Assessment of suitability of an LBC-detector-based system for identification and quantification of radioactive nuclides in samples irradiated by cyclotron beams

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A gamma-spectroscopy apparatus was purchased from Berkeley Nucleonics Corporation (BNC) [1] in order to improve quality of radiation surveys of the devices irradiated at the TAMU Radiation Effects Facility [2]. All the devices under test (DUTs) must be surveyed upon beam exposure and only those that show activity at or below the typical background level can be released back to their owners immediately. Initial survey is normally done using a portable hand-held Geiger-Müller counter and most of the DUTs normally satisfy the above-mentioned criterion. Those that do not pass this test have to be kept and eventually surveyed at a later time in hope that the excess activity is short-lived and so these DUTs will pass the test then. Predicting that time is difficult without some kind of automation of the process.

Furthermore, DUTs irradiated with protons at a flux typical for the K150 cyclotron runs often show long-term activity. Those DUTs must be kept indefinitely or shipped back to the owner. The latter is preferable in order to prevent accumulation of radioactive material (RAM) at our facility. However, it is possible only if the owner is licensed to possess RAM and if the shipment has a declaration that specifies the radioactive nuclides present and their respective activities.

The new apparatus can address both of the above-mentioned problems, but first it has to be characterized in order to assess its capabilities and limitations. The characterization includes assessment of intrinsic background, energy scale, resolution as a function of energy, efficiency, and capabilities of the software for data acquisition and analysis.

Our system includes a lanthanum bromochloride (LBC) scintillation detector with a 2" diam. x 2" thick crystal, coupled to a photomultiplier tube (PMT) [3, 4]. The tube is plugged into the socket of a PMT base integrated with a power supply, an amplifier, an analog-to-digital converter, and a multichannel analyzer (MCA) with multichannel scaler (MCS) capability [5]. The data acquisition and analysis software [6] and a personal computer were purchased separately. The crystal and the PMT are encapsulated in 0.4 mm thick aluminum to protect the detector from ambient light and moisture and to reflect the scintillation light.

Scintillation material in our LBC detector is $LaBr_{2.85}Cl_{0.15}$:Ce. Its density is 4.90 g/cm^3, its photoelectron yield is about 1.30 times that of NaI, and its primary decay time is about 26 ns, which is an order of magnitude less compared to NaI. LBC detectors have superior mechanical strength and energy resolution compared to all other scintillation detectors. Typical energy resolution of an LBC detector is 3% FWHM at 662 keV. It is not as good as that of a typical semiconductor detector (0.2% for high-purity germanium), which limits complexity of a spectrum that can be effectively analyzed. However, an LBC detector is much cheaper, sturdier, and more versatile. Also, it does not require cooling by liquid nitrogen and is virtually maintenance-free.

Another limitation of the LBC detector usefulness comes from its relatively high intrinsic background, as illustrated in Figure 1, with about 200 counts per second (cps) total, and with about 100 cps between 33 keV and 220 keV for the 103 cm^3 crystal. Namely, natural lanthanum contains 99.911%

of stable ^{139}La and 0.089% of ^{138}La that has a half-life of 1.05×10^{11} y and undergoes β^- decay with 34.4% fraction and electron capture decay with 65.6% fraction, in each case emitting a single gamma-ray having energy of 789 keV and 1436 keV, respectively. The 1436 keV gamma-ray peak is prominent in the background spectrum (at 12 cps), while the 789 keV gamma ray peak blends with the 1436 keV gamma-ray Compton distribution. Because of the distributed nature of the source uniformly all over the crystal, the 1436 keV gamma-ray peak has a low-energy tail due to likely escape of scattered gamma rays and secondary x rays as well as a high-energy bump (stronger than the gamma-ray peak) due to coincidence summing with barium x rays. The 789 keV gamma-ray peak has a prominent high-energy tail because scintillations are produced by gamma and beta radiation combined (with beta end-point energy of about 255 keV) and because of coincidence summing with cerium x rays. Also prominent in the background spectrum are the overlapping peaks due to *K* x rays of barium and cerium, emitted following *K* electron capture and β^- decay of ^{138}La, respectively. Peak at 36 keV (50 cps) in Fig. 1 is predominantly due to Ba *K*β x rays. The Ba *K*α peak at 32 keV was cut off from the spectrum by setting a high discriminator lower level. Some lanthanum *K* x rays are expected to contribute to the spectrum as well, due to fluorescence by higher-energy gamma rays.

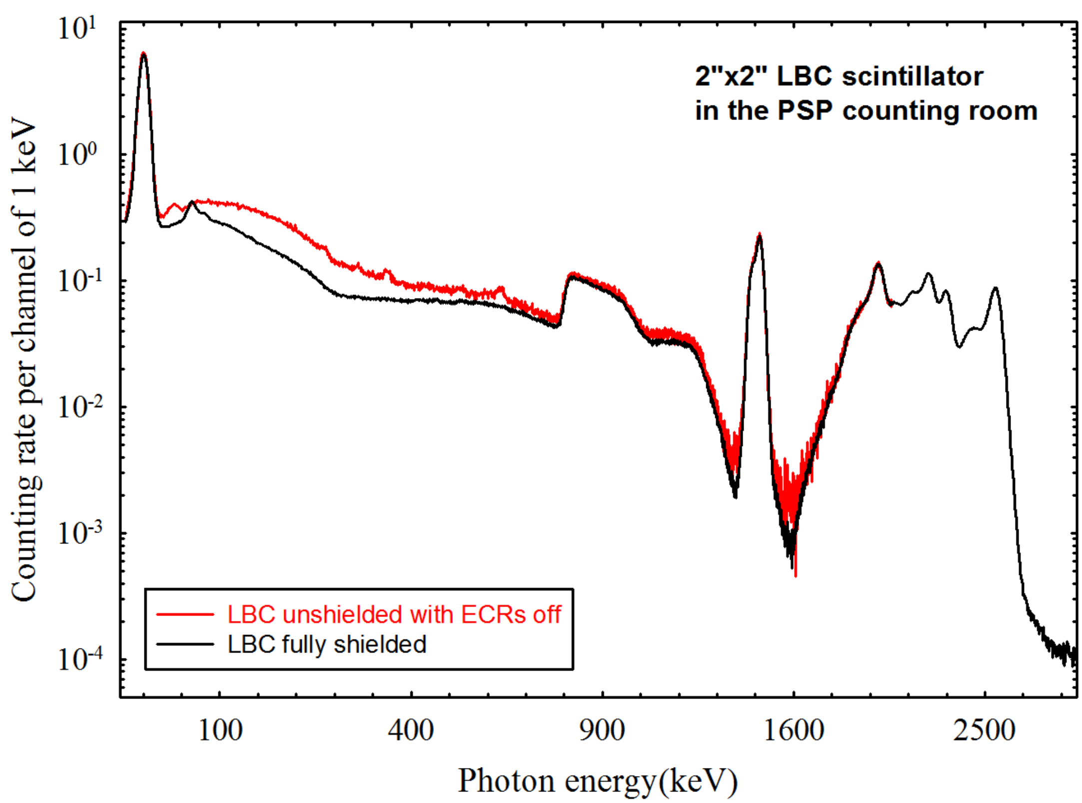

Fig. 1. Spectrum of the LBC detector background.

According to the manufacturer, the peak structure at energies from 1.6 MeV to 2.7 MeV (at about 32 cps) is predominantly "^{227}Ac-related" and possibly "^{235}U-related" (since ^{227}Ac is a progeny of ^{235}U), as these impurities are typically present is $LaBr_3$ and $LaCl_3$ raw materials [7]. Ac and La are homologous elements and therefore are extremely difficult to separate by chemical means, while the average number of ^{227}Ac atoms per La atom is estimated to be on the order of 10^{-14} [8]. Hull *et al*. [9] reiterate that these peaks are due to alpha decays in the ^{227}Ac (^{235}U) series and demonstrate that their position in the spectrum varies depending on the composition of the scintillator, unrelated to the photon energy calibration. Major individual peaks in the structure are attributed to alpha decays of ^{227}Ac progenies, specifically those of ^{223}Ra (5716 keV), ^{211}Bi (6623 keV), ^{219}Rn (6819 keV), and ^{215}Po (7386 keV) [10].

The two curves in Figure 1 illustrate the effect of shielding the detector with a lead-brick structure built around it and the contribution from nearby sources of radiation at the measurement location (PSP counting room). Apparently, shielding is most effective in the energy range from 45 keV to 700 keV, although it gives rise to peaks due to *K* x rays of lead in the region between 70 keV and 90 keV. It should be noted that radiation levels in the lab increase significantly when the ion sources and the cyclotrons are in operation, resulting in a broad distribution in the background spectrum, extending up to about 250 keV and peaking at about 85 keV. Therefore, the setup normally should be shielded by at least one lead-brick wall. It was found that shielding the PMT is just as important as shielding the LBC crystal.

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[6] https://www.berkeleynucleonics.com/bgamma-software-package
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