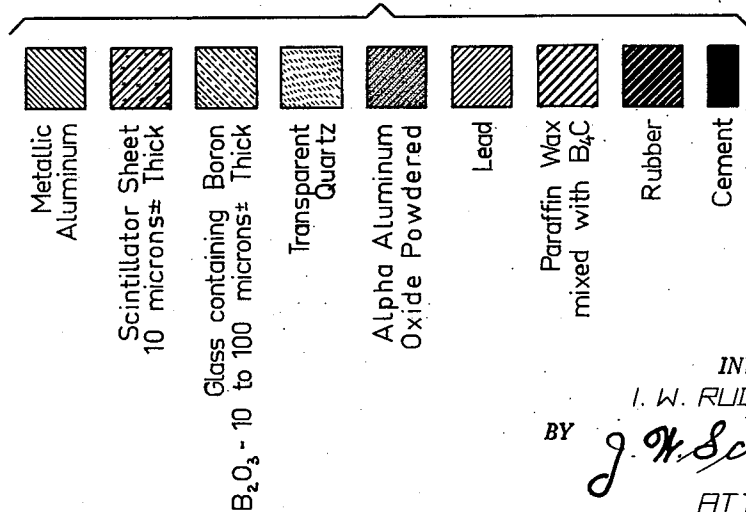


FIG. 2



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NEUTRON SCINTILLATION COUNTER

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Application September 3, 1953, Serial No. 378,381

20 Claims. (Cl. 250—71.5)

This invention relates to methods of and means for the detection and counting of neutrons and more particularly to the counting of neutrons by methods and means which enables effective discrimination against gamma rays which frequently are present with neutrons, and still more particularly to the counting of neutrons by means and methods which combine the advantages of being effective, discriminating against gamma rays, i. e. enabling neutrons to be counted separately, at least to a very large extent, from gamma rays coexisting therewith and accomplishing these results by apparatus of long life and small size.

Before discussing the subject matter comprising the invention the practices of the prior art and their insufficiencies will be reviewed.

The detection of neutrons is conventionally accomplished by means of a proportional counter filled with gaseous boron trifluoride. A neutron captured by a boron atom in the counter causes the ejection of an alpha particle having an energy of 2.78 Mev. 93% of the time and 2.40 Mev. 7% of the time. The alpha particle being a heavy ionizing particle produces a discharge, and a rapid sequence of the resulting pulses can be amplified and detected with a conventional scaler. Because the boron isotope with a mass of 10 is responsible for the high stopping power of boron for neutrons and normal isotopic boron contains only 18.4% B¹⁰ and 81.6% B¹¹, it has become usual to fill counters with BF₃ in which the boron is all of mass 10. This results in an increased counting efficiency. Coating the walls of the counter with a thin layer of B¹⁰ is also used to increase the counting efficiency of BF₃ counters by increasing the number of B¹⁰ atoms in the counter. The layer of B¹⁰ must be thin, however, because the alpha particles liberated on neutron capture have only a short range and must leave the boron layer if they are to produce a discharge in the counter. Increased pressures of B¹⁰F₃ have also been used in order to increase the number of boron atoms in the counter. In spite of these improvements the B¹⁰F₃ counter retains the disadvantages of large bulk and of relatively low efficiency, particularly for epithermal neutrons, generally of energy greater than ½ ev.

Scintillation crystals containing boron or lithium have been proposed for the detection of neutrons. While Li⁶ has only 23% of the stopping power for neutrons as does B¹⁰, a crystal containing Li⁶ offers the possibility of a highly efficient detector for neutrons. Bernstein and Schardt, in the Phys. Rev. 85, 919 (1952), and Schenck and Heath in the Phys. Rev. 85, 923 (1952), and Schenck in Oak Ridge National Laboratory Report 1365 issued January 9, 1953, have shown that lithium iodide crystals activated with tin or europium are useful neutron detectors. These crystals for this purpose suffer the disadvantage, however, of high sensitivity to gamma radiation. Because there is almost always a high background of gamma radiation in the vicinity of a neutron source, activated LiI crystals are useful only if the gamma pulses can be sorted out from the desired neutron pulses. Lithium iodide crystals activated with europium are now com-

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mercially available and show promise as neutron detectors.

Little success has attended many attempts to utilize boron-containing scintillating detectors. Boric acid glass mixed with an organic phosphor such as anthracene, or a mixture of boric oxide and a zinc sulfide phosphor have been proposed by Sun and Shoupp (Rev. Sci. Inst. 21, 395, 1950), but these detectors when used with presently known electron multiplier tubes and known circuits give only small pulses for neutrons as compared with pulses due to gamma background counts. They also have low efficiency because of poor transparency to the fluorescent radiation emitted.

Liquid scintillators containing the phosphors p-terphenyl and diphenylhexatriene dissolved in a solvent such as phenylcyclohexane to which is added an equal volume of boron esters such as methyl borate have been proposed by Draper in the Rev. of Sci. Instruments 22, 543 (1951), and Muehlhause and Thomas in the Phys. Rev. 85, 926 (1952), as neutron detectors but these counters are highly sensitive to gamma radiation if the active thickness is made big enough to be efficient for neutron detection.

In achieving the object of this invention to provide a scintillation detector of small compact size having high sensitivity to neutrons and low sensitivity to background gamma radiation, advantage is taken of the fact that an alpha particle released when a neutron is captured by a boron nucleus has an energy of 2.78 Mev. and can travel only a short length of the order of 9 microns (1 micron=1 mm. $\times 10^{-3}$) in aluminum before being stopped, while a gamma ray in traversing matter will produce one or more electrons whose range depends upon their energy but in general is much longer than the range of an alpha particle. For example, let us suppose a 2.8 Mev. gamma ray passes through matter and ejects a photoelectron of the same energy. This electron will have a range of approximately 5000 microns in aluminum.

The range of an alpha particle of given energy in a phosphor such as described is generally of the order of its range in aluminum. Alpha particles produced by capture of a neutron by elements other than boron will have other energies and therefore other ranges; the thickness of the phosphor sheet may be adapted to the energies of the particular classes of alpha rays resultant from the particular capturing elements employed.

Further description of the details of exemplary embodiments of the invention will make reference to the accompanying drawings, in which:

Fig. 1 is a cross sectional showing, somewhat diagrammatic, of the essential elements of the detector and scintillation creating apparatus of the invention;

Fig. 2 is a ready reference cross section chart; and

Fig. 3 is a second embodiment also shown in cross section from which the outer shielding of lead and paraffin plus boron carbide powder already shown in Fig. 1 are omitted; Figs. 1 and 3 do not purport to be drawn to scale either as to relative height and width or as to number of sheets.

An exemplary embodiment of the invention as indicated in Fig. 1 comprises a stack of alternate sheets of extreme thinness consisting of a boron-containing glass 2 and a transparent fluorescent sheet 1 of plastic material. This stack may be bounded and protected at each end by a quartz sheet or plate 3, 7 which may be one or two millimeters thick, one end serves for the ingress of neutrons to be counted and the other is fitted to an electron multiplier tube 10 of the photo-cathode type. The stack is surrounded on the sides and ingress end with powdered alpha-aluminum oxide (Al₂O₃) designated 4 then in turn by an aluminum can 5. The can 5 may have an enlarged end furnishing a shoulder 6 into which the end plate 7 of quartz or equivalent may be sealed with cement 8 to maintain the consolidated assembly of sheets firmly

in place. The can may be shielded around the sides by one or more layers each of lead 13 and paraffin wax 12 mixed with boron carbide (B_4C), the first of which is a shield for gamma rays and the second for neutrons because better results will be secured with neutrons passing through the sheets of the stack rather than parallel with the sheets. The number of sheets may be quite considerable and numerous variant forms are possible. In one of these (Fig. 2) as will be described below quartz sheets of a thickness such as 100 microns may be interspersed at intervals such as each 10 to 25 or other number of the plastic and glass sheets.

The detector therefore consists of alternate layers or sheets of a transparent scintillator 1 and a transparent boron-containing substance 2. The thickness of each layer is or may be approximately 10 microns i. e. 10^{-2} millimeters. The scintillator is preferably a transparent plastic phosphor. A plastic scintillator suitable for the purpose may be prepared by polymerizing a solution of vinyltoluene monomer containing 3% of p-terphenyl and 0.02% of 1,1',4,4' tetraphenylbutadiene-1,3. This phosphor when properly made has a luminescent efficiency approximately 40% that of a fine anthracene crystal as measured by the relative integrated light output from a photo-multiplier tube sold by the Radio Corporation of America under the code No. 5819 for the photo electric peak from Cs^{137} . Such fluorescent material can be drawn out while hot and cooled into very thin sheets such as are required for present purposes.

There are numerous compositions which are qualitative equivalents for the purpose and these include such substances as may be polymerized or otherwise caused to become solids transparent to their own spectral radiation produced by scintillation when having dissolved or otherwise suitably distributed therein small percentages, in the general range of $\frac{1}{10}$ to 5%, of any one or a combination of numerous fluorescent or activated organic or inorganic materials and which may be drawn into or otherwise formed into adequately thin stackable sheets. Reference is made to U. S. Patent application Serial Number 304,119 filed August 13, 1952.

The boron-containing sheets 2 may consist of a borosilicate or any transparent boron-containing glass which contains approximately 13% by weight of boron as B_2O_3 . Such glass is sold as "Pyrex" a registered trademark of the Corning Glass Works, Corning, N. Y. for a borosilicate glass. Thin sheets of such glass can be prepared by blowing large thin bubbles therefrom and cutting the bubbles into the desired sheets. Any other transparent boron-containing sheet may, however, be used, if of the necessary thinness.

Although uniformity is preferred it is not necessary that the thickness of either the scintillator sheets or of the glass be precisely uniform as considerable variations in the same sheet or same stack are permissible. The sheets need not necessarily be flat but may be curved or concave but preferably not to the extent of offering a path parallel to any part of their surface to the impinging radiation.

The total number of sheets employed in a stack will depend upon the counting efficiency desired but may be considerable. For example, 100 sheets would be only one millimeter thick, exclusive of quartz end plates. It is desirable to protect the ends of the assembly with a thin quartz plate 3 or 7 at each end. The shape and size of the sheets will depend upon the particle counting requirements, but in general they will be circular or square in cross-section.

The stacked assembly of sheets may be held together by means of a transparent cement, for example, polystyrene or poly-methyl-methacrylate applied at the edges of the sheets, preferably while the assembly is somewhat

compressed, so that a compact mass is obtained. The edges and one face are covered and surrounded with a reflecting material described above as alpha-aluminum oxide powder 4 but which may be replaced by polished aluminum foil, magnesium oxide powder, or any other suitable reflector for the light emitted by the scintillator.

A neutron entering the assembly of sheets shown in Fig. 1 has a good probability of being captured by a boron atom in a glass sheet 2 with the ejection of an alpha particle having an energy of 2.78 Mev. This alpha particle will in most cases travel out of the boron-containing sheet and strike the transparent phosphor sheet 1, giving rise to a light flash or scintillation. This light can traverse the transparent assembly of sheets and strike the photocathode surface of the photomultiplier tube thereby producing an electrical pulse which can be amplified and counted on a conventional scaler. Pulses due to alpha particles of 2.40 Mev. are neglected as they are relatively few in number and the difference between 2.78 Mev. and 2.40 Mev. is considerable.

A background gamma ray striking the detector can eject one or more electrons anywhere along the path of the gamma ray. However, since the range of these electrons is very much longer than that of the alpha particle emitted by the boron atom capturing a neutron, these electrons will pass through many layers of boron-containing sheets 2 and scintillator sheets 1. However, only the energy dissipated in the scintillator sheets will produce scintillations; the energy dissipated in the boron sheets is converted to heat and does not appear as light. Consequently, the magnitude of pulses produced by gamma rays in an associated output circuit will be considerably reduced. Of occasional electrons passing laterally or almost laterally through a scintillator sheet this will not be true but these are few in number.

To effect even greater discrimination against gamma rays two courses may be pursued. The boron-containing sheets may be made much thicker than the scintillator sheets. With increased thickness an increased part up to nearly all of the energy of electrons produced by the gamma rays will then be dissipated in non-phosphor material. For example, the boron glass sheet may range up to possibly 10 times the thickness of the scintillator sheets. These would still be thin sheets of only $\frac{1}{10}$ millimeter thickness. The relative thickness will be such as will give the desired discrimination against gamma rays consistent with the wanted efficiency in counting neutrons because this will involve some loss in efficiency since some alpha particles will be unable to penetrate the boron-containing sheet and reach the scintillator; these alpha particles will therefore not be counted. However, in some applications it may be more important to eliminate or greatly reduce the gamma background than to count all the neutrons.

Figure 3 illustrates a means effective for a second method of reducing the gamma background that does not involve any loss in neutron counting efficiency. The structure involves the interposition at regular intervals in the assembly of boron-containing sheets and scintillator sheets with other sheets 15 of a barrier material that is transparent both to the light emitted by the scintillator and to neutrons. $CaCO_3$ crystals would do but they are expensive. Quartz is an example of a material that meets these requirements. The barrier sheets 15 act as a medium for degrading electrons produced by gamma rays without the emission of light. Let us take, as an example, an assembly consisting of alternate 10 micron thick sheets of a boron-containing material 2 and a scintillator 1 with a 10 micron thick sheet 15 of quartz interleaved after every sixth boron sheet. Most of the 2.78 Mev. alpha particles generated in a boron sheet can easily reach a transparent plastic scintillator sheet and give rise to light flashes. A stray gamma ray of the same 2.78 Mev. energy may produce a 2.78 Mev. photo-

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electron anywhere in the assembly. However, this photo-electron has a range of approximately 5000 microns, and will hence dissipate most of its energy in the quartz and the boron sheets. The energy dissipated in the scintillator will be very small and hence only a small amount of light will result and only a small current output pulse will be obtained from the photo-electron multiplier tube. These pulses can be readily distinguished from the much larger neutron pulses by means of a properly biased amplifier.

The thickness of these quartz barrier sheets and the relative number used will depend upon the background conditions under which the counter is to operate.

The bottom end of the assembly of Fig. 1 is indicated as coupled by any suitable means such as a layer of high polymer silicone or by direct light transmission through air to the photo-cathode window 11 of a photo-multiplier tube 10. This coupling may be by conventional means and a rubber ring 14 or equivalent means may surround the junction of the assemblage and the whole or any necessary part of the tube to keep out light when the device is used in an illuminated place.

These elements are not disclosed in Fig. 3 but such or their equivalents are implied.

Suitable tubes 10 are commercially available and because their output circuits, associated amplifiers, scalars or count indicators are commercially available and their modes of use are known, they will be implied and not illustrated or described.

In operation the neutrons enter the stack at the top and most of the alpha rays produced by their capture will enter an adjacent sheet 1 and produce a scintillation whereas most of the energy of electrons produced by gamma rays will be consumed in intervening sheets 2 whereby the scintillating light energy produced by them will be greatly reduced. The resultant light will be collected by the reflecting layer 4 and caused to act on the photo-cathode 11 of the tube 10 which is selected to be sensitive to at least one specific range or ranges of spectral radiation produced by the particular phosphor employed. For better light collection the upper part of the stack may be dome shaped or paraboloid and the surrounding layer 9 will correspond.

The foregoing description is based on boron as a preferred capture element for neutrons. Lithium, cadmium and indium form a group which, when included in a transparent body such as a lithium silicate, operate in a similar manner and to the extent that they or their mixtures may be formed into transparent sheets of the required or suitable thinness not only these metals but the rare earth metals such as gadolinium or their mixtures are qualitatively similar. Many of these are now too expensive or rare or both and are not readily available. Their rarity or excessive cost does not eliminate them as equivalents; obviously, all these elements are not quantitatively equally effective.

What is claimed is:

1. A neutron detector consisting of a multiplicity of stacked sheets of a transparent material having the characteristic property of scintillating with emission of radiation in or adjacent the visible range, when traversed by alpha particles, interspersed with sheets of transparent material containing material of the group consisting of boron, lithium, cadmium, indium and rare earth metals, said first named sheets having a thickness in the general range of the maximum distance which an alpha particle produced by capture of a neutron of the general range of energies of the neutrons to be detected will have when produced by the capture of such a neutron by the particular contained material of the said group will traverse, and said second named sheets having a thickness ranging from about one to ten times the thickness of said first named sheets.

2. A neutron detector as per claim 1, in which the

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thickness of the first named sheets is appreciable but less than 20 microns.

3. A neutron detector as per claim 1, open for the impingement of nuclear including neutrons radiation in a direction generally transverse to the sheets but shielded against impingement of such radiation in a direction approximating parallelism with the sheets.

4. A detector as per claim 1, having a photo-cathode of a light sensitive space discharge tube coupled for spectral radiant energy transmission from the stacked sheets in a direction transverse to the sheets and shielded from stray spectral radiant energy.

5. A detector as per claim 1, having a non-scintillating plate transparent both to light and to neutrons at the ends of the stack.

6. A detector as per claim 1 having a quartz plate at each end of the stack.

7. A detector as per claim 1, having a photocathode of a light sensitive electronic space discharge tube coupled for light transmission to the stacked sheets through means including an intervening quartz plate.

8. A detector as per claim 1, having the edges of the stack of sheets cemented to form a compact mass including the plates.

9. A detector as per claim 1, having the edges of the stacked sheets and one end of the stack presented to a body of high reflecting power for radiation in or near the visible range.

10. A detector as per claim 1, contained in a metallic can-like container open at one end.

11. A detector as per claim 1, in which the second named sheets of the stack consist of silicates.

12. A detector according to claim 1 in which the first-named sheets are composed of a solid organic transparent body activated for scintillation by the inclusion of activating material.

13. A detector according to claim 1, having interleaved between the sheets an occasional thicker sheet of material transparent to light and to neutrons which when invaded by traveling electrons consumes their kinetic energy without production of spectral radiation.

14. A detector according to claim 13 in which the occasional sheets are composed of quartz.

15. A detector according to claim 1, in which the stack of sheets is surrounded on the sides in turn by highly reflective material, a metal wall of a container, and any number of layers each of heavy metal of the class of lead and a shielding of high stopping power for neutrons.

16. A detector for nuclear radiation comprising at least several alternately stacked sheets of transparent material containing metal of high stopping power for neutrons interleaved with sheets of transparent scintillating material, said first named sheets being of an order of thickness not over 100 microns and said second named sheets being of an order of thickness not over 20 microns.

17. A detector according to claim 16, in which the metals are of the group consisting of boron, lithium, cadmium, indium and their mixtures.

18. A detector according to claim 16, in which the metal is boron.

19. A detector according to claim 16, in which the first named sheets consist largely of a boron silicate.

20. A neutron detector consisting of a multiplicity of stacked sheets of transparent material scintillating with spectral emission when traversed by alpha particles, interspersed with sheets of transparent material containing material of the group consisting of boron, lithium, cadmium, indium, and rare earth metals and mixtures thereof, said second named sheets being of the order of 5 to 100 microns in average thickness and said first named sheets being of any thickness appreciably above zero but generally on the average not in excess of the thickness of said first named sheets.

(References on following page)

References Cited in the file of this patent

UNITED STATES PATENTS

2,186,757	Kallmann et al. -----	Jan. 9, 1940
2,272,375	Kallmann et al. -----	Feb. 10, 1942
2,274,900	Kallmann -----	Mar. 2, 1942
2,550,610	Smith et al. -----	Apr. 24, 1951
2,666,145	Eversole et al. -----	Jan. 12, 1954

OTHER REFERENCES

"Two liquid scintillation neutron detectors," Muel-
haue et al., Nucleonics, January 1953, pp. 44-45.
5 "Fluorescent liquids for scintillation counters," Kall-
man et al., Nucleonics, March 1951, pp. 32-39.