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The Development of a Single-Crystal Fiber-Array Scintillator Area Detector

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Abstract

The scientific output of a neutron instrument is directly proportional to the effectiveness of its detector system – coverage of scattering area, pixel resolution, counting efficiency, signal-to-noise ratio, life time and cost. The current neutron scintillator detectors employ mainly ^{6}Li-doped glass and ZnS, both of which present well-know limitations such as low light output, high gamma sensitivity in the case of ^{6}Li-glass and optical opacity in the case of ZnS. We aim to develop a position-sensitive, flight-time differentiable, efficient and cost-effective neutron detector system based on single-crystal scintillator fiber-arrays. The laser-heated melt modulation fiber growth technology developed at NASA provides the means to grow high-purity single-crystal fibers or rods of variable diameters (200 μm to 5 mm) and essentially unlimited length. Arrays of such fibers can be tailored to meet the requirements of pixel size, geometric configuration, and coverage area for a detector system. We report a plan in the growth and characterization of scintillators based on lithium silicates and boron aluminates using Ce as activator.

1. Introduction

The major criteria of a high-performance scintillator for application as neutron detectors for scattering experiments at reactor and pulsed-source based facilities are given in Table 1. At present, there is no scintillator material proven to fulfill all the criteria. The currently employed scintillator materials are mainly ^{6}Li-doped glass and ZnS.[1-5] The main drawbacks are the low light output and high gamma sensitivity of ^{6}Li-glass and the optical opacity of ZnS. Considerable R&D efforts on scintillator technology have been on-going at ISIS (UK), Jülich

(Germany) and Delft (Netherlands) for many years. Currently, more than two-third of the instruments at the ISIS facility employ scintillation detectors.[1] Recently, Lnitel and co-workers [6] have evaluated a number of candidate scintillator systems, although most of them were based on polycrystalline materials. Other reported candidate scintillators are $LiGd(BO_3)_3$[7], BaB_2O_4[8], $Li_2B_4O_7$ [9], $CsLiB_6O_{10}$ [10], Y_2SiO_5 [11]and Gd_2SiO_5 [12, 13]. All of them use Ce or other rare-earth elements as activator.

Table 1. *Important design parameters and criteria for scintillator-based position-sensitive and time-differentiable detectors for thermal neutrons*

	Property	Practical Criterion
1	Neutron-capture cross section (barn)	940 for ^6Li(n, alpha) or 3837 for ^{10}B(n, alpha)
2	Light yield (photons/neutron capture)	4000 (minimum) – 10^5 (preferable)
3	Decay time (ns)	10-100, 5d-4f luminescence e.g., Ce^{3+} activator
4	Gamma sensitivity	$< 10^{-7}$, low-Z elements
5	Intrinsic background	alpha inactive
6	Spatial resolution	1-2 mm^2 pixel, traverse thickness < 2 mm
7	Optical Mechanical Chemical	Optical clarity, index of refraction < 1.8 Void-free, strength tangible to atomic bonding Stable in ambient atmosphere and humidity, resistant to radiation damage
8	Costs	< \$1000 per 100 cm^2 (scintillator alone), moderate cost for assembly and electronics

The consensus of scintillator technology favors the application of large, optically clear, high-quality single crystals. Single crystals free of voids or grain morphology leads to optimal light transmission and the long-range atomic order provides better control of the activator sites and energy levels, resulting in high quantum efficiency. The difficulties reside in the high costs and long lead-time in producing such single crystals (e.g., by the Czochralski technique or flux-growth method). This approach is unlikely to satisfy the full demand of scintillator-based detectors for future instruments of the next-generation neutron sources such as the US Spallation Neutron Source (SNS). Alternative scintillator media such as glass, polycrystalline compact, or microcrystals-binder composite inherently downgrade the efficiency and impose severe optical, mechanical and other constraints that are detrimental to the performance.

Using an assembly of single-crystal fiber arrays provides an attractive means in achieving high performance without the requirement of large bulk single crystals. This technology combines the high strength and high quantum efficiency intrinsic to single crystals and the flexibility in large solid-angle coverage of variable geometry. In principle, single-crystal fibers of continuous length can be laid out to cover a large area featuring desirable curvature and pixel resolution. The fiber configuration is readily adaptable to existing technologies of light collection and position decoding. Thus far it lacks an R&D program to apply the state-of-the-art fiber growing methods to produce and characterize new scintillator fibers. In the paper we present a plan and a status report on the development of single-crystal scintillator fibers for neutron detector application.

2. Fiber Growing Method

Single crystal fibers are fabricated by a novel melt modulation technique[§] developed at the Lewis Research Center of NASA. This melt modulation fiber growth method was described in detail elsewhere.[14, 15] The technique provides the capability of growing single and/or multiple single-crystal fibers, as well as single-crystal rods (up to ~5mm in diameter). It is superior to the conventional laser growth techniques that are limited to growing small diameter crystals, typically 200 μm or less. The growth of fiber crystals, which may be viewed as being nearly one dimensional, has been motivated, in large part, by the high yield and the fibers' extraordinary quality (e.g., dislocation- and void-free) for use in a variety of optical applications. Since the melt modulation technique permits higher pulling rates, the cost of growing fibers is substantially less than the cost of growing an equivalent volume of bulk crystal. In contrast to bulk crystals, the tensile strength of fibers is very high and they are flexible. Thus, they can be directly incorporated as fiber array scintillator device. The typical apparatus and examples of the grown fibers are shown in Figures 1 and 2, respectively.

[§] R & D -Award: R & D magazine selected the Pioneer II, *Melt Modulation Fiber Growth System,* as one of the 100 most technologically significant new products of the year 1993

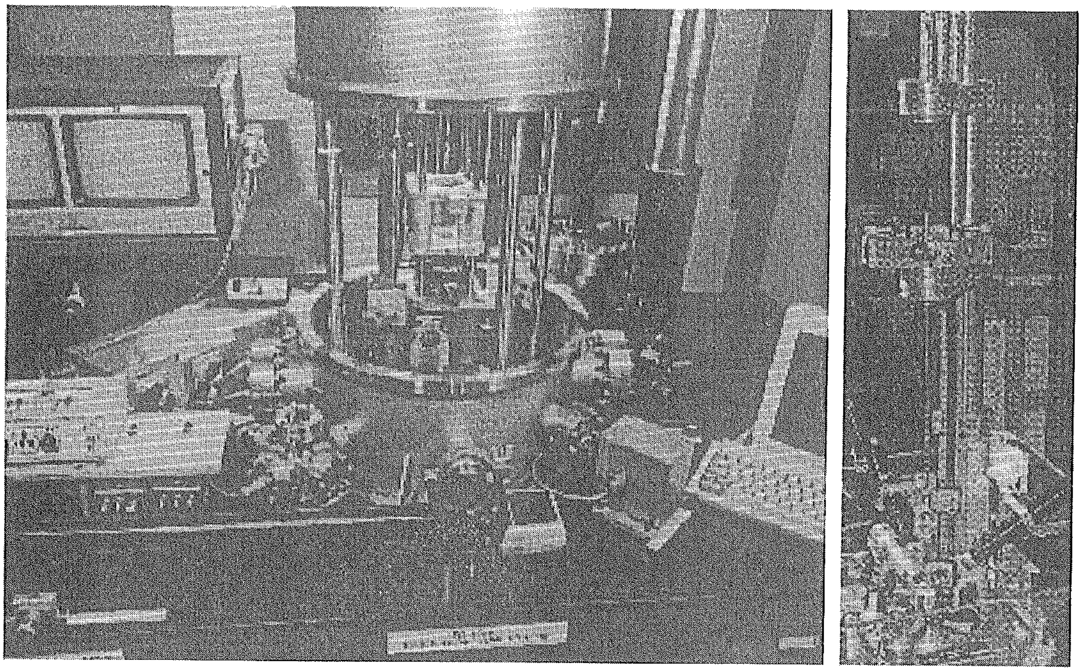

Figure 1. Melt modulation fiber growth units at the Lewis Research Center of NASA.

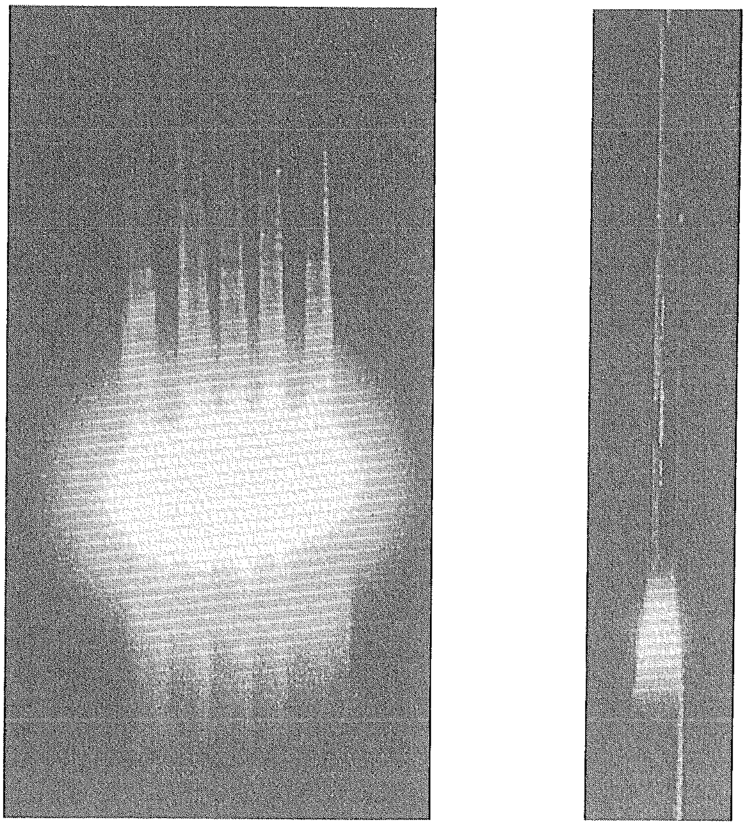

Figure 2. Examples of fibers being grown by the melt modulation technique.

3. Targeted Scintillator Materials and the Present Status

The progress in searching and testing new inorganic scintillators for detection of thermal neutrons has been reviewed by a number of workers.[1-6] In particular, Knitel *et al.* have presented the results of a systematic characterization of a variety of scintillator systems and identified the best candidates as: $LiYSiO_4:Ce^{3+}$, $YAl_3B_4O_{12}:Ce^{3+}$, $La_{1-x}Ce_xB_3O_6$, and $LiGd(BO_3)_3:Ce^{3+}$, and $LiBaF_3$ (possibly adding Ce^{3+} activator). [16] Currently, we are concentrating on the fiber growth of Li-Y-Si-O, Y-Al-B-O and Y-Ga-Al-O systems using Ce as activator. A test station consisting of a scintillator, light-coupling and photomultiplier assembly and a pulse-processing electronic assembly for measurements of pulse-height spectrum and light yield of these fibers from neutron-capture events was set up at IPNS. Our goal is to correlate the scintillator performance with the crystal-growth parameters so that improvement of the neutron-detection characteristics can be made systematically and expeditiously. We hope to demonstrate the feasibility of this single-crystal scintillator technology and to improve the scintillator performance to an acceptable level. Our long-term goal is to produce practical scintillator detectors for world-class neutron instruments for materials research.

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References:

1. N. J. Rhodes, A. J. Boram, M. W. Johnson, E. M. Mott, and A. G. Wardle, in *International Collaboration on Advanced Neutron Sources ICANS - XII*, edited by U. Steigenberger, Y. Broome, G. Rees and A. Soper (Rutherford Appleton Laboratory, Abingdon, Oxfordshire, U.K., 1993), Vol. 1, p. 185.
2. W. Schäfer, E. Jansen, A. Szepesvary, R. Skowronek, G. Will, R. Reinartz, and K. D. müller, in *International Collaboration on Advanced Neutron Sources ICANS - XII*, edited by U. Steigenberger, Y. Broome, G. Rees and A. Soper (Rutherford Appleton Laboratory, Abingdon, Oxfordshire, U.K., 1993), Vol. 1, p. 200.
3. M. G. Strauss, R. Brenner, H. P. Chou, A. J. Schultz, and C. T. Roche, in *Position-Sensitive Detection and Thermal Neutrons*, edited by P. Convert and J. B. Forsyth (Academic, New York, 1983), p. 175.

4. R. K. Crawford, J. R. Haumann, and G. E. Ostrowski, in *International Collaboration on Advanced Neutron Sources ICANS - XI*, edited by M. Misawa, M. Furusaka, H. Ikeda and N. Watanabe (KEK Report 90-25, Tsukuba, Japan, 1990), Vol. II, p. 943.
5. R. K. Crawford and N. Niimura, in *International Collaboration on Advanced Neutron Sources ICANS - XI*, edited by M. Misawa, M. Furusaka, H. Ikeda and N. Watanabe (KEK Report 90-25, Tsukuba, Japan, 1990), Vol. II, p. 975.
6. M. J. Knitel, P. Dorenbos, J. T. M. de Haas, and C. W. E. van Eijk, Nucl. Instrum. Methods **A 374**, 197 (1996).
7. J. B. Czirr, G. M. MacGillivray, R. R. MacGillvray, and P. J. Seddon, Nucl. Instrum. Methods **A 424**, 15 (1999).
8. S. C. Sabharwal and Sangeeta, J. Cryst. Growth **187**, 253 (1998).
9. V. D. Golyshev, M. A. Gonik, and V. B. Tsvetovsky, J. Cryst. Growth **198-199**, 501 (1999).
10. K. Bai and S. T. Jung, J. Cryst. Growth **186**, 612 (1998).
11. S. Zhang, S. Wang, X. Shen, H. Wang, H. Zhong, S. Zhang, and J. Xu, J. Cryst. Growth **197**, 901 (1999).
12. D. Konishi, Y. Uozumi, H. Yoshida, and M. Matoba, Nucl. Instrum. Methods **A 420**, 467 (1999).
13. P. L. Reeder, Nucl. Instrum. Methods **A 353**, 134 (1994).
14. A. Sayir and S. C. Farmer, Mat. Res. Soc. Symp. Proc. **365**, 11 (1995).
15. A. Sayir and S. C. Farmer, Mat. Res. Soc. Symp. Proc. **365**, 22 (1995).
16. M. Knitel, Ph.D Thesis (Delft University of Technology, 1998).