

Detection of Water Content in Aluminum Scrap with a Fast Neutron Source (Cf-252)

Miting Du¹, Qingyou Han², and Ray D. Peterson³

¹Nuclear Science and Technology Division, ORNL, Oak Ridge, TN 37831

²Metals and Ceramics Division, ORNL, Oak Ridge, TN 37831

³IMCO Recycling, Inc., Rockwood, TN 37854

Keywords: moisture detection, aluminum recycling, fast neutron source, thermal neutron sensor

Abstract

Aluminum scrap and remelt secondary ingot (RSI) may contain water in their internal voids. The presence of water can be a serious safety issue for the aluminum remelting industries. This article describes a method that utilizes neutrons for the rapid detection of water in solid aluminum materials based on the fact that neutrons interact with hydrogen atoms in the water molecules. A fast neutron source (²⁵²Cf) and a portable “slow-down” (backscattering) neutron detector are used for moisture detection. Initial experiments carried out with aluminum turnings indicate the detected slow neutron counts can be correlated to the hydrogen or water levels in aluminum scrap. The method has the potential of being used by the aluminum remelting industries for rapid, non-destructive detection of water in aluminum scrap or RSI.

Introduction

Aluminum scrap and remelt secondary ingot (RSI) may contain water in their internal voids. If water enters the cavities during storage and is not completely driven off by preheating or some other means, an explosion can occur when the RSI or scrap metal is charged into molten metal. RSI has been known to explode in a furnace when inadequate preheating occurs because of the trapped moisture. The violence is greater when RSI explodes under molten metal [1]. Because it is often impossible to visually ascertain whether aluminum scrap or RSI contains moisture, furnace drying is the only way to ensure that the materials are thoroughly dry prior to charging. Furnace drying is an expensive procedure, but the risk of explosions is unacceptable [2,3].

Another processing alternative to drying is to detect moisture or water in the scrap metals and RSI before they are charged into molten metal. If the method was valid, it would be possible to ensure that no water was charged into furnace. Methods of rapid detection of water could also be used to reduce the use of a dry furnace since it would not be necessary to dry RSI which contains no water. Current methods for detection of water involve weighing samples before and after drying. These methods are slow and are only appropriate for light gauge materials.

A promising way of detecting water in aluminum is to use neutrons. Detection of water by neutrons has a long history in well logging [4] and ground-water exploration [5]. Large neutron devices have been used for moisture detection in coal, cement and other non-homogenous materials. Portable neutron gauges have also been applied for direct measurement of water content inside

flat roofs [6]. This paper describes some initial experimental results on detecting water content in light gauge aluminum scrap.

Neutron - Hydrogen Interactions

A fast neutron source emits high energy neutrons that collide with the atomic nuclei of the elements of the test material and scatter in all directions. Figure 1 illustrates the elastic collisions of fast neutrons with the nucleus of hydrogen and with the nuclei of heavier elements such as aluminum. Neutron moisture detection is based on the fact that the fast neutrons (>0.5 MeV) from a neutron source (eg. Cf-252) can be greatly attenuated to slow-down or thermal neutrons (<0.5 eV) by interaction with hydrogen atoms in water, but not with other substances in aluminum scrap. The back-scattered slow-down neutrons can be easily detected by a thermal neutron counter.

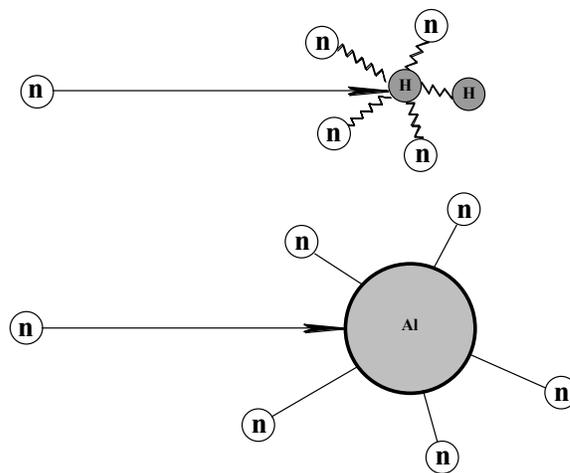


Figure 1: Schematic illustration showing the elastic collision of fast neutrons (n) with nucleus of hydrogen (H) and aluminum (Al).

Figure 1 illustrates the mechanism by which neutrons are “selectively” sensitive to surrounding water. Detectable thermal neutrons are formed more easily by a fast neutron collision with a hydrogen nucleus in water or other hydrogen-containing materials than with the nuclei of other elements. We know that when a neutron collides with the nucleus, the neutron loses some of its energy. The amount of its energy lost depends upon the particular nucleus with which the neutron collides. The amount of energy lost is a function of the ratio of the mass of the neutron to the mass of the nuclide with which it collides and of the angle of

back-scattering. A hydrogen atom has an atomic mass number ($A = 1.007825$ u) very close to that of a neutron ($A = 1.008665$ u). Imagine two different results for a flying ping pong ball: 1). when hitting a wall it changes direction, but is slowed down very little; 2). when hitting another ping pong ball at rest it transfers its kinetic energy to the other ball and is slowed down with less kinetic energy.

The following equation [7] gives us an estimated average number, N , of collisions necessary to attenuate a neutron with an initial energy E_0 to the energy E :

$$N = \ln(E_0 / E) / \zeta \quad (1)$$

where $\zeta = 1 + \alpha \ln \alpha / (1-\alpha)$ or $\zeta \approx 2(A + 2/3)$ when A is large; $\alpha = [(A-1)/(A+1)]^2$ or $\alpha \approx 1 - 4/(A+2)$ when A is large.

If we want fast neutrons of 2 MeV to be attenuated to 0.0253 eV, some average numbers of collisions (slowing-down parameters) are calculated for various substances using Equation 1 and the results are listed in Table 1. The calculation results indicate that it takes only 18 collisions between a neutron and hydrogen atoms to generate thermal neutrons when a small amount of water exists along the pathway of incident fast neutrons. Also we notice that the fast neutrons are almost “transparent” to high A substances. Aluminum scrap or RSI requires 252 collisions to generate thermal neutrons, meaning that very few thermal neutrons are generated along the pathway of the incident fast neutrons. As a result, water and other hydrogen-containing materials can be detected non-destructively in real time with a fast neutron source, if a thermal neutron detector is used to count the slow-down neutrons while discriminating against the fast neutrons. Detection of slow-down (thermal) neutrons vs. fast neutrons can be done by Boron-10, Li-6, and He-3 detectors.

Table 1: Slowing-down parameters of various substances

Parameter	Scattering Substances							
	H	Li	C	N	Al	Ar	Fe	U
A	1	7	12	14	27	40	56	238
α	0	0.5625	0.7160	0.7511	0.9048	0.9048	0.9311	0.9833
ζ	1.000	0.2603	0.1578	0.1363	0.0723	0.0492	0.0353	0.0084
N	18	70	115	133	252	370	515	2170

Experimental

A Boron-10 thermal neutron counter was used for the detection of thermal neutrons. The counter was filled with highly enriched $^{10}\text{BF}_3$ gas. Incident neutrons have a nuclear reaction with Boron as $^{10}\text{B}(n, \alpha)^7\text{Li}$, while the detection efficiency, ε , for a neutron of energy E (in eV) is expressed as [7]:

$$\varepsilon = 1 - \exp(-1.7 \times 10^{-2} pL / \sqrt{E}) \quad (2)$$

where p is the gas pressure (atm) and L is the length (cm) of gas chamber. When $p=1$ atm. and $L=20$ cm, the detection efficiency, $\varepsilon \approx 0.9$ for neutron of 0.0253 eV but is only 0.03 for neutrons of 100 eV. This selectivity allows the detector to achieve the discrimination of fast and slow neutrons in its signal counting.

A simple setup was assembled [8]. It consisted of a ^{252}Cf neutron source, a calibrated portable thermal neutron counter ($^{10}\text{BF}_3$), and containers filled with a mixture of Al scrap and varying known amounts of H-containing material. The Experimental Setup is shown in Figure 2. A sealed radioisotope (^{252}Cf) neutron source (17.8 μg) was inserted into a cavity of a well shielded container (Atkinson Steel #263). The ^{252}Cf (half-life 2.6 y) source emitted neutrons of 2.2 MeV in 4π directions, but due to the shielding only a beam of fast neutrons centered along the container cavity were emitted out of the container when the top plug of the container was removed. A bucket holding pre-weighed dry Al scrap was placed on the top of the shielded container with spacers in between. The distance between the ^{252}Cf source and the bottom of the bucket was 38 cm. The direction of the fast neutron beam was set upwards to minimize the background counts due to neutron scattering from the floor.

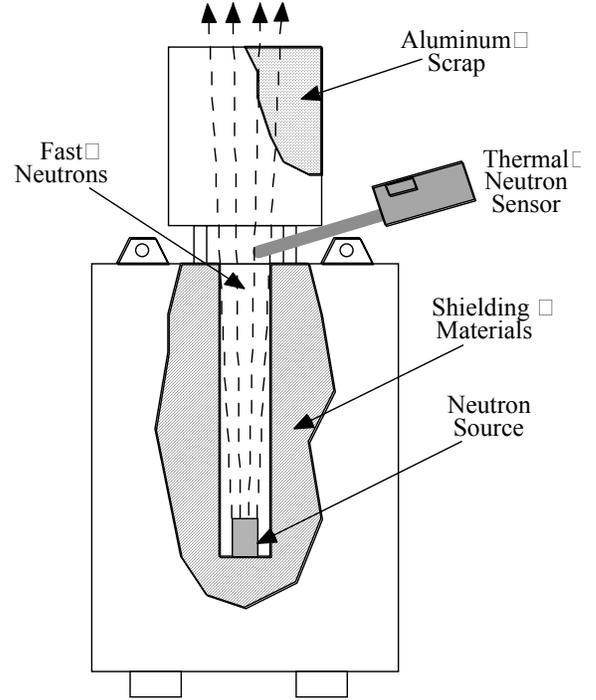


Figure 2: Schematic illustration of the experimental setup for measuring moisture content in aluminum scrap.

The experimental steps are: 1.) weigh Al scrap (pre-dried in a 200°C oven for 10 hours) with a bucket (metal or plastic); 2.) remove the top plug of the neutron shielded container, place wood spacers around the opening of the cavity and place the bucket of

Al scrap on top of spacers (so as to keep the distance between the ^{252}Cf source and the bottom of the bucket constant at 38.0 cm); 3.) take readings of the thermal neutron counts (background counts) with a calibrated portable thermal neutron counter (Q-2824-1, counts/cm²/sec) at the position between the bucket and the shield container, where a maximum count of thermal neutrons was observed; 4.) weigh desired amount of hydrogen-containing material, either polyethylene beans (Poly-beans, hydrogen content: 14.3%) by an electronic balance or water (hydrogen content: 11.1%) by a graduated cylinder and convert to weight based on 1.0 g per mL; 5.) remove the bucket away from the ^{252}Cf source and mechanically mix the Al scrap with pre-weighed polyethylene beans; in the case of water addition, the pre-measured water was evenly sprayed on the top surface of the Al scrap in the bucket and a few minutes of waiting was necessary; calculate and record the percentage of H-containing material weight vs. total mass of the mixture inside the bucket; 6.) place the bucket back onto the

spacers and take readings of the thermal neutron count at a position where a maximum of thermal neutrons is achieved; 7.) repeat steps 5) and 6) to add more H-containing material to the desired percentage ratio and take the back-scattering thermal neutron counts correspondingly; 8.) put the plug back onto the neutron shielded container after each run of the experiment to confine the neutron beam in the shielded container.

Table 2 lists experimental conditions of four runs. In the first two experiments, water was used to demonstrate that moisture could be detected using the thermal neutron detector. In experiment No.1, five glass cylinders were placed in the aluminum scrap. Water was added into the cylinders. In experiment No.2, water was sprayed on the top of the aluminum scrap. In the last two experiments, poly-beans were used because they could be mixed uniformly in the aluminum scrap.

Table 2: Experimental conditions

No.	Al Scrap(g)	Bucket (cm)	H-Containing Material
1	13600	Paper board $\Phi 38 \times 50.8$	Water
2	600	Metal $\Phi 15.5 \times 16.5$	Water
3	2000	Plastic $\Phi(21.5-25) \times 24$	Poly-beans
4	600	Metal $\Phi 15.5 \times 16.5$	Poly-beans

Results

Figure 3 shows the results of the No.1 experiment listed in Table 2. Each point on Figure 3 represents one experimental date. The background noise contributed to a thermal neutron count of about 10. With the addition of a small fraction (0.00037) of water, the thermal neutron count jumped to 25, 250% higher than that of the background noise. This result indicates that neutrons are extremely sensitive in determining the existence of moisture in aluminum. When the moisture fraction in aluminum scrap was 0.0037, the detected thermal neutron counts were 35. The thermal neutron counts then increased slightly with further increases in the addition of water in the scrap aluminum, a bit discouraging since we were expecting somewhat linear response of the thermal neutron counts with moisture content in the aluminum scrap. This could be resulted from the fact that water distribution was highly non-uniform because, in this experiment, water was added in the glass cylinders placed in the aluminum scrap.

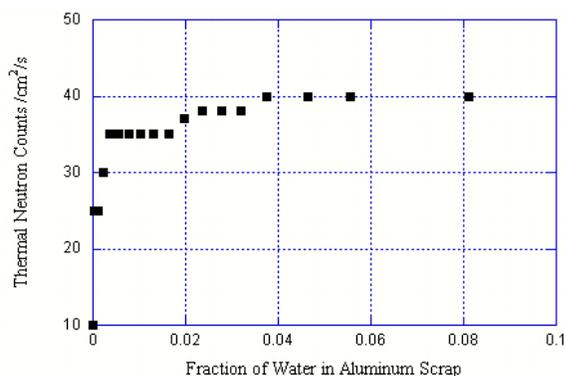


Figure 3: The relationship between the thermal neutron counts and the amount of water in aluminum scrap contained in a paper board.

In order to improve the distribution of water in the aluminum scrap, a second experiment was designed to spray water on top of the aluminum scrap. A small container was used. Figure 4 shows the relationship between the thermal neutron counts and the water content obtained in the No.2 experiment. The detected thermal neutron counts increased monotonically with the increase of water in aluminum scrap. This was an improvement in term of the shape of the curve comparing with that in Figure 3. However water distribution in the No.2 experiment was also non-uniform. Figure 4 also shows that the background noise is higher than that shown in Figure 3. This was due to the fact that the spacers supporting the container collected some moisture before the No.2 experiment started.

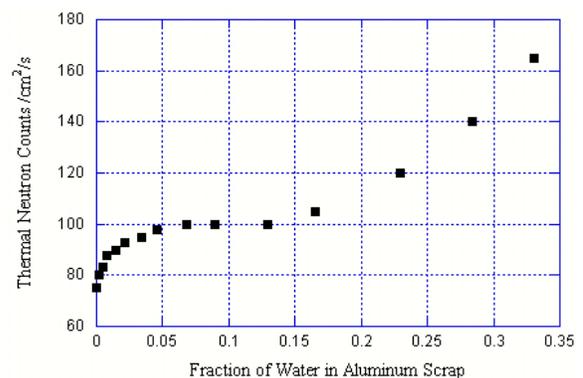


Figure 4: The relationship between the thermal neutron counts and the amount of water in aluminum scrap contained in a metal basket.

Figure 5 shows the results of the No.3 experiment listed in Table 2. In this experiment, small poly-beans were used because these beans contain hydrogen that can be detected in the same way as

water can be detected using neutrons. Besides the small poly-beans do not evaporate and can be mixed uniformly in aluminum scrap. Figure 5 indicates that there is a sharp increase in thermal neutron counts with a small amount of addition of poly-beans in the aluminum scrap. The thermal neutron counts then increased linearly with further increases in the addition of poly-beans in the scrap aluminum.

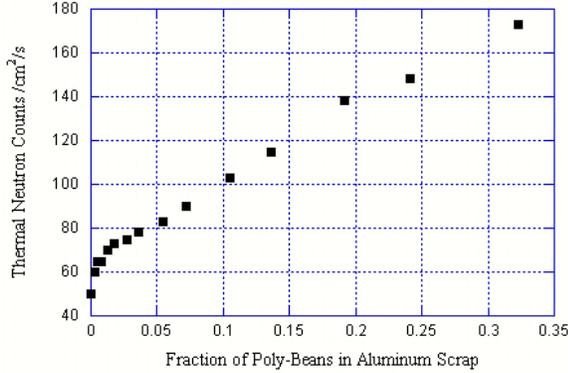


Figure 5: The relationship between the thermal neutron counts and the amount of added poly-beans in aluminum scrap contained in a plastic basket.

Figure 6 shows the results of the No.4 experiment listed in Table 2. In this case, a smaller container was used. Similar to the No.3 experiment, thermal neutron counts increased sharply with the addition of a small fraction of poly-beans, indicating that the thermal neutron counts are sensitive to hydrogen in the test basket. The results shown in Figures 5 and 6 represent the cases where hydrogen from the poly-beans is uniformly distributed in the aluminum scrap. In these two cases, the detected thermal neutron count increases with increasing hydrogen concentration following a simple relationship. These two tests are a strong indication that the neutron detection method can be used for quickly detecting and measuring hydrogen content in aluminum scrap.

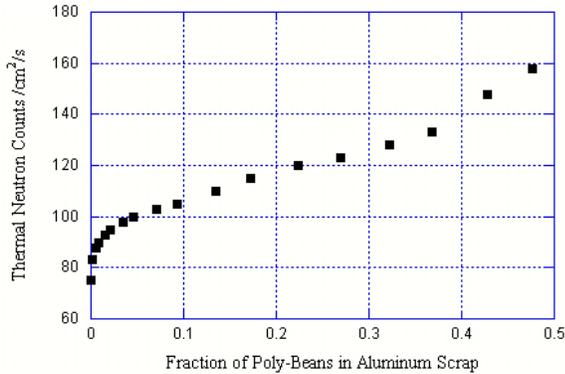


Figure 6: The relationship between the thermal neutron counts and the amount of added poly-beans in aluminum scrap contained in a metal basket.

Discussions

If the ²⁵²Cf source at the bottom of the cavity of the shielded container is considered as a point source, the fast neutrons emit out of the container upwards as a beam of diameter of $\Phi 10$ cm (the diameter of the open cavity of the container). With the beam

of fast neutrons interacting with the polyethylene beans or water inside the test bucket, back-scattering thermal neutrons are detected with a portable thermal neutron counter while maxima readings are obtained at the position between the bucket and shielded container. The more H-containing material in the bucket, the more back-scattering thermal neutrons are detected under the same experimental conditions. This tendency is evident in all four runs of the experiment. When the percentage of H-containing material in the test bucket changes from zero to 32% or 47%, the detected thermal neutron counts increase from a background count (50 – 70 counts/s/cm²) up to 160 – 170 counts/s/cm².

Fast neutrons lose energy and become thermal neutrons due to their interactions with the hydrogen atoms. The number of interaction “events”, Ψ , is theoretically proportional to the fast neutron beam intensity, J (the number of neutrons which travel across one cm² surface perpendicular to the beam direction per second) and the hydrogen density, D (the number of hydrogen atomic nuclei per cm³) [7]:

$$\psi = \sigma \times J \times D \quad (3)$$

where the proportionality constant σ is termed as interaction (scattering and absorption) cross section.

Equation 3 indicates that either J (fast neutron intensity) or D (hydrogen density in the tested mixture) must be increased in order to increase Ψ (the number of interaction “events”). In this work the intensity of the fast neutron source, J , was kept constant (the same source and the same distance between source and tested object), but the hydrogen density, D , in the test bucket was changed with the addition of the poly-beans or water. However we don’t observe a single slope or linear correlation when plotting detected thermal neutron counts (Ψ related) vs. the hydrogen density, D , in the test bucket as indicated by Equation 3.

The complexity of the real experimental conditions is beyond what Equation 3 governs. Equation 3 describes a relationship in a homogeneous system. The material tested in this work was a heterogeneous system of aluminum scrap and either poly-beans or water. Equation 3 can be extended to describe a multi-component system:

$$\psi = J(\sigma_H D_H + \sigma_{Al} D_{Al} + \dots) = J\Sigma \quad (4)$$

Plotting thermal neutron counts vs. D_H using Equation 4 will still yield a linear correlation if the densities of the components (D_H , D_{Al} , etc) are constant, when J and σ ’s are known constant coefficients. With different grain sizes or specific gravities of aluminum scrap and polyethylene beans, mechanical mixing won’t ensure uniform distribution of the two components as a semi-homogeneous mixture.

Distribution of water in our experiments was highly non-uniform. After being sprayed onto the top surface of the aluminum scrap inside the bucket, water started permeating the aluminum scrap and gradually accumulated at the bottom of the test bucket. The No. 1 experiment listed in Table 2 is another example of a non-homogenous system. An assumption for D_H is that it is uniformly represented by a percentage of the total water added into the bucket vs. the 13605 g of the total aluminum scrap. But D_H is not a constant value when the water is not uniformly distributed in the aluminum scrap and when the fast neutron beam of $\Phi 10$ cm (the

size of the cavity opening) is aimed at the test bucket bottom of $\Phi 38$ cm. This can account for the non-linear nature of the relationship between the thermal neutron count and the water content in aluminum scrap.

In most cases in the aluminum remelting industry, the water in aluminum is non-uniform. Distinct spots or locations of water exist in the voids of aluminum RSI or in the aluminum scrap. Detection of water with a neutron moisture gauge could be used to scan the object with fast neutrons and measure the thermal neutron counts with a thermal neutron counter. Any spot of water that is caught by scanning of the fast neutron beam will generate a substantially higher thermal neutron reading on a thermal neutron counter than the pre-measured background counts. Thus neutrons can be used to check if aluminum scrap or RSI contain moisture. This will be useful for preventing water explosions during remelting of aluminum alloys.

It is worth noting that a neutron source much smaller than the 17.8 μg of ^{252}Cf source used in this work can be used if the distance of neutron source to the tested object is shorter than 38 cm. A 17.8 μg ^{252}Cf source generates fast neutrons at the rate of 4.12×10^7 n/s/cm² at a distance of 1.0 cm, but only of 2.85×10^4 n/s/cm² at a distance of 38 cm because the neutron intensity is inversely proportional to the square of the distance ($I = 1/r^2$). A simple calculation shows that a 0.8 μg ^{252}Cf source would generate fast neutrons at same intensity at a distance of 8 cm as that of a 17.8 μg ^{252}Cf source at a distance of 38 cm. The use of a small ^{252}Cf source can lead not only to a lower purchase cost for the neutron source, but also to a much smaller size of the shielded source container. This may be an important consideration for designing portable neutron detectors for measuring water content in scrap aluminum alloys.

Conclusions

A neutron source (^{252}Cf) has been used for the detection of moisture in aluminum scrap. Preliminary experimental results indicated that neutrons are sensitive in detecting hydrogen (moisture) in aluminum. The detected thermal neutron count increases sharply with the addition of a small fraction of hydrogen-containing materials such as water or polyethylene beans. When hydrogen (water) is distributed uniformly in the aluminum scrap, the detected neutron counts increase linearly with increasing hydrogen content. In aluminum scrap with non-uniform distributions of water, the relationship between the detected thermal neutron counts and the water content deviates from a linear curve. Even under these conditions the detected neutron count increases with increasing water content. This work indicates that neutrons have the potential to be used by the aluminum remelting industries for rapid, non-destructive detection of water in aluminum scrap or RSI.

Acknowledgment

This research was supported by the United States Department of Energy, Office of Energy Efficiency and Renewable Energy, Industrial Technologies Program, Industrial Materials for the Future, Aluminum Industry of the Future, Materials Processing Laboratory Users (MPLUS) Facility, under contract No. DE-AC05-00OR22725 with UT-Battelle, LLC. The authors would like to thank IMCO Recycling, Inc. for providing the aluminum scrap.

References

1. S. G. Epstein, "Update on Molten Aluminum Incident Reporting," *Light Metals 1997*, ed. R. Huglen, (Warrendale, The Minerals, Metals & Materials Society, 1997), 887-895.
2. A. Giron, and J.E. Jacoby, "Design of Molds to Minimize Internal Shrinkage Cavities in Sows," *Light Metals 1993*, ed. S.K. Das, (Warrendale, The Minerals, Metals & Materials Society, 1992), 855-861.
3. R. D. Peterson, R. D. White, and R. Downie, "Development and Verification of the DELPEC Sow Mold," *Light Metals 1992*, ed. Euel R. Cutshall, (Warrendale, The Minerals, Metals & Materials Society, 1991), 999-1005.
4. H. J. Paap, and H. D. Scott, "The Use of ^{252}Cf as a Neutron Source for Well Logging," *Neutron Sources and Applications* (Proceedings of the American Nuclear Society National Topical Meeting, Augusta, Georgia, CONF-710402, Vol. III, April 19-21, 1971), 30-42.
5. V.I. Ferronsky, "Neutron Techniques in Ground Exploration," "Neutron Physics," *Neutron Sources and Applications* (Proceedings of the American Nuclear Society National Topical Meeting, Augusta, Georgia, CONF-710402, Vol. III, April 19-21, 1971), 48-58.
6. Bonin, H. W. and Thorp, C. J., "Design of a Neutron Gauge for the Detection and Measurement of Water Ingression in Flat Roofs," *Nuclear Technology*, 95(1991), 337-348.
7. Beckurts, K. H. and Wirtz, K., *Neutron Physics* (Springer-Verlag OHG, New York, 1964), 3, 117-121.
8. Q. Han, M. Du, and R.D. Peterson, "The Measurement of Moisture/Water Content in Recycled Aluminum Scrap and RSI," MPLUS Report, Metals and Ceramics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 2004.