Boron Nitride, A Neutron Scintillator With Deficiencies

R. Engels, *Member, IEEE*, G. Kemmerling and J. Schelten

***Abstract*–During an electron beam evaporation accidentally sintered boron nitride was discovered to be a bright scintillating material [1]. The high density boron nitride HDBN of Henze [2] showed most promising results concerning light emission due to 8 keV electron radiation, due to bombardment with 5 MeV alpha particles, and due to 240 nm UV radiation. The light pulses generated in HDBN are similar intense as those generated in the well known ^{6}Li glass scintillator. Because of the extremely strong absorption of thermal neutrons and the weak interaction with MeV gammas the material is an exciting new inorganic scintillator candidate for the detection thermal and epithermal neutrons. A series of properties of HDBN was measured with various methods and evaluated. Many results are in favour an intended use of HDBN as scintillator. A few observations reveal deficiencies of this new scintillator and ask for further investigations.**

I. Introduction

A material which heavily absorbs thermal neutrons and which emits a bright light pulse due to such an absorption event can be utilized as scintillator for the detection of neutrons. The scintillators are optically coupled with photomultipliers or semiconductor diodes which then generate an electrical pulse for each absorbed neutron. For a reliable, position sensitive detection with high spatial resolution a series of properties must exist. They are listed in the following:

A. Absorption probability

A well designed scintillator should stop thermal neutrons with a probability of 70%. At this level the ratio of signals generated by thermal neutrons to noise generated by fast neutrons is optimised.

B. Colour of scintillation light

For blue light with a wave length of about 400 nm the light guidance as well as the generation of photo electrons and of ion pairs in photomultipliers and photodiodes, respectively, are performed most efficiently.

C. Number of photons per absorbed neutron

If a neutron is absorbed in ^{6}Li, ^{10}B, and ^{3}He nuclei an energy of 4.78 MeV, 2.31MeV, and 0.78 MeV is released, respectively. Only a small fraction of typically less than 1% is converted into light energy. The vast majority of energy dissipates as heat. At least 1000 photons per neutron should arrive at the photo multiplier or photo diode in order that a well defined event position can be deduced.

D. Duration of light pulses

The decay time of a generated light pulse should be not more than 100ns in order that pile-up effects can be ignored even for pulses which rapidly follow each other. In addition, such short pulses do not affect the temporal time structure as determined by choppers and pulsed sources.

E. Optical transparency

For optically transparent scintillators one can expect sharp light pulses. In non-transparent scintillators the pulse height spectra become broad due to multiple reflection and self absorption. With very broad spectra neutron pulses occur which fall beyond a lower level discrimination threshold and are not counted.

F. Gamma insensitivity

In scattering experiments with thermal neutrons a strong flux of gamma quanta with energies in the MeV region cannot be avoided. This flux is generated in the neutron source, which might be a fission reactor or a spallation source; it is also generated when the primary beam is monochromatized, pulsed, collimated, guided and scattered in the sample. A residual sensitivity of 1ppm seems to be tolerable, a value of 100 ppm is not acceptable as was experimentally verified quite often. Usually low gamma sensitivity of scintilators is achieved by pulse height discrimination. Attempts to obtain additional gamma discrimination by pulse shape analysis were successful in special cases with a huge amount of electronic equipment.

G. Light background

A continuous weak after-glow in the scintillator material leads to a strong wing at low pulse height in the pulse height spectrum. There, the pulse rate decreases exponentially and rapidly towards zero with increasing pulse height. Usually a discrimination threshold against gamma rays is set so high that it completely repels these signals due to this light background.

H. Large detector areas

Scattering experiments can profit from the circumstance that the sources suffer from weak neutron fluxes but offer big source areas. The profit can be orders of magnitude in scattered intensity provided large area detectors are available. Large means that the detector cross sections are of the order of 100 to 10000 cm^2. So far the largest scintillation detectors are not more than 2500 cm^2.

Manuscript received November 7, 2005.

R. Engels, and G. Kemmerling with the Zentralinstitut für Elektronik; Forschungszentrum Juelich GmbH, 52425 Juelich, Germany (email: r.engels@fz-juelich.de; g.kemmerling@fz-juelich.de).

J. Schelten is with the Institut für Schichten und Grenzflächen, Forschungszentrum Juelich GmbH, 52425 Juelich, Germany (email: j.schelten@fz-juelich.de)

I. Scintillator cost

A discussion of the cost-to-benefit relation for neutron detectors asks for large area detectors. In view of this, a scintillator of a low specific price (cost-per-unit area) is extremely welcome.

A scintillator material which simultaneously meets all these nine requirements does not exist. The following brief discussion demonstrates, that only very few scintillator types are useful for neutron detection.

1) Li glass

This is essentially the only scintillator which has been used in large area scintillation detectors for thermal neutrons. The neutron absorption takes place in ^{6}Li nuclei. By blending ^{6}Li-enriched Li_2O into the glass typical to an amount of 18% by weight about 1mm thick scintillator plates are able to stop thermal neutrons with the required probability of 70%. The product is commercially available from Applied Scintillation Technologies [3].

2) LiGdBorate

A brighter neutron scintillator was developed by Photogenics, Salt Lake City, Utah, USA. The material is ^{6}Li^{158}Gd^{11}Borate.It is the role of the isotope ^{6}Li to absorb thermal neutrons,while the isotopes ^{158}Gd and ^{11}B are chosen to prevent neutron absorption by Gadolinium and Boron. The crystalline borate powder is bedded into an epoxy resin which has the same refractive index as the borate [4]. Due to such a match an optical transparency should be achieved. In an ultimate step, the epoxy resin must be deuterated in order to avoid the strong incoherent neutron scattering of hydrogenous materials. The development of this scintillator was subsidized for more than ten years. Meanwhile the financial support has stopped [5]. So far the scintillator was used once in a large area scintillation detector at ISIS because of its intense light response on a neutron absorption event.

3) Li-Iodide

A scintillator with excellent physical properties is single crystalline ^{6}Li-iodide. A few single crystal disks of 4cm diameter which are encapsulated by glass do world wide exist.

For a long time no further LiI single crystals were grown and no new ^{6}LiI neutron scintillators were on the market.

4) BC523A

This liquid scintillator is commercially available from Bicron, now Saint Gobin, Crystals and Detectors [6]. The neutrons are absorbed by the ^{10}B-enriched boron component of the organic liquid. The liquid is optically transparent and chemically instable. Its light output is weak. Again, the hydrogenous organic molecules must be deuterated in order to get rid of the strong incoherent neutron scattering.

5) Other scintillators

There are more organic liquids and small single crystals which absorb neutrons to some extend and scintillate upon ionising radiation. In some respect they have fantastic properties, however, they exist only as laboratory samples and are used for scintillator studies only.

More information is given by C.W.E.van Eijk [7].

In view of the nine requirements which are described in the first part of this chapter all five scintillators listed above have more or less severe deficiency.

The main disadvantage of the Li glass (1) scintillator is its insufficient gamma insensitivity which is in the order of 100 ppm for MeV gammas. One would like to have more intense light pulses in order that there are more than the measured 2000 photons per neutron absorption event at the photo cathode which directly influences the spatial resolution in Anger type detectors [8]. In addition, the scintillator cost with 3000 EUR / 100 cm^2 is large.

For the ^{6}Li^{158}Gd^{11}Borate scintillators the deficiencies are more severe. The poor transparency causes broad pulse height spectra in order that a pulse height discrimination against gamma quanta and electronic noise affects already the detection of neutrons. The used Gd isotopes ^{158}Gd and ^{160}Gd are extremely expensive and they are available in only small quantities. A homogenisation of the powder/resin mixture was not successful on microscopic scale. Financial support of further developments had come to an end and a commercial use is not foreseen.

The powerful semiconductor industry has lead to single crystalline silicon waver with 300mm diameter. A similar development is not expected for lithium iodide. It is more likely that the growth of ^{6}LiI single crystal has stopped for ever and that the development of neutron scintillation detectors must look after alternatives of ^{6}LiI scintillators.

With liquid scintillators the light signals are too small, the pulse height spectra are too broad and the differences in pulse heights between neutron and gamma signals are too little.

In this context a new inorganic scintillator is introduced. The scintillation property of high density boron nitride (HDBN) which is fabricated by sintering BN powder at high isostatic pressure and elevated temperature was discovered fortuitously by Angelika Pracht while she was making gold pads on HDBN plates by electron beam evaporation. The gold pads were used for transport measurements in HDBN. A day later a piece of Li glass scintillator was mounted next to a HDBN sample in the electron beam evaporator. With the electron beam switched on but the shutter still closed both samples emitted light visible with the bare eyes. The HDBN piece appeared brighter than the well known ^{6}Li glass scintillator. The scintillation in both samples was probably caused by X ray Bremsstahlung which is generated in the electron beam heater and which circumvented the shutter above the evaporation source. The observation stimulated a lot of experiments in order to get to an evaluation HDBN as neutron scintillator.

It is the purpose of this paper to describe these experiments and to evaluate the results. This ends with a list of favourable properties of HDBN, with some recognized deficiencies for using HDBN as neutron scintillator and a few suggestions how to proceed.

It is hoped that the presentation of these preliminary results will lead to some stimulating discussions and fruitful cooperation.

II. Experimental Results

In a Scanning Electron Microscope SEM the cathode luminescence detector was used to measure the spectral distribution of the emitted light as generated by the radiation with 8 keV electrons in HDBN and Li glass. In both cases well pronounced intensity peaks accrued at 400 nm with a FWHM of ~100 nm due to this near-surface excitation. The peak heights of both scintillators were also comparable.

In the next experiment the two materials were radiated by 5 MeV alphas from a radioactive ^{241}Am source in front of a photo multiplier. The near-surface excitation generates light pulses which are half the size in HDBN compared to Li glass. The pulse height distribution is peaked in both scintillators, however in the transparent Li glass the distribution is much sharper. Fig. 1 shows the alpha peak as measured with the HDBN.

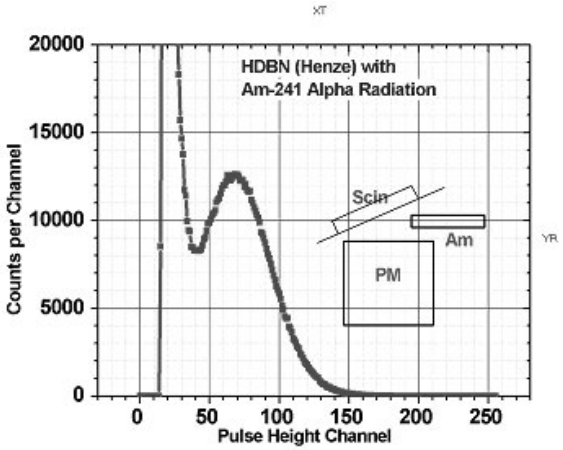

Fig. 1 Pulse height spectrum caused by alpha radiation of HDBN in the indicated 45° geometry

Because of the non-transparent HDBN the 45° geometry was chosen. The strong wing at low pulse height is an after-glow effect.

Day light exposure leads to the strong after-glow which is visible by eye many hours later in the dark. The decay of this after-glow occurs with different rates over hours, days, and even over weeks.

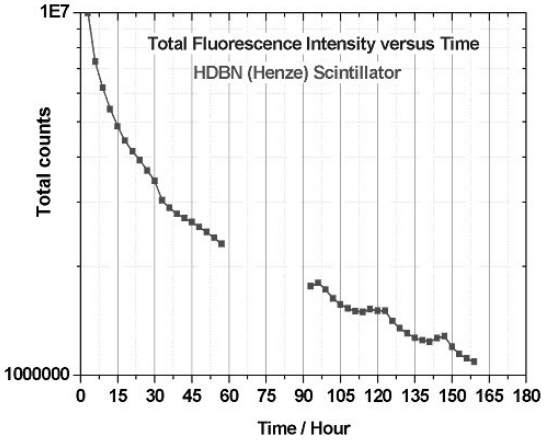

Fig. 2 Long time decay of after glow in HDBN

Fig. 2 shows the long time behaviour as measured with a photo multiplier.

The temporal width of a light pulse due to an alpha trace is very short, a half width of the temporal peak is only 2.5 ns as indicated by the oscilloscope diagram in Fig.3.

An annealing of HDBN at 500C for 1 hour destroys the after-glow completely.

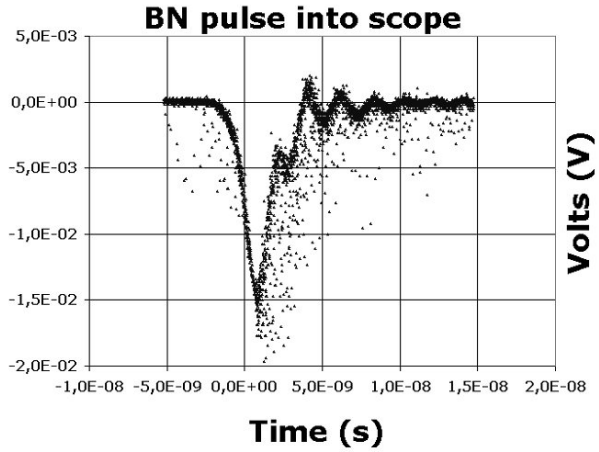

Fig. 3 Temporal pulse shape as measured with a digital storage microscop

Further experiments were done with frequency-doubled laser light of 5.1 eV energy and 240 nm wave length. The source power was 17mW. The main results are:

1. HDBN fluoresces orders of magnitude stronger than BN powder which is the starting material of HDBN
2. HDBN fluoresces similarly strong as Li glass or ordinary quartz glass
3. Part of the light generated in HDBN decays very slowly within hours or even days. Such behaviour was not observed in other substances.

Some properties of boron nitride are compiled in Table 1. An extremely favourable property of HDBN is its efficient neutron absorption. HDBN with ^{10}B-enriched boron has an absorption length of 50 µm. The ranges of the two nuclei, which are released when a neutron is absorbed, are 3.9 µm and 2.15 µm. HDBN is a low Z material with 12 electrons per unit cell containing a boron and nitrogen atom. The scintillation light has an energy of ~3 eV which is smaller than the gap energy. Thus there is no self absorption in pure BN. Pure crystalline BN has a gap energy of ~7.5 eV [9] and is probably an indirect semiconductor.

From a full chemical analysis no components were found which could play the role of activation centres. Contaminants were found only in the ppm range. In other inorganic crystalline scintillators such activation centres are deliberately created with additives grown into the crystals. In the case of HDBN it is more likely that activation centres are formed by lattice defects in the sintering zone near the surface of the small crystalline BN grains. There the lattice periodicity is gradually distorted which leads to a reduction of gap energy. Without a drastic reduction of the gap energy one would not reach excited states by 240nm UV radiation. In addition the defects form localized lattice states where the transition from an excited state to its ground state can occur more likely via the emission of light.

TABLE 1
DATA OF BORON AND BORON NITRIDE

Boron Isotope		^{10}B	^{11}B
Abundancy		20%	80%
Absorption cross section for 2200 m/sec (thermal) neutrons	σ	3835 barn	0.0055 barn
Molecular weight of Boron Molecular weight of BN	M_B M_{BN}	10.80 24.80	
HDBN mass density	ρ	2.10 g/cm3	
Atomic number density	n	$0.051\ 10^{24}\ cm^{-3}$	
Absorption coefficient	μ	$195.5 cm^{-1}$	
Scintillator thickness for natural boron nitride	d	255µm	
Scintillator thickness for ^{10}BN	d*	51µm	
Neutron reaction to 94%		$^1_0n + {}^{10}_5B \rightarrow {}^4_2He + {}^7_3Li^* + 2.31$ MeV	
Energy distribution		E(He) = 1.47 MeV	E(Li) = 0.84MeV
Range of reaction nuclei in boron nitride		R(^{4}He)= 3.9 µm	R(^{7}Li) =2.15µm

III. SUMMARY AND CONCLUSIONS

In the introduction nine requirements are discussed which should be met by an ideal neutron scintillator. In view of these the HDBN solid is evaluated.

1) Absorption Probability

HD^{10}BN is a very strong neutron absorber. The absorption length is only 50 µm.

2) Colour of scintillation light

The mean light wave length is 400nm, ideal for photomultipliers and semiconductor diodes.

3) Number of photons per absorbed neutron

A preliminary estimate leads to 6 to 8 time less intense signals compared to ^{6}Li glass. This, however, is not yet based on a direct measurement. So far no attempts are being made to increase the light output.

4) Duration of light pulses

The temporal width is extremely small, only 2.5 ns which is sufficient for any time of flight measurement and is favourable in avoiding pile-up effects.

5) Optical transparency

1mm thick HDBN appear white like salt in a bag. Day light is reflected in HDBN at the sintered BN grains like it is reflected in salt bag at the piled-up salt grains. In both cases there is no light absorption

6) Gamma insensitivity

There are sound arguments that the gamma sensitivity is very low. BN is low Z material and the required scintillator thickness is extremely small. Thus the probability of a photo effect, Compton process or pair production is small; in addition, the created high energy electron can hardly deposit its energy in the thin scintillator. The gamma sensitivity has not been measured yet.

7) Light background

The after- glow is very strong, however, one gets rid of this light emission after annealing the scintillator at 500C for 1h. This procedure might be inconvenient, however, it is very successful.

8) Large detector areas

HDBN is available in large quantities of different shape and size. HDBN can be shaped by cutting, milling, lacing, polishing, and drilling.

9) Scintillator cost

The material HDBN is cheap compared to any other scintillator.

At present it is too early to give more detailed information on this topic.

The deficiencies of this scintillator material are:

The low light output per absorbed neutron which is serious drawback.

1. The strong after glow which disappears after annealing
2. The optical non transparency based on reflection but probably not on absorption at grains.
3. For the next experiments which are under preparation HDBN disks of 2mm thickness are thinned down to 0.05 mm.

With such thin scintillators we should be able to measure directly the pulse height and pulse height distributions of light pulses generated by neutron absorption. Before exposure to a neutron beam the thin HDBN disks will be annealed in a light tight container and mounted in front of a photo cathode of a photo multiplier under IR illumination.

In an other experiment the transparency of the thin HDBN disks will be investigated in order to show that there is no absorption but solely reflection.

In further experiments the light response is investigated by high temperature treatments where atomic diffusion can take place.

ACKNOWLEDGEMENT

We are thankful to H. P. Bochem for cathode luminescence measurements, to Dr. G. Crecelius for the UV radiation experiments and valuable discussions, to Dr. David E. Holcomb, ORNL for performing two interesting experiments with HDBN and for his interest in this subject.

REFERENCES

[1] Angelika Pracht, private communication about her discovery of the scintillation of HDBN, Mai 2001.

[2] HDBN is manufactured by the Henze, Boron Nitride Products GmbH Heisinger Str. 12, 87437 Kempten (Germany). For more details see http://www.henze-bnp.de/

[3] ^{6}Li glass scintillators are manufactured by Applied Scintillation Technologies[3], Harlow (UK). For more details see http://www.apace-science.com/ast/

[4] J.B. Czirr, G.M. MacGillivray, R.R. MacGillivray, P.J. Seddon, “Performance and characteristics of a new scintillator”, Nucl. Instr. Meth., A 424 (1999), 15-19
[5] Dave Merrille Photogenics, Salt Lake City, Utah (USA) private communication (2004)
[6] Information on liquid scintillators see http://www.detectors.saint-gobain.com/
[7] C.W.E.van Eijk , Review on “Inorganic Scintillators and Storage Phosphors for Position Sensitive Neutron Detection” in the Conference Proceeding of the workshop on Position Sensitive Neutron Detectors, Berlin, Germany, June 28-30 (2001)
[8] R. Engels, E. Jansen, R. Reinartz, P. Reinhart, W. Schäfer, and J. Schelten IEEE Transactions on Nuclear Science, 45 (1998) 502-504 "Realization of a Small-Size High Resolution Linear Scintillation Detector”
[9] Sze, S.M., Physics of Semiconductor Device, New York, Wiley Interscience Publication, 1981, pp. 848-849.