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Test Adapters Based on Natural Lutetium –

a Discussion of Benefits versus Conventional Check Sources

Abstract:

Recently an innovative series of test adapters based on high purity natural lutetium oxide was developed. The rare earth element lutetium contains the isotope Lu-176 with $3{,}6*10^{10}$ years half-life and a natural abundance of 2,6 %, which yields a specific activity of the pure element lutetium of about 50 Bq/g. The gamma and X-ray spectrum shows several peaks essentially in the range between 50 keV and 300 keV, the maximum beta energy is 600 keV. The available adapters contain up to 200 g of high density Lu_2O_3 ceramic material shaped to different dimension in order to optimize the response for a number of typical instruments. Depending on the particular design, these adapters are ideal to verify the beta radiation response of surface contamination monitors, the gamma radiation response of scintillation detectors and the energy calibration of gamma spectroscopic instruments. Each test adapter of a specific design represents the same and constant activity and activity distribution. The activity can simply be controlled by the weight and dimensions of the test adapters. Thus it is now possible to directly compare the response of sensitive radiation detectors – without the usual uncertainty in respect to the individual activity and surface emission rate of conventional check sources. Furthermore, due to their natural origin and low specific activity, in respect to many national and international regulations these adapters are not considered as radioactive material. The combination of all these unique features can help to facilitate the important frequent on-site performance verification of sensitive radiation detection equipment world-wide. These new test adapters will thus contribute to a reduction of calibration cost and instrument downtime, as well as to an increased user confidence and familiarity with "his" or "her" instrument.

KEYWORDS: Calibration; check source; lutetium; radiation detection.

1. Introduction

Suitable radioactive check sources are indispensable for the reoccurring functional verification of radiation detectors and instruments. While most dose rate detectors need to be checked and calibrated in calibration laboratory or calibration jigs, this is not necessarily required for highly efficient gamma detectors and portable surface contamination survey meters. Since these detectors are sensitive to even very small amounts of radiation, check sources of extremely low activity in the order of a few 100 Bq to 10 kBq are sufficient for a basic verification of these instruments. Especially gate monitors installed at steel mills, scrap yards and international border crossings as well as alarming personal radiation detectors operated by law enforcement officers and first responders can and should be checked by this category of radioactive sources.

Although in many cases the activity of these sources is quite low and sometimes even below the exemption values of the respective isotope given by national and international regulations for the transport and notification, a lot of inappropriate and potentially dangerous resistance against the use of radioactive check sources can be observed. One aspect may be the concern regarding the associated paperwork and hidden cost (e.g. of disposal) that are usually related to the purchase and possession of even the smallest check source consisting of man-made radioactivity. To some extent however this reservation to use radioactive check sources is certainly driven by the human psychology and the related fear of radiation. Unfortunately, the terms “radioactive source” or “radiation source” cover an incredible wide activity range [1]. The potential risk of operation, loss or damage of these sources ranges from disastrous to virtually non-existing. Talking about a “Cs-137 source” can refer to an irradiator in the million TBq range or to a large area reference source of a few hundred Bq. This huge span that is inherent in the word “radioactive source” apparently exceeds the human imagination capabilities and may explain both the mentioned resentments and the perceived overregulation and excessive paperwork that are often related to check sources of small activity.

In addition to these regulatory and psychological aspects, conventional test sources suffer from a number of inherent physical and procedural problems. Since every source is an individual and unique item regarding activity and surface emission rate, these data need to be known in order to quantify the results of an instrument check. For most isotopes the radioactive decay must be taken into account, which results in yet another error prone step that needs to be performed. For some isotopes different sources may have different spectra of the emitted particles depending on the details of the design and production process. Furthermore beta and alpha emitting large area test sources may have variations of the emission rate over the different sections of the surface and the thin active surface layer is a delicate part of the source that must be handled with special care.

As an alternative, materials and objects containing naturally occurring radioactivity (NORM) have always been used as workaround check sources. The dials of old watches and instruments (radium), incandescent lantern mantles (thorium), glazed ceramics (uranium) are typical examples of such surrogates. Although these items are sufficient to test the basic function of a radiation detector, they are not suitable for quantitative calibration purpose. The amount, composition and distribution of radionuclides is usually not defined and varies from item to item. In some cases, the associated dose rate and potential contamination risk of using these surrogates may even be considerable [2]. Quantitative measurements can be performed using natural potassium compounds. E.g. it is established practice to use KCl in a Marinelli beaker in order to calibrate a gamma spectrometer at 1,46 MeV. Unfortunately due to the low gamma emission rate and high gamma energy of K-40, in conjunction with the low specific activity and low density of KCl a large sample volume and long counting time is required.

This paper discusses the design requirements and key features of “check sources” based on another primordial radionuclide (Lu-176). Relative to the mentioned potassium based natural sources, these test adapters prevail the safety advantages of a well defined chemically pure natural material of very low specific activity and inherently low surface dose rate, but extend the range of applications to the fast test of even small size gamma detectors and large area contamination detectors.

2. Natural lutetium and the primordial isotope Lu-176

The rare earth element lutetium contains the stable isotope Lu-175 at a natural abundance of 97,4 % and the primordial isotope Lu-176 with $3{,}6*10^{10}$ years half-life and a natural abundance of 2,59 % [3]. Thus, the specific activity of the pure element lutetium is about 54 Bq/g. This specific activity value is quite comparable to the well-known K-40 in natural potassium (30 Bq/g). Per decay Lu-176 emits one beta particle with a maximum energy of 589 keV and approximately 1,7 discrete electrons, thereof 0,18 electrons above 150 keV. As a preferred chemical composition, lutetium oxide Lu_2O_3 was selected. Unlike lutetium metal, it is chemically stable, but still contains a very high and well defined content of lutetium. Lutetium oxide is available in high chemical purity and can be compressed and transformed to ceramics material with a density value of over 9 g/cm³. Test adapters containing this material are well-defined and reproducible. Each test adapter of a specific design represents the same constant activity and activity distribution. The total activity can simply be controlled by the weight of the natural Lu_2O_3 material. The specific surface emission rate of the beta radiation is a constant value for each and every specimen. Thus, it is now possible to directly compare the response of sensitive radiation detectors – without the usual uncertainty in respect to the activity and surface emission rate of conventional check sources. Consequently, these test adapters offer many performance advantages which are exciting not only for the scientist working in the field of metrology, but also for the technician in the field. Table 1 summarizes the basic decay properties of Lu-176 in comparison to a selection of other man-made and primordial radionuclides.

Table 1: Comparison of relevant nuclear data [3] for man-made and primordial beta/gamma radionuclides: Co-60, Cs-137, K-40, Lu-176, Rb-87 and La-138, sorted by the half-life of the isotope.

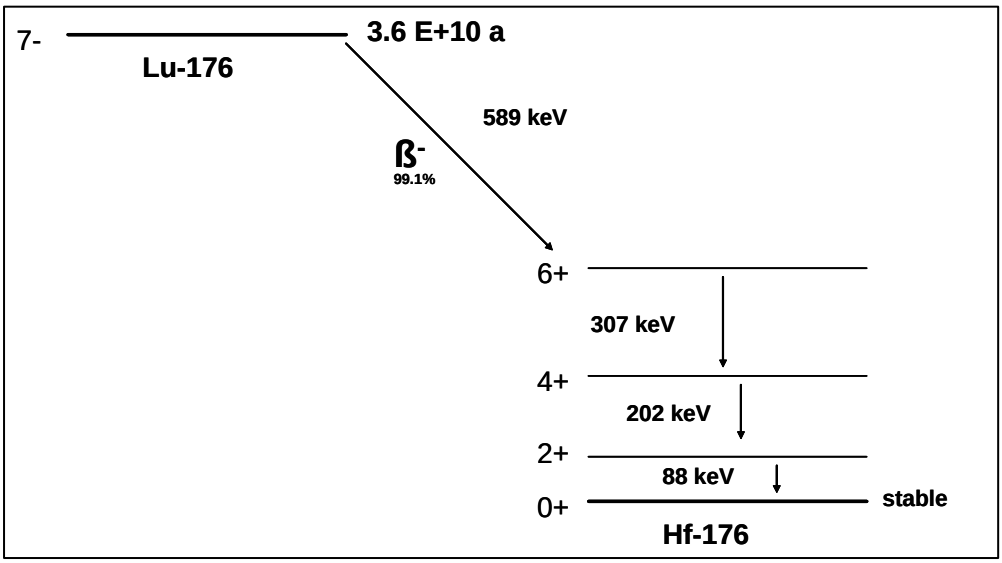
Nuclide	Half-life	Specific activity of isotope (Bq/g)	Natural abundance	Specific activity in natural element	Max beta energy	Gamma & X-rays > 50 keV
Co-60	5,27 a	4,19E+13	NA	NA	318 keV (100%)	1,17 MeV (100 %) 1,33 MeV (100 %)
Cs-137	30 a	3,22E+12	NA	NA	512 keV (95%) 1173 keV (5%)	662 keV (89 %)
K-40	1,28 E+09 a	2,59E+05	0,01%	31 Bq/g	1312 keV (89%)	1,46 MeV (11 %)
Lu-176	3,6 E+10 a	2,09E+03	2,59%	54 Bq/g	589 keV (99%)	55 keV (26 %) 63 keV (6,9 %) 88 keV (13 %) 202 keV (84 %) 307 keV (93 %)
Rb-87	4,8 E+10 a	3,17E+03	27,9%	883 Bq/g	273 keV (100%)	-
La-138	1,05 E+11 a	9,13E+02	0,09%	0,8 Bq/g	253 keV (33%)	788 keV (33 %) 1436 keV (67 %)

It is quite obvious from the data in table 1 that Lu-176 in natural lutetium is the most suitable primordial isotope for both gamma and beta instrument testing (see as well Fig.1). Compared to K-40 it shows a 20 times higher activity related photon yield and, due to the lower gamma energy, the detection efficiency of these photons is significantly higher as well. Furthermore the beta energy is much lower, so that a more critical test of any detector efficiency can be performed.

Rb-87 might become a potential candidate for a large area reference source (pure low energy beta radiation), resulting in an alternative nuclide to Pm-247 with its inconvenient and costly short half life of 2,6 years.

La-138 in natural lanthanum results in a gamma source that is quite similar to K-40 in natural potassium. Thus the same limitations regarding size of a test sample and measuring time apply.

Figure 1: Decay scheme of Lu-176 [4], the half-life of Lu-176 was taken from [3]



3. Application examples using Lu-176 photon radiation

Lu-176 is a very efficient check source isotope, especially for smaller size detectors of portable instruments. Lu-176 photo-peaks are located in a very convenient energy range that is usually covered by the man-made radioisotopes Am-241, Co-57 and Ba-133. Fig. 2 shows an example where the lutetium test adapter is shaped to best match the outer surface of a radiation pager detector. Thus, the lutetium oxide material is brought as close as possible to the detector surface so that the signal to count rate ratio is optimized. In that example only 36 g lutetium oxide are required to induce a net count rate of 100 cps – versus a typical background count rate of 10 cps of the instrument. The corresponding gamma dose rate in a distance of 10 cm caused by this adapter is less than 10 nSv/h.

Figure 2: Special shaped test adapter for optimized response of a personal radiation detector RadEye PRD with NaI(Tl) detector.

Since all photon and beta radiation of Lu-176 is promptly emitted, for tutorial purposes, interesting experiments using different detector to source geometries can be performed, e.g. in order to enhance and suppress sum peaks and coincidence counts. A disc-shaped version can be placed on top of any spectroscopic gamma detector. Depending on the distance, a more or less pronounced sum peak of the 202 keV and 307 keV appears in the spectrum (Fig. 3).

Figure 3: Gamma energy spectrum for Lu_2O_3 (2,5 g/cm²) measured with NaI(Tl) detector.

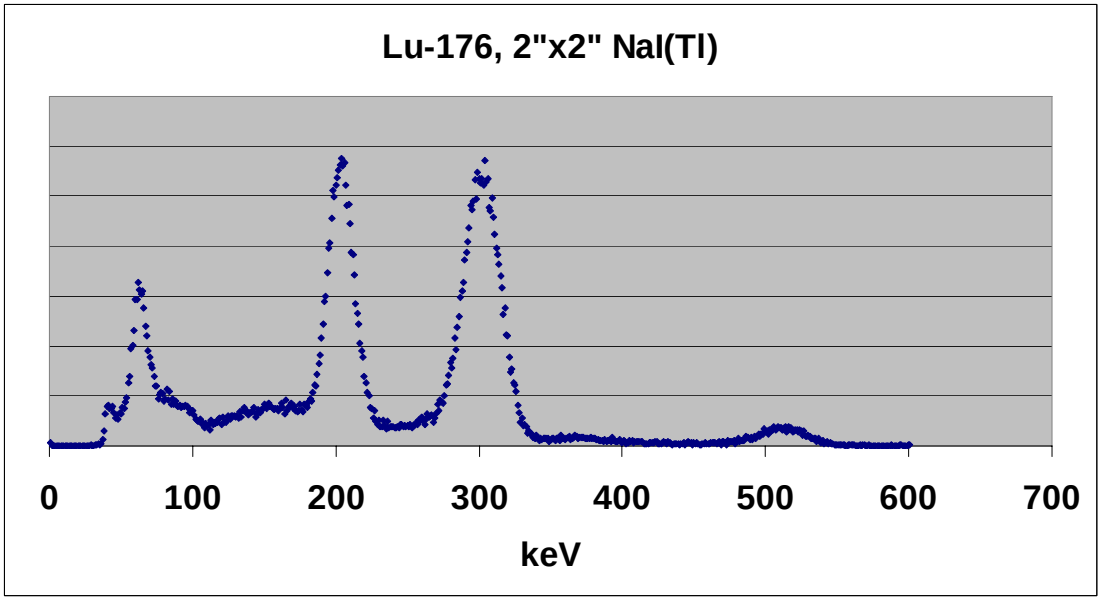
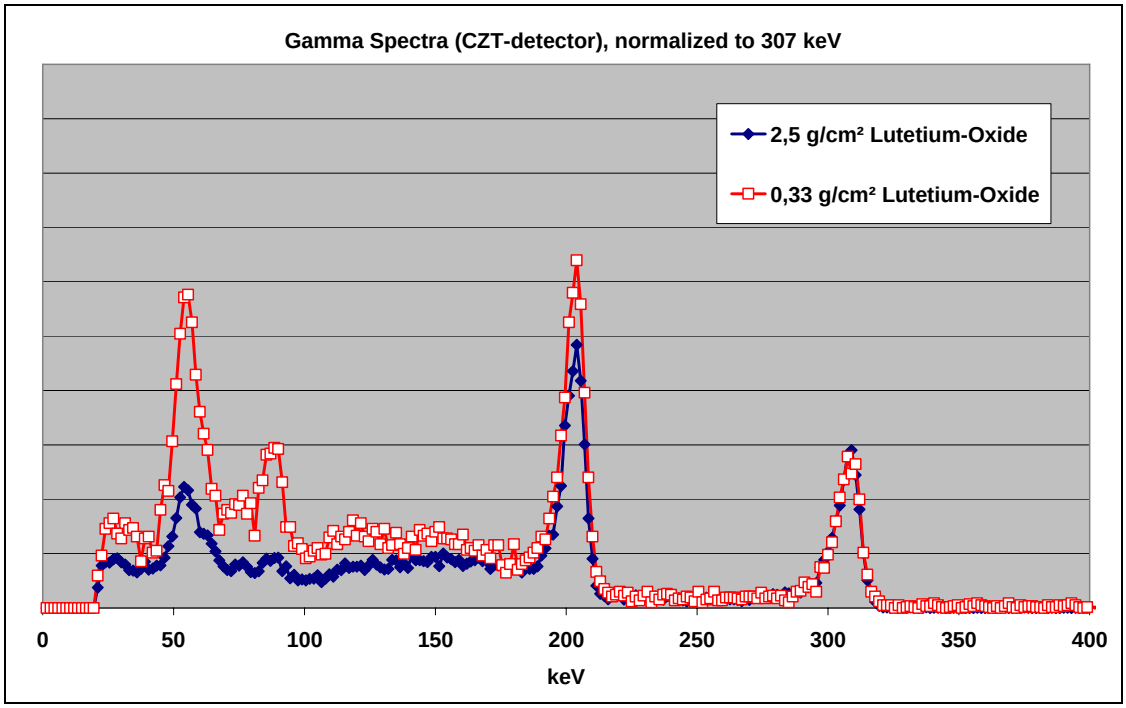


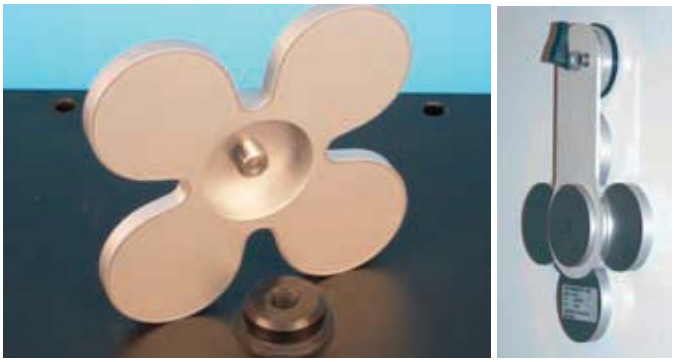
Fig. 4 shows the measured gamma spectra for thin and thick layers of lutetium oxide, taken with a spectroscopic personal radiation detector (Thermo Scientific Interceptor) using a CdZnTe detector. It is interesting to note that the attenuation in the lutetium oxide bulk material is significantly more pronounced for the 88 keV gamma peak (above the K-edge of lutetium) than for the characteristic X-rays (below the K-edge of lutetium at 63,3 keV) [5].

Figure 4: CTZ gamma energy spectra for Lu_2O_3 (2,5 g/cm² and 0,33 g/cm²).



Due to the low specific activity and the high self-absorption for gamma radiation in the Lu_2O_3 matrix (total attenuation coefficient without coherent scattering 0,585 cm²/g at 202 keV and 0,244 cm²/g at 307 keV [6]), any test adapter suitable for large size gamma detectors needs to be designed as rather thin slabs of significant geometrical dimensions. Fig. 5 shows a 200 g test adapter version that can be used for calibration of installed monitors at border crossings and e.g. the gates of steel and recycling facilities.

Figure 5: Gamma type “check sources” made of low specific activity material require special design considerations. The picture shows a 200 g test adapter for planar detectors. It consists of 4 coupled Al-encapsulated discs of 5 cm diameter each. The threaded bolt in the centre of the arrangement is used for mounting the test adapter to a matching holder of the detector enclosure. The right part of the figure shows the use of a suction cup holding device with which the test adapter can be fixed to any flat detector enclosure surface.



4. Application examples using Lu-176 electron radiation

Due to the short range of beta particles even thin layers of the bulk lutetium oxide ceramic are of "infinite" thickness for the beta emission rate and energy distribution. No coating of the bulk material is required. Unlike conventional man made beta sources, where the active material is embedded and covered by e.g. a thin aluminium layer, there are no variances possible regarding the energy loss and back scattering effects. The advantages of such large area test adapters can be seen in the inherent traceability and homogeneity. Each and every lutetium test adapter is a twin or clone of other test adapters built to the same geometrical dimension. No individual source data like manufacturing date, activity emission rate etc. need to be taken into account.

Square ceramics tiles of 36 mm x 36 mm and a thickness of less than 1 mm are mounted to an aluminium backing and frame in order to form the desired dimensions of an area with totally homogeneous surface emission rate (Fig 6 to 8). Thus wide area reference sources of virtually any dimension with a uniformity much better than 10 % can be produced. Limitations are given by the fact that the beta emission rate is less than 1 per s*cm² and that a decent photon contribution needs to be considered. However, the low emission rate of these wide area reference sources is ideal for training purposes in order to learn how to detect low levels of surface contamination. Each student can work on exactly the same "source" and gets a real feeling of detector response times in respect to the statistical nature of the radioactive decay.

Figure 6: Schematic design of large area test adapters that can be used to measure both the beta and photon response of large area contamination detectors.

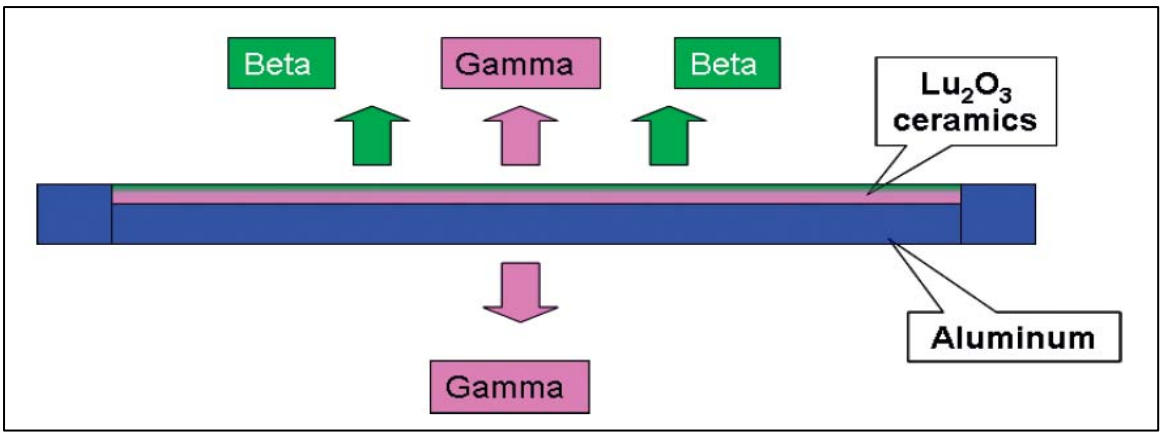


Figure 7: Lutetium oxide test adapter for rectangular large area portable contamination detectors manufactured by Thermo Scientific (FHT 382, RadEye AB100, DP6, HP 380) and corresponding measurement data taken with the RadEye AB 100 for 8 different specimen of this type of test adapters. The variation of the measured counts for a counting time of 300 s is well within the expected range of counting statistics (1 %). The test adapter can be used from both sides – the side of the lutetium oxide ceramics emits both beta and gamma radiation, the other side is photon radiation only. The dimensions 74 mm x 148 mm = 110 cm² of the active surface exceed the detector surface area in accordance with the recommendations of ISO 8769.

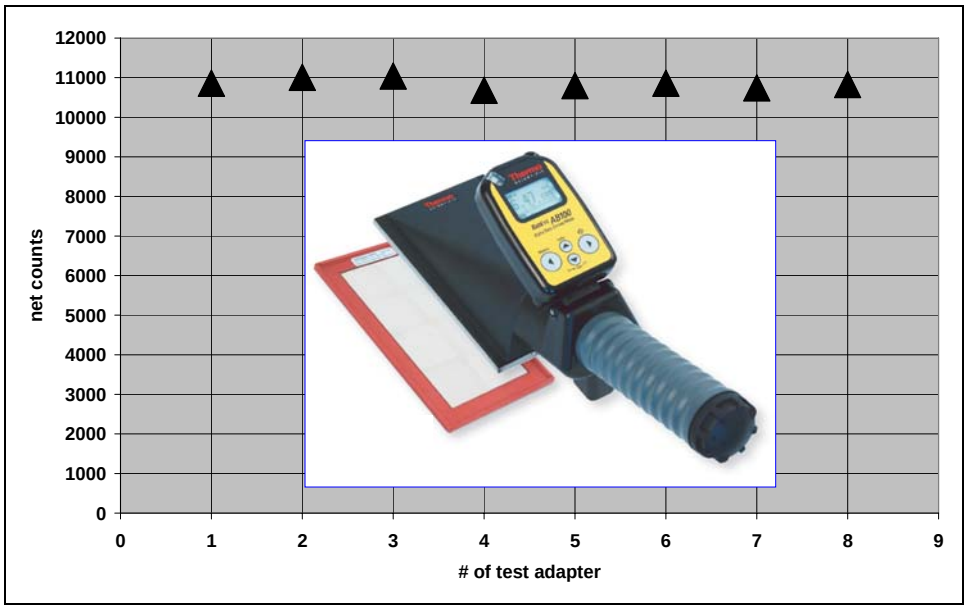
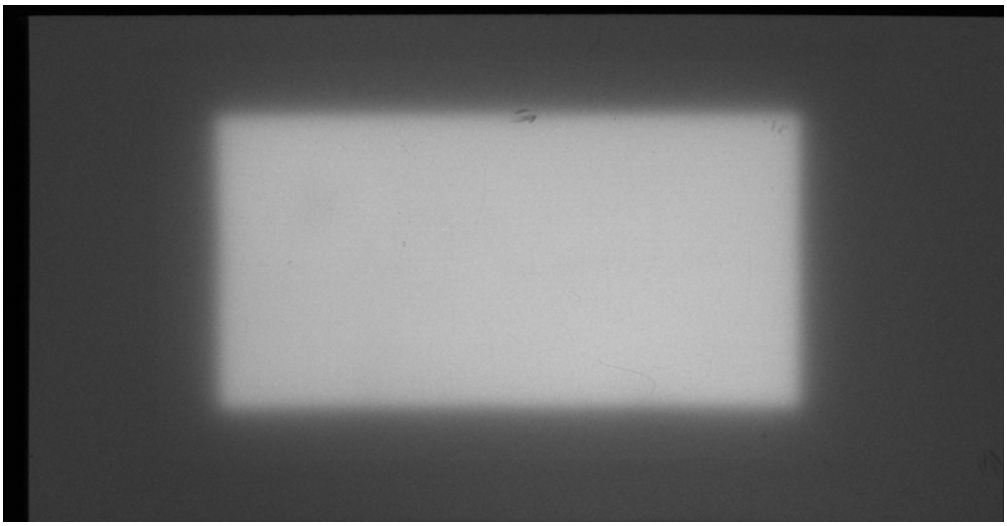


Figure 8: Autoradiography picture of the large area test adapter shown in Fig. 7.



Simple absorption measurements show that the energy distribution for the electrons escaping the lutetium oxide ceramics is altered, such that the effective energy is slightly below Cs-137, but significantly higher than Co-60 (Fig. 9). The reference data were derived from conventional 100 cm² sources.

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