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Absolute determination of small samples of Pu and Am by calorimetry

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ABSTRACT

An extensive measurement campaign has been carried in order to recalibrate and assess the performance of the small sample calorimeter (SSCAL) that was recently upgraded. The measurements have been performed in the Performance Laboratory of the Joint Research Centre's (JRC) Nuclear Security Unit in Ispra (Italy) using calibrated electric heat sources and standard reference nuclear materials. The SSCAL is a heat flow calorimeter which works by measuring the voltage generated by a heat-emitting sample across a thermal gap based on a thermopile cup technology. Results of calorimetry measurements carried out, both inside and outside a well-controlled environment of a climatic chamber, on reference Pu–Ga samples and well-characterised ^{241}Am samples are presented and discussed. The latter samples were produced at the JRC-ITU to be used by the JRC-IRMM for various cross-section measurements (total, neutron capture and ^{241}Am(n,2n) ^{240}Am).

1. Introduction

Calorimetry remains one of the most accurate non-destructive assay (NDA) techniques for materials containing plutonium, when combined with accurate isotopic analysis using high resolution gamma-ray spectrometry. This made it the primary measurement method in the USA during many decades from as early as 1940s when it became arguably the most important part of plutonium accountability. Since, with minor precaution, the magnitude of the heat flux leaving the sample container at equilibrium is not affected by the matrix, the technique is accurate, unbiased and unaffected by geometry and sample matrix effects. Furthermore calorimetry requires no reference standards which are representative for the items of interest.

In the late 90s the JRC's Performance Laboratory (PERLA) in Ispra (Italy) purchased a compact and transportable small sample calorimeter (SSCAL model 601C) [1] shown in Fig. 1. For indication, its left side rack (drawn in Fig. 2) which contains the thermal elements, the plug units through which heat items to be measured are introduced into the sample canisters etc. measures $580 \times 580 \times 800\ mm^3$ height. This device is able to accurately measure samples of Pu bearing materials corresponding to powers of less than 20 mW with a precision better than 0.2% at 10 mW powers. The upper range is about 200 mW. The objective initially sought was to use the calorimetry technique as an NDA measurement tool in order to reduce the number of samples subjected to costly destructive analysis. As described and reported in [2,3] the SSCAL has been extensively tested and its performance evaluated as the first of a new generation of plutonium calorimeters based on thermopile technology. It was subsequently used at the Institute of Isotopes of the Hungarian Academy of Science in Budapest (KFKI) for the characterisation [4], as dictated by IAEA safeguards requirements, of about 250 Pu–Be and Am–Be sources that came into the country mainly during the soviet union era. Recently the SSCAL has been upgraded, recalibrated and its performance re-assessed using calibrated electric samples and standard reference nuclear materials. The reference materials consisted of a set of Pu–Ga samples and seven well characterized $^{241}AmO_2$ samples, which were produced by the JRC Institute for Transuranium Elements (ITU) to be used for cross-section measurements by the JRC Institute for Reference Materials and Measurements (IRMM, Belgium). Six samples of these specimens consisted of $^{241}AmO_2$ dispersed in an Al_2O_3 matrix with approximately 40 mg of ^{241}Am. These samples were characterized by at ITU. The other sample, with a 320 mg nominal content of ^{241}Am, was prepared in a Y_2O_3 matrix and characterized by gamma-ray spectroscopy. These items have been measured in PERLA (Ispra) using calorimetry in conjunction to neutron spectrometry.

This paper reports on the results of extensive measurements and analysis carried in order to recalibrate and assess the performance of the SSCAL. Some aspects of the upgrades and improvements will be outlined together with some general description for completeness. Results of the calorimetry measurements carried out, both within and outside a well-controlled environment of a climatic chamber, on reference Pu–Ga samples

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and ^{241}Am samples will be presented. The results of the calorimetry measurements on Am samples are also compared to the masses which were measured at ITU using various means [5].

2. Description and general considerations

The SSCAL [1], developed by ANTECH (UK), measures heat producing samples with thermal powers in the range 0–200 mW. The SSCAL is one of a new generation of calorimeters that employ thermopile technology in conjunction with traditional nickel resistance thermometry. Thermopiles consist of a serially interconnected array of thermocouples, which produce an offset free Seebeck voltage that is proportional to the temperature difference between the two sites. The SSCAL is a heat flow calorimeter, which works by measuring the voltage generated across a thermal gap, consisting of a thermopile cup, when it contains a heat-emitting sample. The SSCAL has a dual measurement chamber ("twin cell") system of well size with 50 mm diameter by 100 mm height. The wells are designated A and B. The item to be measured is usually introduced in well A whereas well B provides the reference.

Fig. 1. Photo of the small sample calorimeter.

The inner measuring cylinders are made from an aluminium alloy that sits tightly in the thermopile cups. The cups are housed in a 25 kg cylindrical block of CR-100 aluminium alloy, which acts as a heat sink. The heat sink is surrounded by a 330 mm annulus thermopile of nominal sensitivity 0.026 mV/mW. All the thermopile components were provided by the International Thermal Instrument Company of California. The heat sink and the annulus thermopile are insulated at the top and bottom by expanded polystyrene to reduce axial heat leakage. The heat-sink, thermopile and insulation are contained within the outer aluminium alloy cylinder, closed at both ends which has two nickel windings, one for heating and the other for sensing. The cylinder is insulated on all sides by a type of expanded polystyrene. The outer cylinder is temperature controlled and serves to isolate the measurement chamber from fluctuations in ambient temperature. Access to the sample chambers is by a 350 mm long plug unit. Thermal equilibrium of the calorimeter is achieved by maintaining a zero heat flow between the heat-sink and the cylinder surrounding it. If the heat-sink is warmer than the outer cylinder, as is the case when a sample is present, heat will flow outwards from the heat-sink, towards the outer cylinder thus generating a voltage in the annulus thermopile. The voltage generated in the thermopile cups is measured by a dedicated two channel nano-voltmeter manufactured by the Keithley Company, wired directly to the cups. The nano-voltmeter has a resolution of 1 nV and an accuracy of 30 ppm/year. All other measurements are performed by a Keithley source metre via a scanner. Heat flow calorimeters are thus in their nature devices used to extract the mass of radioactive materials by measuring their heat output, and with the increased emphasis on safeguarding of nuclear materials, radiometric calorimetry had become one of the most preferred and powerful NDA measurement technique for plutonium and tritium which provides a reasonably high degree of accuracy and precision. However, calorimetry often requires long measurement times,

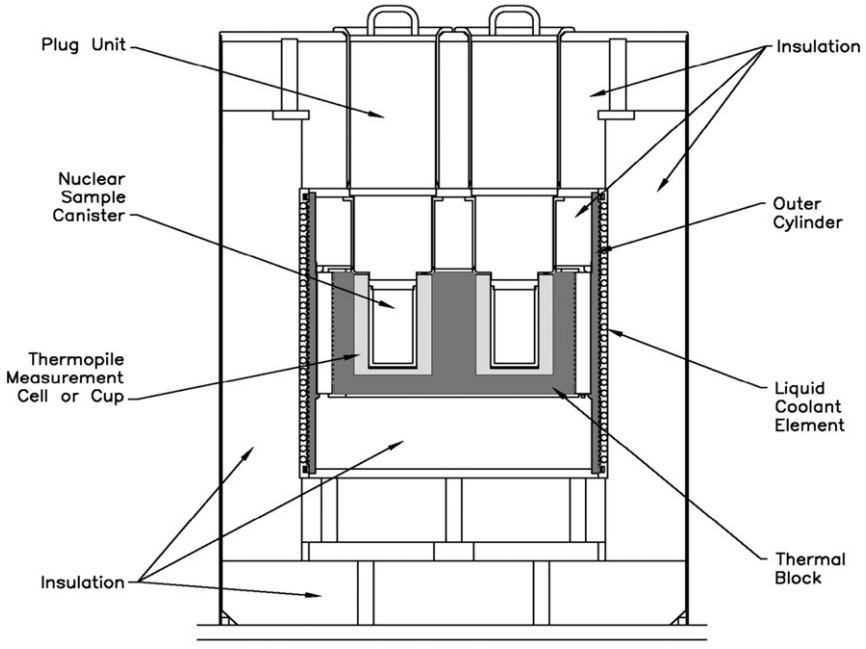

Fig. 2. Cross-section of the upgraded SSCAL 600C Series Thermal elements.