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^{10}B enriched film deposited by e-beam technique on Al_2O_3 substrate for high efficiency thermal neutron detector

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ABSTRACT

In the framework of the research and development activity on He-free neutron detection systems, the solid state physics experimental techniques and characterization tools can provide a valuable contribution for detector assessment and performance improvement. Presently, a He-replacement strategy relies on the use of Li-and/or B-based neutron to charge particle converters coupled to particle detectors (solid state, gaseous, scintillators). In particular, in this paper the detailed structural and chemical–physical characterization of ^{10}B-enriched films deposited by e-beam evaporation onto alumina substrates is reported. X-ray diffraction and photoelectron spectroscopy investigations have revealed that high B content (about 78%) can be achieved on these films, contaminants being mainly C and O most likely coming from the B targets. Microstructural investigation carried out by AFM, SEM and nanoindentation techniques have mainly revealed that B films replicated the alumina substrate microstructural features. The described $^{10}B/Al_2O_3$ samples were successfully tested in a Gas Electron Multiplier based neutron detector with measured detection efficiency in full agreement with the expectations.

1. Introduction

Presently, the worldwide shortage of ^3He induced an interesting and important R&D activity to conceive innovative detection techniques especially for thermal neutrons both for their use at large scale facilities for neutron-based analysis, e.g. neutron scattering or for applications in homeland security, just to cite two important examples [1–5]. In the case of detection systems for neutron scattering facilities, the most common setup is based on the coupling of a neutron-to-charged particle converting material with a (solid state, gaseous or scintillation) particle detector. Converting materials such as boron (^{10}B), lithium fluoride (^6Li enriched), and gadolinium, that feature large absorption cross sections for thermal neutrons, are typically used in the form of thin films (a few microns thickness) on particle detector surfaces.

For solid state converting materials, the main issue to be addressed to develop effective and robust detectors operating over a lifetime in the order of 10–15 years, relies on achieving a complete control of the thin films chemical–physical properties.

In the framework of research and development activity on innovative thermal neutron detector, boron thin films have been recently proposed as a suitable neutron-charged particle converter technology for the development of ^3He-free neutron detectors based on the Gas Electron Multipliers (GEM) [6,7]. If the chosen mechanism relies on the ^{10}B and neutron interaction, that produces ^7Li ions and α particles, the main requirement for this application is, of course, to maximize the ^{10}B content in the films. The maximum in the detection efficiency is expected for 1–3 μm as the film thickness matches the ranges of ^7Li and α [6,8]. The crystalline nature of the film or its microstructure does not play any significant role while the presence of impurities is one of the most relevant concerns to be addressed. In addition, films must exhibit good adhesion to the substrate in order to avoid any delamination. Pure boron and boron carbide films deposited by both physical and chemical vapor deposition (PVD and CVD) methods were proposed and tested for neutron detector application [8–11]. While B_4C is chosen because of its chemical and thermal stability, pure B films are favored because the maximum content in the boron can be reached and can be easily deposited even at room temperature (RT) by PVD techniques. These deposition conditions, if compared to high temperature CVD methods, should, in principle, guarantee a lower film contamination and lower film residual mechanical stress which ultimately promote a better film adhesion.

On this basis, it has been shown that high neutron detection efficiency can be achieved in a detector based on GEM technology in which a set of boron coated lamellae were arranged perpendicularly to the incident neutron beam direction [6,7]. In that detector, the alumina lamellae

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were coated on both sides by ^{10}B enriched films deposited at RT by e-beam evaporation. In this paper, a detailed characterization of the chemical physical and microstructural features of the boron films is reported. It has been shown that film thickness as high as 1 μm was reached with about 78% mass content of boron and some contaminants (carbon and oxygen) coming from the evaporation source targets. The measured neutron detection efficiency was found to be 2.8% for the 300 nm ^{10}B film with 78% of B content, fully in agreement with the expectations based on the Monte Carlo simulations [6]. Thus, higher content of boron and hence higher neutron detection efficiency is expected as soon as target manufacturing will be optimized.

2. Experimental setup

Boron films were obtained by e-beam evaporation technique using ^{10}B enriched target as sources. Targets 3 to 5 mm thick with a diameter of either 12.5 or 25 mm were manufactured from ^{10}B enriched metal powder (isotope $^{10}B = 97\%$, Eagle Picher Technologies, LLc). The powder was firstly pressed at 250 MPa at RT and then subjected to thermal treatment at 1200 °C for 3 h under flowing He gas. For the thermal treatment, tablets were arranged on graphite plates. These tablets were used as evaporation source in a vacuum chamber equipped with a Thermionics 10 kW 4-crucible e-beam evaporation system. The 25 cm^3 crucible was filled with ^{10}B tablets using either graphite or Cu liners. ^{10}B films with 1 μm thickness were grown on both sides of 10 × 50 mm^2 randomly-oriented alumina lamellae (Wollemi International) set at 30 cm from the evaporation source with a maximum growth rate of 0.75 nm/s. Due to the rather large crucible–substrate distance, simultaneous coating of 9 lamellae with film thickness uniformity >90% can be obtained. Alumina lamellae were mounted side by side on a support in such a way that both surfaces could be subsequently exposed to the evaporating boron flux. Substrates were cleaned in acetone and then in isopropanol for $10' + 10'$ in ultrasonic bath before insertion in the vacuum chamber. The typical ultimate vacuum level before boron deposition was $<1 \times 10^{-4}$ Pa. Boron films were deposited at room temperature without heating the alumina lamellae.

Films were investigated by X-ray diffraction (XRD) θ–2θ scans using a 9 kW rotating anode X-ray tube as a Cu Kα source Rigaku SmartLab diffractometer, equipped with a Johansson monochromator on the incident beam.

X-ray photoelectron spectroscopy (XPS) data were acquired in an Ultra-High Vacuum (UHV) system operating at 4×10^{-8} Pa base pressure. It is equipped with both a non-monochromatic SPECS Mg Kα and a VG Al Kα monochromatic X-ray source and with a CLAM2 hemispherical analyzer operated at 100 eV pass energy for survey data and 50 eV pass energy for core-level spectroscopy and calibrated according to Ref. [12]. In order to probe the film composition at different depths, samples were mildly sputtered in UHV with 1 keV Ar ion energy. The Ar etching rate is estimated to be about 4.5 nm/min. The XPS intensities used for extracting the concentration were obtained by normalizing the area of the background-subtracted core level peaks by the CLAM2 analyzer transmission function [13], by the Scofield cross-section [14] corrected for the angular asymmetry [15] and by the electron mean free path [16].

Atomic force microscopy was performed using Park Instruments XE 150 AFM working in noncontact mode.

Roughness and topography were evaluated using SEM imaging and 3D profilometry using a Leica DCM3D profilometer (ISO 25178 and ISO 4287) using a 10× objective. In these experimental conditions the lateral and vertical resolutions are 470 nm (using blue LED light) and 30 nm, respectively. To avoid finite size effect in the roughness estimation, 4 mm long strip was imaged. Over 500 different profiles were extracted from the topography to evaluate roughness of the sample.

Nanoscratch measurements to evaluate film adhesion to the substrate were performed using an Agilent G200 nanoindenter (ISO 14577).

3. Results and discussion

The prepared ^{10}B tablets appeared black like the starting powder and very compact. In Figs. 1 and 2, XRD θ–2θ patterns of ^{10}B enriched metal powder and target are plotted. Powder pattern (Fig. 1) corresponds to the metal orthorhombic β-B phase the most stable form of boron system in standard conditions. On the contrary, new peaks appear in the target spectrum being the most intense ones centered at about 21.5°, 27.9°, 34.7°, 38.8° and 63.4° (Fig. 2a). These peaks can be ascribed to either carbide or oxide phases of boron. Possibly during the high temperature thermal treatment a fraction of boron reacted with carbon and oxygen from graphite supports and/or from flowing gas impurities. These phases are expected to be mostly concentrated on the tablet surface. Indeed, assuming the theoretical density, for 2θ spanning in the range 10°–40°, 95% of the X-ray radiation is diffracted within the 20–75 μm superficial layer which is by far much smaller than the target thickness [17].

It was observed that boron films deposited using such tablets became darker in color (from light to light-brown and finally dark) as tablets were consumed. XRD θ–2θ patterns recorded on B-coated alumina having different appearance (light: labeled S1, and dark: S2) are plotted in Fig. 3. Sample details are reported in Table 1. As it can be seen, apart from Al_2O_3 substrate reflections, only one peak at 27.9° attributed to a B_2O_3 reflection is detectable. The peak intensity decreases as the boron film appearance becomes darker and vanishes in the case of the S2 B/Al_2O_3 sample. The absence of diffraction peaks related to boron metal phases could be explained with the amorphous or nanocrystalline character of films. Similarly, even though no peaks are detected, the presence of boron carbide cannot be ruled out. Coherently, the XRD spectrum reported in Fig. 2b, recorded on targets after several boron deposition runs, reveals that the presence of oxide phases is largely reduced. Related peaks exhibit a very reduced intensity (or undetectable) if compared to the as-sintered target.

These features were deeply investigated by XPS. Both S1 and S2 B/Al_2O_3 samples were considered. In the former, two different regions were analyzed (reported as survey S1_I and S1_II) whereas one region was sampled in the case of the darker boron film.

All the samples showed an appreciable charging. For this reason, the binding energy (BE) has been calibrated according to the position of "adventitious" carbon that was set at 284.6 eV.

XPS survey spectra of all the as-inserted samples showed that the sample surface contains only B, O and C. The relative atomic concentrations (at. %) calculated by XPS analysis are summarized in Table 1. In the S1 sample about 50% of the detected atomic species is B, about 30% is O

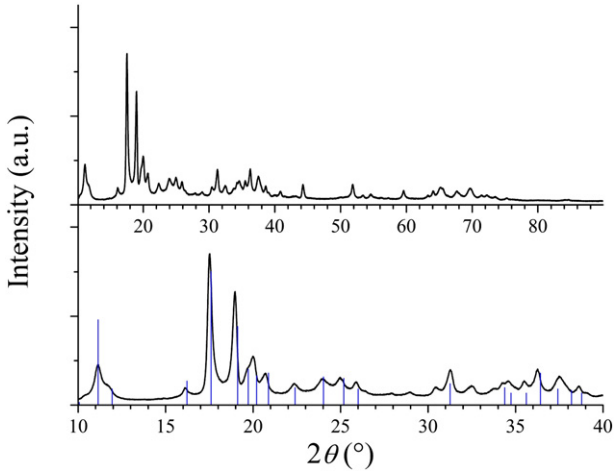

Fig. 1. Upper panel: X-ray diffraction θ–2θ pattern recorded on ^{10}B enriched metal powder. Lower panel: pattern details for 2θ spanning in the range 10°–40°; peak angle values for metal orthorhombic β-B phase (ICDD 31-0207) are marked with vertical continuous lines. The line heights scale with the relative intensities.