Nuclear Instruments and Methods in Physics Research A 784 (2015) 430–437

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Activation analysis study on Li-ion batteries for nuclear forensic applications

Erik B. Johnson [a,*], Chad Whitney [a], Keith E. Holbert [b], Taipeng Zhang [b], Tyler Stannard [b], Anthony Christie [b], Peter Harper [b], Blake Anderson [b], James F. Christian [a]

[a] Radiation Monitoring Devices Inc., 44 Hunt Street, Watertown, MA 02472, United States
[b] Arizona State University, Tempe, AZ 85287, United States

ARTICLE INFO

Available online 19 January 2015

Keywords:
Activation analysis
Nuclear forensics
Lithium batteries

ABSTRACT

The nuclear materials environment has been increasing significantly in complexity over the past couple of decades. The prevention of attacks from nuclear weapons is becoming more difficult, and nuclear forensics is a deterrent by providing detailed information on any type of nuclear event for proper attribution. One component of the nuclear forensic analysis is a measurement of the neutron spectrum. As an example, the neutron component provides information on the composition of the weapons, whether boosting is involved or the mechanisms used in creating a supercritical state. As ^{6}Li has a large cross-section for thermal neutrons, the lithium battery is a primary candidate for assessing the neutron spectrum after detonation. The absorption process for ^{6}Li yields tritium, which can be measured at a later point after the nuclear event, as long as the battery can be processed in a manner to successfully extract the tritium content. In addition, measuring the activated constituents after exposure provides a means to reconstruct the incident neutron spectrum. The battery consists of a spiral or folded layers of material that have unique, energy dependent interactions associated with the incident neutron flux. A detailed analysis on the batteries included a pre-irradiated mass spectrometry analysis to be used as input for neutron spectrum reconstruction. A set of batteries were exposed to a hard neutron spectrum delivered by the University of Massachusetts, Lowell research reactor Fast Neutron Irradiator (FNI). The gamma spectra were measured from the batteries within a few days and within a week after the exposure to obtain sufficient data on the activated materials in the batteries. The activity was calculated for a number of select isotopes, indicating the number of associated neutron interactions. The results from tritium extraction are marginal. A measurable increase in detected particles (gammas and betas) below 50 keV not self-attenuated by the battery was observed, yet as the spectra are coarse, the gamma information is not separable from tritium spectra. The activation analysis was successful, and the incident neutron spectrum was reconstructed using materials found in lithium batteries.

1. Introduction

Nuclear forensics is a deterrent for prevention of the detonation of a nuclear weapon in that the actors involved can be attributed to the weapon. One part of the nuclear forensics analysis is collecting and determining information regarding the neutrons generated in the explosion. The neutron spectrum emitted from a nuclear weapon provides insight into the weapon's mechanical construction, chemical composition, and nuclear isotopic content. A powerful tool would be to provide neutron spectra over the large area exposed to the detonation, requiring the use of a ubiquitous material with known properties. Lithium batteries meet these requirements.

1.1. Nuclear weapons

The unclassified neutron spectrum for nuclear devices is provided in [1,2]. The spectra are based on models from the World War II weapons (Fat Man and Little Boy) and a thermonuclear weapon. From [2], Dr. Preeg provided spectra of Fat Man, Little Boy, and a typical fission spectrum. The fission spectrum peaks around 1–2 MeV, where the spectra from the weapons are significantly attenuated. The spectrum from Little Boy peaks around 1 keV, where the spectrum from Fat Man is dominated by neutrons below 1 keV. The difference in these two spectra from 10 keV up to 1 MeV is roughly 2 orders of magnitude with the Little Boy

* Corresponding author. Tel.: +1 617 668 6886.
E-mail address: ejohnson@rmdinc.com (E.B. Johnson).

http://dx.doi.org/10.1016/j.nima.2015.01.046
0168-9002/

weapon producing more neutrons within this spectral region. The cause of this difference is due to the weapon construction, where the nuclear material in Fat Man is completely surrounded by explosive materials used to implode the plutonium to a supercritical mass.

The process of boosting uses a combination of deuterium and tritium to generate energy through a fusion process. The nuclear materials are compressed to a supercritical mass, with the resulting explosion bringing the D-T to a temperature where fusion can take place [3]. For fusion reactions that produce neutrons, the energy released is 3.7, 17.6 and 11.3 MeV for $^2H+^2H$, $^2H+^3H$, and $^3H+^3H$ respectively. One byproduct of these reactions is neutrons that will carry off a significant fraction of the energy produced. This excess energy adds to the explosive power of the device, as well as creating more fission reactions throughout the fissionable material. There will be a large contribution to these fusion reactions from the $^2H+^3H$ reaction, where 14 MeV neutrons are generated. The resulting neutron spectrum from a fission weapon can have a large contribution from these neutrons, as shown in [1]. The neutron energy spectrum from fission reactions falls off rapidly around 10 MeV, where fusion weapons involving tritium can be identified by detection of high energy neutrons.

1.2. Activation analysis

Correlating nuclear weapons to the output spectrum generated from their detonation provides attribution as the weapon construction can be related to the output neutron spectrum. A method used for reconstructing the incident neutron spectrum is neutron activation analysis. These analysis methods are used to determine constituents within a material of unknown composition [4–6], but nuclear scientists have used this method to determine the incident neutron spectrum based on known material compositions [1,7]. Neutron activation analysis is typically done with a variety of elementally pure metal foils of known mass and volume.

1.3. Lithium batteries

The lithium battery is composed of a cathode and anode with metal substrates, where the anode and cathode are separated by a porous material, such as polypropylene. Ion exchange is done through an electrolyte medium. The cathode is typically made from some lithium oxide, where the anode is usually graphite. The battery casing may consist of aluminum or stainless steel. The battery is inherently a set of metal foils and materials that have a potential to be used for activation analysis. The lithium content might be used to provide additional sensitivity.

All except one of the batteries used in this analysis are coin cell batteries. The majority of coin cell batteries consist of a single layer of cathode and anode. A porous membrane is placed between them. The battery layers are placed in a case along with electrolyte, where the case is pressed closed. A diagram of a coin cell battery is shown in Fig. 1. The other type of battery is an ultrathin cell, consisting of very thin layers of anode and cathode on a steel substrate.

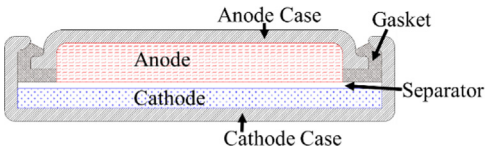

Fig. 1. Diagram of a coin cell battery. The battery consists of multiple layers of material pressed together. The steel anode and cathode cases are spaced with a gasket, while the anode and cathode materials are divided by a porous material (the separator).

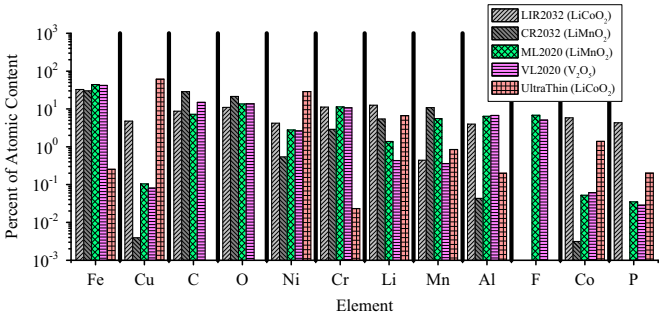

Fig. 2. The percentage of atomic content for the twelve most abundant elements in the batteries used in this analysis is shown.

2. Experimental setup and methods

2.1. ICP-MS results

To conduct activation analysis, the content of each of the batteries needs to be well known. A number of lithium batteries were broken up and dissolved for inductively coupled plasma mass spectrometry (ICP-MS) measurements to determine the chemical content. Though not all of the components of the battery were measured using the mass spectrometer, we conducted the analysis to determine the entire composition. The organic materials could not be measured by ICP-MS, as well as the electrolyte, since the electrolyte evaporated once the battery was opened. The process for determining the battery content is discussed in detail in [8].

A large number of batteries were processed, and the atomic concentration for batteries used in this analysis is shown in Fig. 2. From the ICP-MS analysis, the major constituent is iron for the coin cell batteries, as expected, since the battery casing is stainless steel. In the stainless steel, there is a large concentration of chromium, nickel, and manganese. As shown, there is a large concentration of lithium, yet an important element in small amounts, not shown, is gold, which is present in the ultrathin battery, the ML2020, and the VL2020 batteries. These inputs will be used in the activation analysis.

2.2. UMass FNI

Similar battery models purchased at the same time as those using in the ICP-MS analysis were exposed to neutrons from the University of Massachusetts, Lowell research reactor. A vessel (Fast Neutron Irradiator, FNI) was constructed to allow for fast neutron irradiation. The vessel is placed next to the reactor core and is designed to attenuate gammas and low energy neutrons. The end product is a lower neutron flux, 4.33×10^{11} n/cm^2/s compared to 1.81×10^{13} n/cm^2/s for irradiations in the reactor core basket, but the spectrum is significantly harder. The major advantage to the FNI is that it allows for multiple samples to be exposed simultaneously in a uniform beam, reducing run to run variations [9]. The x or y profile of the fast neutron (>0.1 MeV), thermal neutrons (<1 eV), and gammas is shown in Fig. 3. A diagram of the sample holder is drawn to scale to illustrate that the samples were exposed to a roughly uniform beam. From [9], the beam is also uniform in the z direction for the placement of the samples.

The sample holder was made from nylon foam (McMaster 3623K74), and a number of regions were cut out of the foam to allow the batteries to be press fit into the foam. The batteries were simply held in place from the compression of the foam on the battery. The holder diagram in Fig. 3 is a linearly scaled drawing of the holder, where the cut out regions actually were. The figure indicates a hole for an AA battery for reference, while the coin cell battery holes are pointed out as well. In this diagram, four of the coin cell batteries used in this analysis are drawn. Two of the