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STUDY OF ACID SOLUTION BONDING IN EPOXY MATRIX FOR SEALED RADIOACTIVE SOURCES PRODUCTION.

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ABSTRACT

The present work aims to analyze different resin formulations. These formulations are used in the production of radioactive sealed sources that are used in many fields such as nuclear medicine; environmental analyzes, radiation detectors accuracy check, and so on. These sources can be produced with different radioisotopes and different activities, it all depending on the use they will have. Certain types of resins have the same density water has. This property is appreciated when we consider that radiotracers used in nuclear medicine are applied in aqueous solutions. So the sources used for checking and calibrating equipment must have their radioisotopes sealed in a material having similar properties, thus the measures are reproducible and repetitive. The most important aspect that is brought to attention in this work is the miscibility those resins have with water. The radioisotopes for the production of the sources are supplied in an aqueous form. In case the resin and the radioisotope solution do not mix, the source will not be sturdy enough to seal the radioisotopes in its structure and the source will not be safe. There were prepared different formulations with different amounts of acid solution, and the cured formulations were analyzed by Wipe Test, DSC (Differential Splanatory Calorimetry) and also, the possible volatile aspect of the radionuclide used. All to evaluate the integrity of the sources. The obtained results were satisfactory and show that when the resin is well cured, the radioisotope remains sealed in the matrix, making it possible to produce radioactive sealed sources.

Keywords: brachytherapy, radioisotope, epoxy resin, radioactive sealed Sources

1. INTRODUCTION

Estimates of Globocan 2012, project of the International Agency for Research on Cancer (IARC - International Agency for Research on Cancer), of the World Health Organization (WHO), showed the occurrence of 14.1 million new cancer cases and a total 8.2 million deaths related to cancer worldwide in 2012 [1,2]. In 2030 it is estimated that the incidence is 21.4 million of new cases and 13.2 million deaths as a result of aging [1]. In Brazil, the Instituto Nacional do Câncer José Alencar Gomes da Silva (INCA) estimates that for the years 2014 and 2015 will be occurring around 576.000 cancer cases, including non-melanoma skin cases. The cases of non-melanoma skin cancers are the most frequent, around 182 thousand new cases. Followed by prostate, female breast, colon and rectum, lung, stomach and cervix. Not considering the cases of non-melanoma skin cancer the estimate for other types is 395 thousand new cases of cancer [1].

1.1. Nuclear Medicine

It is a medical specialty within the field of radiology that uses radioactive isotopes, nuclear radiation, charged particles, photons, and biophysical techniques, for preventive purposes, diagnostic, therapeutic and medical research [3].

Radioactive isotopes can be used to determine the target of compounds in the body. These studies begin with a compound that has a radioactive isotope as one of its constituent elements, the union of organic compounds and radioactive isotopes are known as radiotracers [4]. They are injected in patients and bound to tissues and bones according to their chemical affinity; the radiation emitted by these radioisotopes is then analyzed by a scintigraphic camera, generating two-dimensional or tomographic images.

The Comissão Nacional de Energia Nuclear, CNEN, created the standard "Requisitos de segurança e proteção radiológica para serviços de medicina nuclear" CNEN-NE-3.05, April 1996, which states that all nuclear medicine service must have standard reference sources of cobalt-57 and barium-133, for checking their radiation detectors [4].

1.2. Epoxy Matrix

Epoxides are ethers in rings of three members. The method used for their synthesis is the reaction of an alkene with an organic peroxide acidic, a process called epoxidation [5]. The process can be started simply with the addition of a catalyst such as an alkoxide or amine [6]. Solidified epoxy matrices in the glassy state have high compressive (500-700kg/cm^2) and adhesion strength (100kg/cm^2). Investigations of radiation resistance of some compounds, for example, have shown that the matrix remains unchanged with gamma radiation doses up to 10 000 Mrad, but has its elasticity increased and tensile fracture decreased with the radiation dose. No leakage was observed on sources produced with cesium-137 for a period of two years of testing [7].

Epoxy resins are thermosetting materials readily converted through the curing reaction with a variety of chemical compounds (curing agent). Most resins are obtained from the condensation of epichlorohydrin (1-chloro-2,3-epoxy propane), and Bisphenol A [2,2-bis(4-hgydroxyphenyl) propane], known as copolymers of diglycidyl ether bisphenol A or simply

DGEBA. They have high interest to be employed in the manufacture of polymeric immobilization for radioactive material [7,8], because:

- They are among the oldest resins of the epoxy class; they offer lower cost, availability and easy acquisition on the market.
- They have low toxicity and, consequently, low possibility of chemical contamination during handling.
- After the curing process it is obtained a polymeric material with high compressive and adhesion strength [7], with a high radiation resistance [9], as well as high resistance to thermal decomposition, which makes a material with high chemistry stability.
- Also originate, after curing, water-insoluble polymers, either in acid and alkaline environment, which guarantee any leakage or diffusion of the radioactive component [7].
- Commercial epoxy resins, in general, does not contain significant amounts of radioactive impurities, leading to a very low radioactive background, not compromising the total activity and calibration procedures of the sealed source.

1.3. ISO Standards

To ensure compliance with the requirements of radiological protection, standards for the development and manufacture of sealed sources were established by the rules:

- "Radiation protection - sealed radioactive sources - General requirements and classification" ISO 2919 [10].
- "Radiation protection - sealed radioactive sources - leakage test methods "ISO 9978 [11].

According to the standards, the sealed sources must be evaluated on several parameters. They must be classified by analyzing the toxicity of the radioisotope. Subsequently, tests must be performed to determine the performance of the product. These tests consist in exposing the sources to specific temperature, pressure, external vibration and puncture.

The approval in any of the tests will be determined by the ability of the sealed source to keep its sealing properties. After each test, the source must be visually examined for checking its integrity and must also be approved in leakage test, performed according to the standard "International Standard Organization Radiation protection - leakage test methods" ISO 9978 [3].

The leakage test can also be carried out by rubbing a fabric that can be moistened or not with water or ethanol. This tissue then has its activity examined. The activity must not exceed 0.2 kBq (≈5nCi) [12,13].

2. METHODOLOGY

Thermo gravimetric analyses (TGA) were performed on the equipment SDT Q600 from TA Instruments. We opted to use a nitrogen atmosphere (N_2) for no oxidation of the resin at a rate of 50 mL / min in a platinum pan. The temperature increase was 10°C / min up to 600°C. This equipment has the ability to perform thermo gravimetric and differential scanning calorimetry (DSC) analysis. To test the tightness of the source, there was prepared a piece of 5cm x 5cm with 10% of iodine-125 in the formulation.

The source was allowed to cure in a Petri dish lined with plastic wrap inside a chapel with laminar flow and air filter. The wipe test was performed according to the standard "Radiation protection - Sealed radioactive sources - General requirements and classification." And also with ISO 2919 standard "Radiation protection - Sealed radioactive sources - leakage test methods" ISO 9978 [10,11]. The standard suggests immersion tests in hot, boiling, cold, room temperature and liquid scintillator. Also suggests helium test, bubbles and wipe test. In wipe test, most commonly performed, the activity of the fabric used for wiping may not show higher activity than 0.2 kBq (≈ 5 nCi) [11].
The possibility of iodine-125 to volatilize and escape from the epoxy matrix was also evaluated in the source. For this test, we measured the activity of the source from time to time and compared the theoretical activity that the source should present at the time of measurement considering the decay of iodine-125. For activity measurements we used an ionization chamber detector Capintec CRC 15R.

3. RESULTS

The initial objective of thermo gravimetric analysis was to investigate the possible evaporation of water during the temperature increase achieved by the analysis itself. This evaporation is related to tightness of the source, if any volume of water evaporates, it could drag radioactive material or destabilize the epoxy matrix and cause leakage of the source.
Then it was investigated the weight change of samples made with formulations varying the amount of iodine-125. Figure 1 shows the curves obtained in thermo gravimetric analysis.

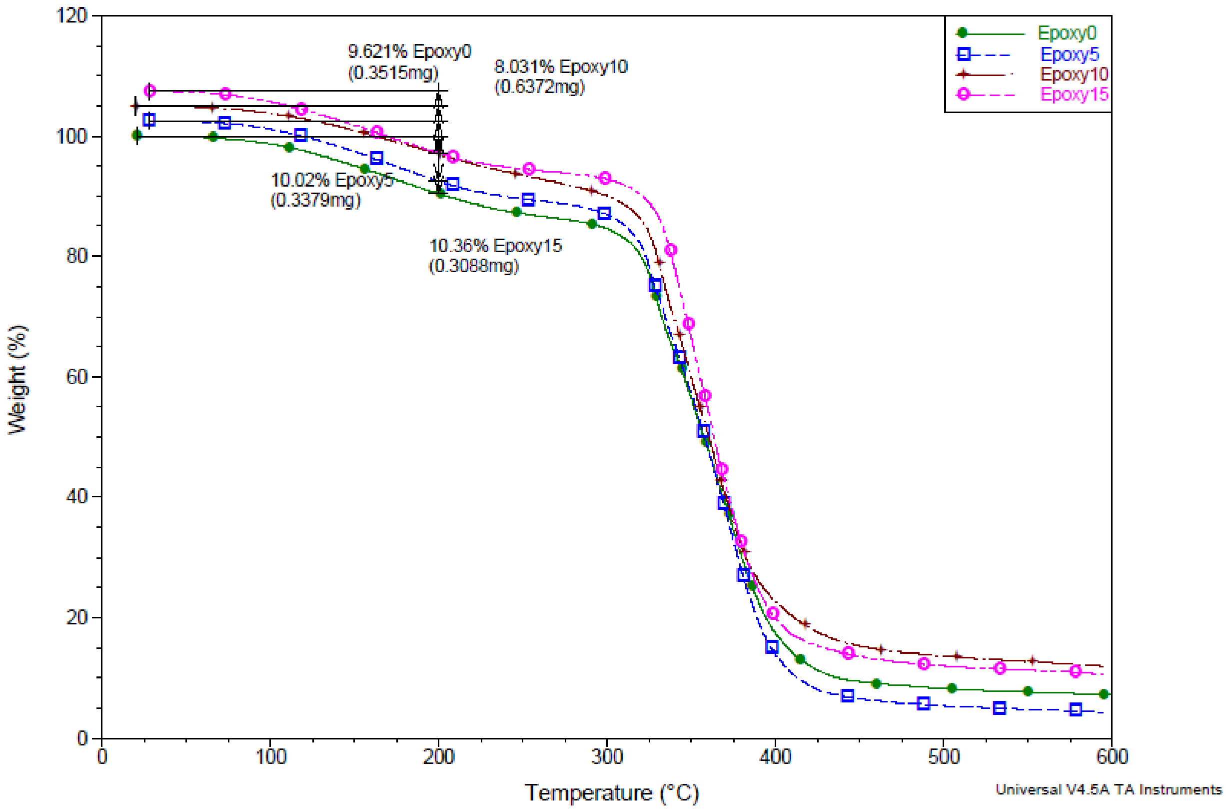

FIGURE 1. Degradation degree at 200°C

There was no proportional difference in the weight change for the different concentrations of acid solution added to the formulations. The added water is chemically bound to the matrix and thus all formulations show they are tight in temperatures up to 200°C. Table 1 shows the values of the degradation degree obtained in thermo gravimetric analysis.

TABLE 1. Degradation degree of different epoxy formulations at 200ºC

Formulation	Weight change at 200ºC (%)
Epoxy0	9.621
Epoxy5	10.02
Epoxy10	8.031
Epoxy15	10.36

During data analysis, it was noticed that the values of the derivative TGA graph of Epoxy15 sample had a different behavior from the other derivatives. Thus, it was decided to repeat the analysis of the sample Epoxy15. The data obtained is shown in Figure 2.

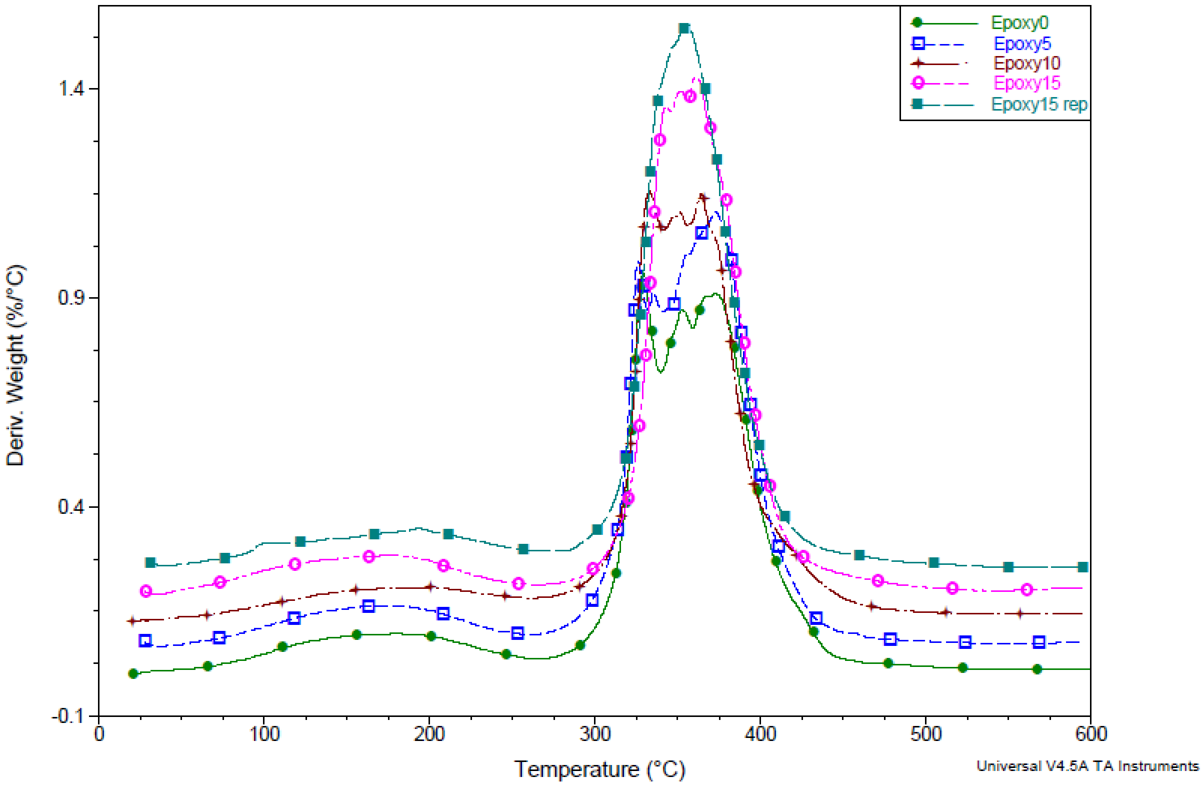

FIGURE 2. Behavior of the derivatives of weight change curves.

A sample analysis repetition was performed in Epoxy15 about a month after the first analysis, and shows a tendency to form a single peak. Unlike the first one that had three small and close peaks the other samples showed different peaks. This fact showed that the resin may not have had a complete cure at the time of the first analysis. Even though it cured for 24 hours, as suggested by the manufacturer.

The results of the activity measurements of the source and the wipe test in Table 2.

TABLE 2. Results of activity and wipe test measurements.

Measurement date	Measurement time	Measured acitivity (µCi)	Theoretical activity (µCi)	Variation (%)	Wipe test (µCi)
April 22	10:23	**641**	-	-	0
April 23	15:43	598	631.92	5.67%	0
May 5	15:40	537	549.37	2.30%	0
June 10	15:07	368	361.05	-1.89%	0
June 13	15:32	339	348.56	2.82%	0

For the calculations we adopted the first measurement of the source completely cured. There was not any parameter of a satisfactory variation in activity between the measured and theoretical activity. But we can consider that the lack of knowledge of the best way to perform the activity measurement of this type of source, as well as the interference between the way that the detector is calibrated and geometry of the source, may have interfered for the percentage of variation presented.
However, all wiping tests showed no activity in the detector, assuring that the source is considered tight.

3. CONCLUSIONS

We see by the thermo gravimetric analysis that the added water from the iodine-125 solution is chemically bonded to the structure and is not released with increasing temperature, up to 200ºC. As the amount of iodine-125 is increased in the formulation, the behavior of the weight change derivative changes, it may show a tendency of properties loss of the resin.
The monitoring of the possible evaporation of iodine-125 system and wipe tests showed that the source is tight. However dosimetry studies must be done to decreased the errors related to the geometry of the source during detection.

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