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Alpha Emitter Intrinsic Concentration in Copper required for Nuclear Spectrometry Application

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Abstract Low-level radioactivity content in copper are employed for bolometric thermal radiation sensors and astro-nuclear spectrometers. The required lowest achievable alpha emitters concentration, for treated and untreated surfaces, are measured by Double Sided Silicon Strip Detectors in a high vacuum chamber and provide information on its intrinsic NORM content. Results shows that copper alpha emitters content can be achieved in the range below 0.01 (counts. $keV^{-1} \cdot kg^{-1} \cdot y^{-1}$) adequate for specific nuclear spectrometry applications.

Keywords: alpha emitter; copper; nuclear spectrometer, self radiation

1. INTRODUCTION

Copper with low-level radioactivity is often employed as structural matter for active volume in nuclear detectors and cope well for most spectrometry applications. On the other hand, for some specific cases, e.g. as required for detectors employed to record rare nuclear events or to search for unknown forms of mass or energy (like dark matter), commercially available high purity copper do no cope with matter purity requirements. In fact, the demanded metal ultra-low intrinsic radiation background is not met, for instance sach as in the case of bolometric thermal radiation sensors or astro-nuclear spectrometers; these require high detection sensitivity and therefore even low concentration of

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radioactive matter or NORM (Naturally Occurring Radioactive Matter) pose limitation on its employment [1]. Copper characteristic for detector supporting structure should be conveniently below ~ 0.153 (counts · $keV^{-1} \cdot kg^{-1} \cdot y^{-1}$). In order to achieve the lower attainable specific count rate, two main techniques are at hand: employing a proper (multi-layer) shielding assembly or selecting matter closer to the required elemental and radioactive purity; in the latter case, ultra-low concentration of NORM and Technically Enhanced one or TE-NORM [2] is of concern. For instance even under carefully controlled metallurgical manufacturing, Unat and Th is found on copper metal surfaces in a measurable amount and that constitute the so called self background together with the ubiquitous Radon (from radioactive families). These are unavoidable particularly radon daughters $^{218,214}Po$ as frequently plate out on surfaces exposed to the environment. The accompanying ^{210}Pb (last step in the decay chain) increases further the bulk copper background that are quite difficult to be removed [3]. Therefore in order to have a conveniently low radioactive background copper matter is necessary to employ a high purity one then, carry out a purpose made cleaning process under controlled environment. The cleaning effectiveness in removing surface encrusted NORM-alpha-emitters can be determined for instance by a Double Sided Silicon Strip Detectors (DSSD). In this study it was employed to measure copper samples in a high vacuum chamber operated at LNL target station. The spectrometric system, to collected alpha radiation energy and frequency, provides information on copper matter intrinsic contamination level and hints of its influence on detector performance. This work is related to copper commercially available and ultra clean bulk matter that was surface treated to assess copper suitability for nuclear detector structure for rare nuclear event.

2. COPPER WITH NORM CONCENTRATION

Minerals that contain naturally-occurring radioactive elements often are related to potassium (K) and radioactive families of uranium (U), thorium (Th) and in minor degree Np family, generally referred to as NORM or in the case of anthropogenic influence as TE-NORM; their concentrations in finished product depend on the exploited mine geological characteristics other than metallurgic processes and metal manufacturing. Relative radioactive substances can be mostly removed; nevertheless recycled scrap metal may contain Technically Enhance NORM in a wide range of concentrations for instance Nguyen reported in copper ore radiation doses levels of 582 nGy/h, [4]. There is evidence that also man made radioactive materials may concentrate in raw copper e.g. 241Am and other alpha or gamma emitters related to the 2053 nuclear weapons tests, oil exploitation, mining or other anthropogenic disrupting activity. Therefore, it may also be accidentally found even in high purity copper metals. Some detectors employing Cu-metal with NORM or TE-NORM, may be limited in their application concerning e.g. detectors technical performance; sensitivity or Lower Detection Limit (LLD) could be restricted by copper intrinsic concentration of radioactive isotopes or self radiation background. This aspect, has been taken into consideration so far, only in particular detectors design, since the over all background contribution due to environmental and cosmic radiation activation, in most spectrometry application, has negligible effect. Here we would stress the importance of surface treatment to reduce the radioactive isotope concentration (copper self-background) to attainable or convenient levels for rare events detection. Fig. 1 shows a typical alpha spectrum for bulk copper layers without surface treatment.

Preliminary studies did put in evidence, that alpha emitters random spatial distribution conveniently represented for arbitrary layers below surface as given in Fig.1, has an intrinsic NORM concentration. In the mentioned figure for the energy region above 3.5 MeV shows an average count rate of 10 counts per energy channel (spectrum on the right). The count rate diminishes exponentially only below the fourth layer corresponding to a thickness of ~ 20µm. The knowledge of alpha emission rate for each layer is of advantage for planning the surface cleaning procedure. A particularly effective NORM-reduction technique was developed recently by Camacho [5]; the applied sequence to remove thicknesses of copper layers is given in Fig.2; for technical details see the reference [5].

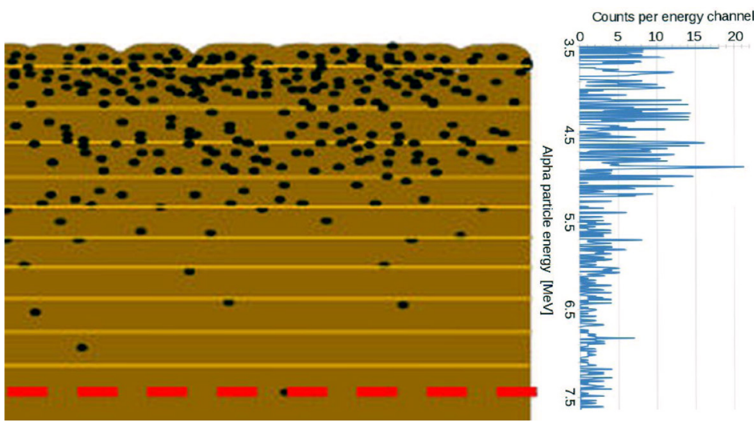

Figure 1: Bulk copper content of NORM-alpha emitter. On the left, an artistic sketch showing micrometer thick layers containing radioactive matter (black spots) and on the right experimentally observed alpha spectrum or the copper self-background, [5] (Camacho 2017).

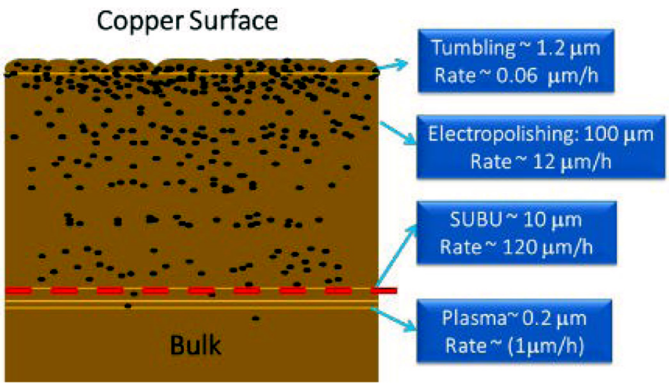

Figure 2: Schematic copper structured by layers to indicate their removal rate by different techniques suitable employed to reduce intrinsic NORM-alpha emitters from bulk matter adapted from [5].

That also indicates the removal rate characteristic for each ultra-cleaning technique providing a copper surface with a radioactive counting rate of (0.072±0.008) counts. $keV^{-1}\cdot kg^{-1}\cdot y^{-1}$ in the energy region of 2.7−3.9 MeV [6]. As an observation we would like to mention that the energy spectrum indicate for NORM matter tendency, an accumulation near the surface; this phenomenon is well known as observed also during crystal purification. This result is very important since shows that surface treatment may be applied to improve copper radioactive characteristics. Among the numerous techniques applicable for surface cleaning we may observe that the best procedure to reduce NORM concentration is primarily the electro-polishing treatment. However to obtain the most effective cleaning processes it is necessary to establish the effectiveness of a single or the best combination of available and tested techniques (viz. chemical etching, electro-polishing, tumbling, magnetron sputtering and plasma cleaning by DC magnetron) given in Fig. 2 on the right.

3. EXPERIMENTAL SETUP

Several copper pieces of regular geometry have been measured for self radiation background employing, as mentioned above Double Sided Silicon Strip Detectors based charged particle spectrometer [7]. The experimental system is conveniently placed in a suitable vacuum chamber operating at low pressure (~ 10^{-5} mbar). To trap gases and vapors, that often are retained inside the counting chamber that could interfere with measurements even in a very low concentration, a cold surface is employed to provide condensation. The DSSD is cooled down to the operational temperature of 5 °C, by a *Peltier cell*; the acquisition system block diagram is given, for illustration purposes, in Fig.3.

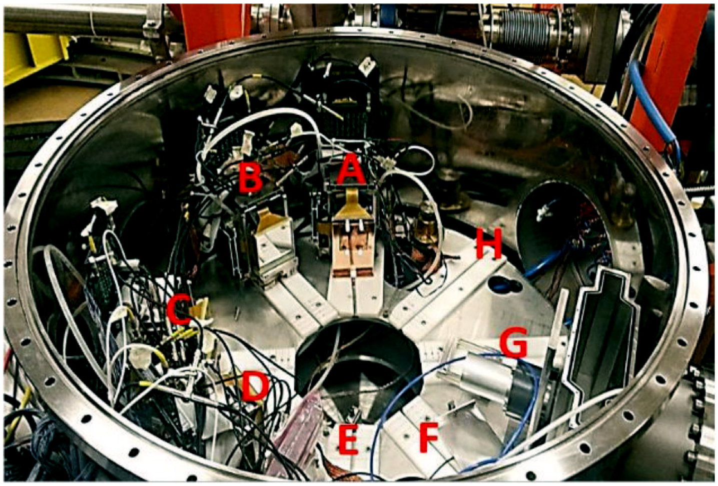

Figure 3: Spectrometer data acquisition system illustrating the operational principle. The pulse shape generated by the incident alpha particle ionization, is shown, as a bar histogram on the lower left square.

The low noise DSSD indicated by the first block in the flow-diagram of Fig.3, is described elsewhere, here we mention that is biased at ±25 VDC by a exceptional low-noise performance power supply. As the energetic charged particle cross the detecting active volume an electric pulse is generated by induced leakage current; the time dependent signal is reported as a bar histogram in the same figure below the detector. The out-put signal is proportional to the deposited energy and frequency i.e. NORM-concentration; further the spectral shape provide information on concentration related to each unit layer as mentioned above. The active area (48.5 mm × 48.5 mm) of the detector, is segmented into 16 strips per side, thus defining a 3 mm × 3 mm pixel structure displaying an energy resolution at 59.5 keV of 1.6 keV and 2.8 keV (FWHM) for the P and the N-side, respectively, as reported by Pierroutsakou [8]. This device and the sample with its holder, are positioned in the counting chamber shown in Fig 4, in which vapital letter A indicates the position of the detector facing the sample surface under test and D the measuring DSSD device to assess the vacuum chamber radiation background .

Further technical details of the signal processing, data acquisition including the custom ADC, and the spectrometer energy calibration is given elsewhere op. cit. [5]. Worth to mention that the data acquisition system is related to charge collection in a multi-strip array, therefore it is to be expected that the electrons-holes pairs formed by passing energetic alpha particles, may be swept in two strips, instead of a single one. To reduce the accompanying uncertainty

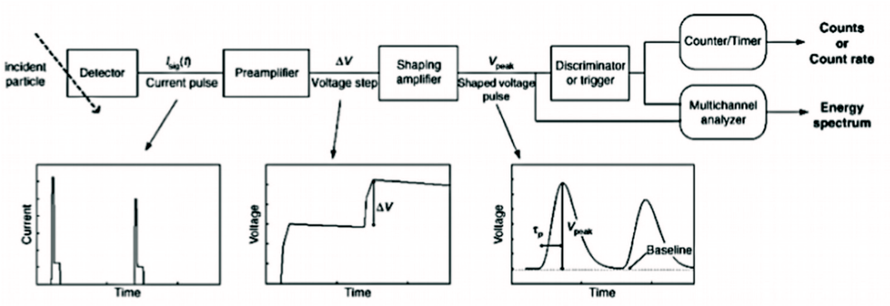

Figure 4: Vacuum chamber is housing detectors located in A and D is the spectrometer position to measure the activity of the copper samples and the vacuum chamber respectively. The other letters are not relevant in our case.

that could lead to large errors, the data are processed with multiplicity = 1, so that only those events are counted that are registered as signal induced by one strip per time interval unit.

4. RESULTS

As mentioned, the vacuum chamber matter also has to some extent, radioactive contamination and it is expected to contribute to some degree to the measured alpha-background. In fact, spectra related to the sample holder and the vacuum chamber, shown in Fig. 5, evidence surface radioactive contamination (peak

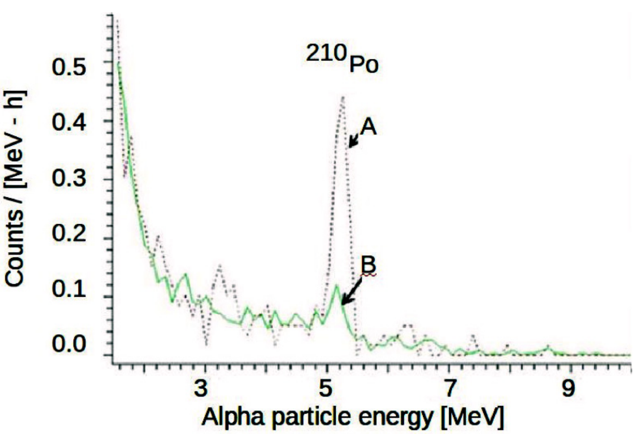

Figure 5: Alpha particle spectrum of sample holder and vacuum chamber radioactive contamination before and after surface cleaning.

A) and the residual NORM. comparison of similar spectra is indicative of the best and effective cleansing methodology (peak B).

We may observe that the peak corresponding to polonium-210 is significantly lower by a factor of 3 that qualify the followed surface treatment (B vs. A). The consequence of having in the vacuum chamber low background rate is advisable since a better discrimination is attainable for sample count rate. Since it was observed a relatively low interference in the spectrum from the chamber no further attempt was made to reduce the wall radioactive contamination. In Fig. 6 for illustration purposes we show the background subtracted alpha spectrum on the left and the vacuum chamber background spectrum on the right.

In the spectra of Fig. 6, on the left, the background subtracted data is shown which is higher in comparison to the background spectral area given on the right side. The outcome of the comparison betwee treated and untreated bulk copper, is that a significant reduction of radioactive matter content could be achieved by applying a selected cleansing sequence. Further, in the background subtracted spectrum, we identify a region with negative counts corresponding to alpha particles emission related to the vacuum chamber surfaces; that indicate a background rate higher than that corresponding to copper surface in the lower energy region. The positive region up to 7.6 MeV, shows the opposite case i.e the alpha emission rate is higher, as already mentioned, for the copper surface in comparison to the chamber background by almost a factor of three. The energy interval covers alpha particles corresponding to decays chain families of ^{238}U and ^{232}Th suggesting that the widespread distribution as shown in Fig. 7, is related to registered alpha particles originating from different depth below the copper surface.

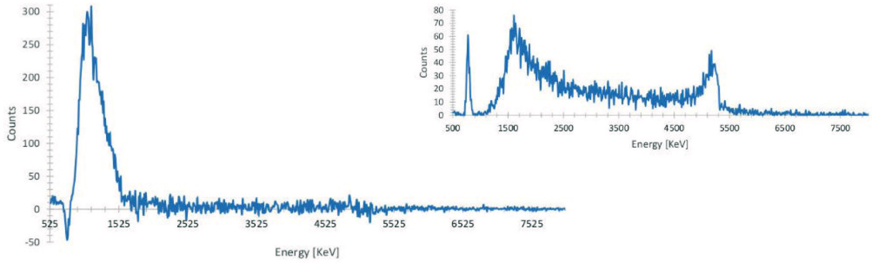

Figure 6: Background subtracted alpha spectrum from high purity copper bulk untreated surface (left) and typical background of the vacuum counting chamber (right).

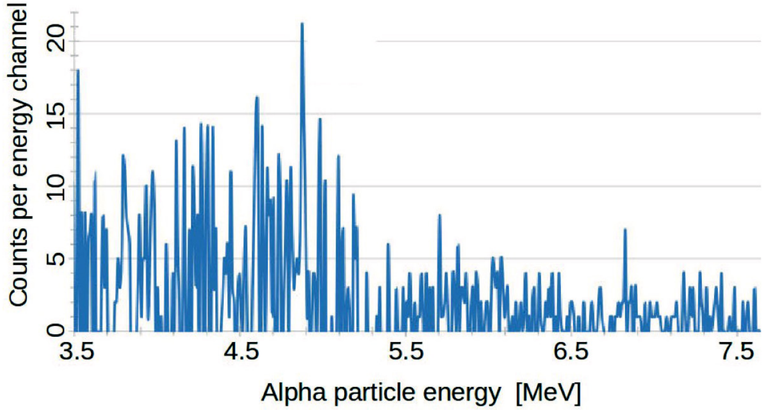

Figure 7: Typical alpha spectrum from treated copper surface (low concentration of NORM or TE-NORM).

Fig. 7, shows count rate for energy region 3.5 – 7.5 MeV, that is attributed to alpha particles originating from ^{234}U, ^{226}Ra and their decay products mentioned above. The corresponding energy loss of alpha particles in copper layers, is simulated using the Stopping and Range of Ions in Matter (SRIM®) software [9], which describes the transport properties of ions in matter. Resulting values, provide insight on the residual alpha energy (that absorbed in the detector active volume) and those lost by passing through different layer depths. Therefore, alpha particle emission rate in function of copper depth could be correlated with spectral data as it was shown previously in Fig.1.

5. DISCUSSION AND CONCLUSIONS

The manufactured samples of copper-OFHC (Oxygen Free High Conductivity Copper) is produced by the direct conversion of high quality copper cathode. Worth to mention that the high purity (99.99%) product is obtained by melting-casting under controlled conditions with CO gas as deoxidizer. In spite of the copper high quality, radioactive matter exists in a measurable amount. As mentioned, isotope ^{238}U, ^{232}Th and their decay products are of concern and it was objective of this study to establish copper suitability for specific nuclear spectrometry. Alpha emitters from NORM and TE-NORM, were determined in several samples that underwent chemical and physical surface treatments such as degrease, mechanical cleaning, electro-polishing, chemical etching and plasma cleansing. The surface intrinsic contamination was determined by a low level alpha spectrometer from which the charged particles emission rate was determined including the radioactive matter density in function of subsurface layers. It was shown that the NORM-matter concentration per unit layer was not a constant one, observing that the subsurface layer lowest content could be reached by removing a surface layer of 100mm thick. In fact the 5.2 MeV alpha particle, was detected with an energy of 500 keV originated from a depth at 81.26 mm thickness. This result was successfully achieved by tested method in that electrochemical etching provided the highest removal rate of radioactive contaminants from bulk and treated copper surface. A complex optimized etching sequence to reach a specific count rate of 0.0109 (counts. $keV^{-1} \cdot kg^{-1} \cdot y^{-1}$) was attained summing the 16 spectra obtained for each strip of the detector. Measurements were carried out during ~26 days for samples with a high purity metal surface and mass around 321g. Further this study did show that the higher alpha count rate corresponds to a layer at o depth of 6.7 mm and several times this thickness must be removed. The main conclusion is that employing a proper surface (chemical and physical) treatment sequence, copper metal radioactive content can be reduced to meet stringent self-background requirement to be employed in low level nuclear spectrometry. The best signal to background ratio was obtained after removing a thickness layer greater than 80 mm. This study pointed out that alpha particle spectrum, based on DSSD spectrometer, provide a proper technique to determine copper metal suitability for nuclear measurements where self background is of concern. We understand that a lower specific count rate is advisable; that certainly can be achieved employing natural radiation shielding e.g. storing bulk copper deep in a cave where the muon flux reduction could be around six orders of magnitude lower ($2.58x10^{-8}$ counts . $s^{-1}.cm^{-2}$) than the measurable value at manufacturing premises. Consequently the cosmogenic muon activation will be reduced accordingly providing bulk coper matter with lower self background.

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