

## APPLICATION OF RADIOLOGICAL IMAGING METHODS TO RADIOACTIVE WASTE CHARACTERIZATION

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### ABSTRACT

Radiological imaging technologies are most frequently used for medical diagnostic purposes but are also useful in materials characterization and other non-medical applications in research and industry. The characterization of radioactive waste packages or waste samples can also benefit from these techniques. In this paper, the application of some imaging methods is examined for the physical characterization of radioactive wastes constituted by spent ion-exchange resins and activated charcoal beds stored at the Radioactive Waste Management Department of IPEN. These wastes are generated when the filter media of the water polishing system of the IEA-R1 Nuclear Research Reactor is no longer able to maintain the required water quality and are replaced. The IEA-R1 is a 5MW pool-type reactor, moderated and cooled by light water, and fission and activation products released from the reactor core must be continuously removed to prevent activity buildup in the water. The replacement of the sorbents is carried out by pumping from the filter tanks into several 200 L drums, each drum getting a variable amount of water. Considering that the results of radioanalytical methods to determine the concentrations of radionuclides are usually expressed on dry basis, the amount of water must be known to calculate the total activity of each package. At first sight this is a trivial problem that demanded, however some effort to be solved. The findings on this subject are reported in this paper.

**Keywords:** waste, characterization, imaging, radioactive

### 1. INTRODUCTION

Imaging methods have been widely used for the characterization of various materials such as polymers and rocks. These methods such as Nuclear Magnetic Resonance (NMR), x-ray and Neutronography enables visualization of both heights, as the present compounds (through the use of NMR spectroscopy). In this paper, the application of some imaging methods is examined to evaluate the masses height stored in drums with radioactive waste at Radioactive Waste Management Laboratory (RWL), situated at the Energy and Nuclear Research Institute (IPEN), São Paulo, Brazil.

The RWL is responsible for performing the temporary storage and treatment of waste generated by the Nuclear and Energy Research Institute (IPEN), located in São Paulo, Brazil. The IPEN possess a nuclear research reactor pool type, called the IEA-R1, which is used for the production of radioisotopes and irradiation experiments and has beds of ion exchange

resin and activated carbon, which are responsible for cleaning and retreatment of water present in the reactor. These materials have particular characteristics and optimal for the occurrence of purifying water in the reactor. Activated charcoal is used due to its high surface area and its ability to adsorb a wide variety of components, and presents lower cost when compared to other materials sorbents. And the ion exchange resins are used for their ability to form complexes with heavy metals [1, 2].

The water in the reactor is used as coolant and as moderator biological shield. Fission products, released by the reactor core, as well other impurities in the coolant are retained in the resin and coal to maintain water quality within acceptable operating limits. Waste is generated when the absorptive capacity of the bed is no longer enough to keep the reactor operation within the established parameters, and then these beds of ion exchange resin and activated charcoal are disposed of as radioactive waste and replaced with new beds [3].

The RWL has the responsibility to direct the correct treatment of the wastes so that they comply the safety requirements established by Brazilian legislation. Therefore, several steps are necessary, including the characterization. In characterization stage there are some steps, including the determination of radioisotopes contained in the tailings. To determine this level, it is necessary to know quantitatively what is liquid, solid and interstitial phases in the waste. One method is to perform the measurement of heights, is to calculate the masses of liquid and solid. However, the method of observation alone can generate large margins of error, since it is possible that there are air bubbles or gaps in waste drums [3].

Currently, the designation of the content of radioisotopes in dry basis is performed by radiochemical and radiometric analysis; however, the tailings that are stored in the RWL are not characterized in this way, since they have adsorbent and water. Also, these masses are at many different levels compared between the drums themselves, due to the method used for pumping empty the reactor beds. Therefore, it is necessary to analyze each waste drum individually, requiring an optimization process and for that, imaging methods were used. This work used two imaging methods, which are Nuclear Magnetic Resonance (NMR) and Neutronography.

The attributes of NMR originate in the interaction between an atom in an external magnetic field, more precisely, is a phenomenon in which particles containing angular and magnetic momentum exhibits a precession movement when under the action of a magnetic field. A magnetic resonance imaging is the result of the interaction of the strong magnetic field produced by the equipment with the hydrogen protons creating a condition to send a pulse of radio frequency and, after collecting radiofrequency modified through a coil or receiving antenna. The collected signal is processed and converted into an image or information [4, 5].

The Neutronography is a technique that involves three components: an appropriate neutron flux, the object to be investigated and a medium containing radiographic film recorder with a neutrons converter in secondary radiation sensitize the film, just as in conventional techniques using  $\gamma$  rays and X-rays [6, 7].

The objective of this study was to observe the behavior of the samples using non-destructive testing such as MRI technique and radiographic imaging with neutrons to characterize the content present in the samples.

## 2. EXPERIMENTAL

To prepare samples of activated charcoal was necessary the saturation of the material. Therefore, coal was immersed in distilled water for about 96 hours (4 days). Soon after the saturation process, activated charcoal and the supernatant passed through vacuum filtration, using a vacuum pump, quantitative filter paper black belt (for rapid filtration) and gelatinous precipitates, Büchner funnel.

For the samples of ion exchange resin (IRA-410), the process was the same, changing only the saturation time which was 72 hours (3 days).

To prepare the samples, we used non-radioactive materials.

### 2.1. Samples for NMR

Ten samples were prepared for use in NMR, five of ion exchange resin and activated charcoal with five different weights, both of adsorbent material as water with packaging made in plastic.

**Table 1: Activated charcoal samples (weight adsorber and water)**

	Dry Charcoal (g)	Saturated Charcoal (g)	Water (g)
Sample 1	100	0	0
Sample 2	0	187,4	0
Sample 3	0	180,1	84,1
Sample 4	0	124,1	107,3
Sample 5	0	102,8	229,7



**Figure 1: Samples of activated charcoal**

**Table 2: Ion exchange resin samples (weight adsorber and water)**

	Dry resin (g)	Saturated resin (g)	Water (g)
Sample 1	224	0	0
Sample 2	0	191,2	0
Sample 3	0	200,1	59,5
Sample 4	0	191,5	45,9
Sample 5	0	159,2	142,3



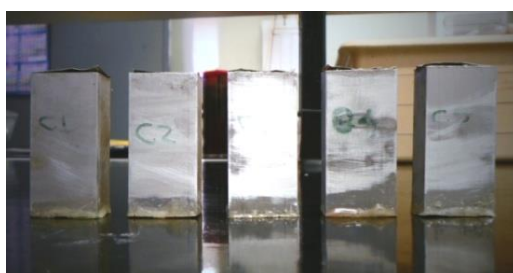
**Figure 2: Samples of ion exchange resin**

## 2.2. Samples for neutrongraphy

As for NMR, ten samples were also prepared, differing only in the storage of the adsorptive material and water. In this case, containers comprise aluminum tube with 1.52 mm thickness and 5 cm height.

**Table 3: Activated charcoal samples (weight adsorber and water)**

	Dry Charcoal (g)	Saturated Charcoal (g)	Water (g)
Sample 1	1,9	0	0
Sample 2	0	4,9	0,8
Sample 3	0	4,6	1,2
Sample 4	0	4,7	3,4
Sample 5	0	3,4	0



**Figure 3: Samples of activated charcoal**

**Table 4: Ion exchange resin samples (weight adsorber and water)**

	Dry resin (g)	Saturated resin (g)	Water (g)
Sample 1	224	0	0
Sample 2	0	191,2	0
Sample 3	0	200,1	59,5
Sample 4	0	191,5	45,9
Sample 5	0	159,2	142,3



**Figure 4: Samples of ion exchange resin**

### **2.3. NMR method**

To perform the NMR samples was used a device from Siemens of 3 Tesla.

The samples (Figs. 1 and 2) were submitted to analysis being accommodated inside a skull coil in a vertical position.

Image acquisition was performed in two stages, initially positioning the coal samples and then immediately samples of resin and subjecting both in acquisitions for T1 and T2. Later in PD.

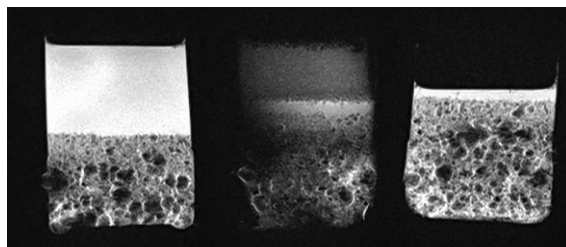
### **2.4. Neutronography method**

To perform the Neutronography, was used the IEA-R1 reactor. The samples (Fig. 3 and 4) were placed in a buffer suitable for Neutronography and images were obtained from 3,45.10<sup>13</sup> a beam of neutrons / cm<sup>2</sup>.s, 12 cm in diameter.

After exposure, it was necessary to wait for the radioactive decay due to the risk of neutron activation, with subsequent monitoring, ensuring the removal of samples from the reactor.

## **3. RESULTS**

With the evaluation method of magnetic resonance imaging, sagittal sections of charcoal samples 2, 4 and 5, these images were obtained:

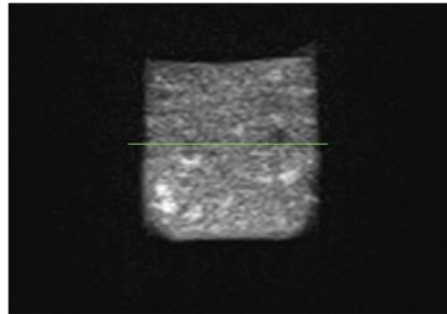


**Figure 5: Activated charcoal samples by NMR method in T1 ponderation**

Performing the weighted T1 and T2, we are not able to see water levels below the adsorbent material. Conducting tests positioning of samples and settings for image acquisition, we reached Proton density weighting, also known as PD.

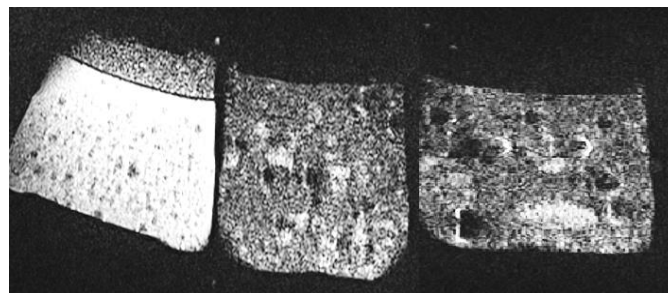
Through this ponderation was possible to visualize the water levels below the adsorbent material, in this case coal.

The acquired image (Fig. 6) shows delimited in line with the green water level (below the level of adsorbent material) in sample 3.



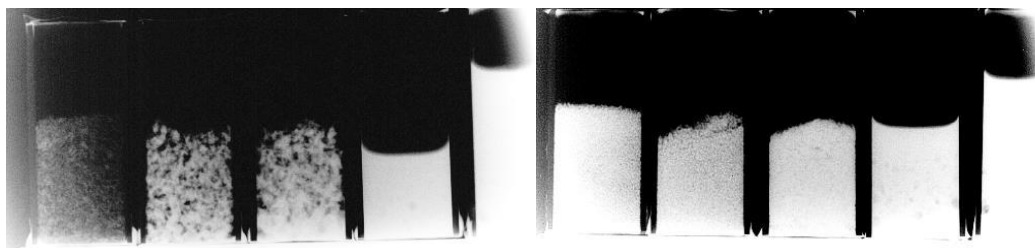
**Figure 6: Activated charcoal sample by NMR method in PD ponderation**

For the ion exchange resin, were imaged directly in PD. Clearly delimiting the water level below the adsorbent material in the first left image (sample 5).



**Figure 7: Ion exchange resin samples by NMR method in PD ponderation**

By the method of Neutrongraphy, these images were obtained:



**Figure 8: Activated charcoal and ion exchange resin samples by neutrongraphy method**

Although Neutronography makes good use for displaying the levels of adsorbent material, it was not possible to see the water levels in a completely clear way. In some pictures, it was possible to see some irregularly levels but due to its unclear way, give rise to an erroneous interpretation of the levels.

#### 4. CONCLUSION

Two techniques for image acquisition of simulated waste samples constituted of spent ion-exchange resins and activated charcoal were tested and compared. From the results it was possible to conclude that imaging techniques can be an effective method for physical characterization of radioactive waste. The NMR with proton density (PD) acquisition mode allows us to see the water levels in the samples, even when they are below the upper surface of adsorbent material. The NMR technique has a high cost (equipment and support), and there is the problem of interaction of the magnetic field generated by the equipment with steel of the waste packages, preventing its use, unless waste samples are drawn.

It is necessary some changes and adjustments in the neutronography technique, allowing a better view of the water levels in the samples. More studies are needed for the used techniques to achieve adjustments in image acquisition.

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