DEVELOPMENT OF RADIOACTIVE SOURCES FIXATED IN EPOXY MATRIX TO VERIFICATE NUCLEAR MEDICINE EQUIPMENTS

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ABSTRACT

The aim of the present work is to study and develop commercial resins for manufacturing solid sealed sources. The sources are produced with radionuclides of barium-133, cesium-137 and cobalt-57. They are used in radiation detectors verification. For the immobilization of the radionuclides in the epoxy matrix, it is made use of emulsifying agents that ensure the miscibility between resin and aqueous radioactive solution, as well as curing agents for controlling, curing and sealing the standard radioactive solution completely. As a result, it is expected to obtain standard water-equivalent sealed sources (the radioisotopes used in Nuclear Medicine are supplied in an aqueous form and the resin applied must have a very similar density comparing to the water). We used DGEBA epoxy matrix and DETA polyamic modified catalyst, and obtained great results. The epoxy resin cure can be improved in relation to the cure at room temperature, with use of irradiation, since it is used a dose around 37kGy during curing.

1. Introduction

The activity detectors, shown in Fig. 1, are devices widely used by nuclear medicine services, to assess the activity of the radioisotopes used in diagnostic and therapeutic purposes. The measurement of the activity of these radioisotopes must be done with accuracy because it will be injected in a patient.[1]

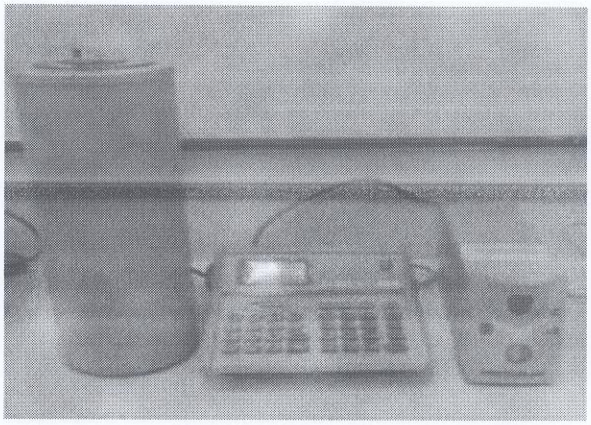

Figure 1: Example of an Activity Detector, Capintec CRC 15R.[2]

Using radioactive sealed sources makes the process of checking detector's accuracy easier, due to the fact that the production of these sources with more available raw-materials and processes facilitates the supply of the local demands. These sources are prepared with radioisotope solutions of cobalt-57, cesium-137 and barium-133, with the final activity of 185 MBq, 9.3 MBq and 5.4 MBq, respectively, a picture of the sources is shown in Fig. 2. [2]

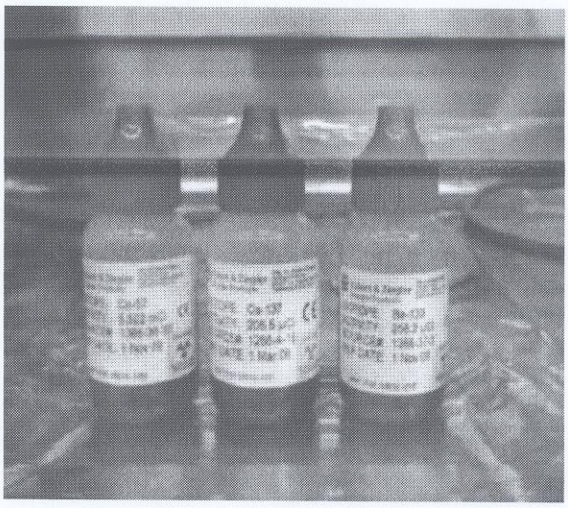

Figure 2: Sealed radioactive sources, sealed in an epoxy matrix (brown colour), produced with cobalt-57, cesium-137 and barium-133. [2]

These capsules and materials must be sturdy enough to prevent any radioactive material leakage under normal use.[3] After producing, the source must be visually exanimated to ensure its integrity and must also be approved in a leakage test, performed according to the standard "International Standard Organization Radiation protection - leakage test methods" ISO 9978. [2]

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.[4]

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. [2] They are injected in patients and bound to tissues and bones according to their chemical affinity; the radioactivity generated by these radioisotopes is then analysed by a scintigraphic camera, generating two-dimensional or tomographic images.

The National Nuclear Energy Commission, CNEN, created the standard "Requirements for Radiation Protection and Security for Nuclear Medicine Services" 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. [2]

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/cm²) and adhesion strength (100kg/cm²). 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, [10] 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[11]
- "Radiation protection - sealed radioactive sources - leakage test methods "ISO 9978.[12]

According to the standards, the sealed sources must be evaluated on several parameters. They must be classified by analysing 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. [2]

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). [3, 10]

2. Methodology

The immobilization of radionuclides in sealed sources using matrices prepared with epoxy resin DGEBA stumbles into two problems:

- The miscibility of the epoxy resins with aqueous solutions. The sources of barium-133, cesium-137 and cobalt-57 are supplied commercially in acidic aqueous solutions. They are rarely supplied in the form of solid compounds. The epoxy resin and the aqueous solution are not easily miscible.
- When the aqueous solution is added to the resin, it decreases the mechanical and chemical resistance of the resin,

Both problems can be overcome. The miscibility issue can be minimized with emulsifiers and curing agents that are miscible in the epoxy resin. For the related loss of properties, especially the curing efficiency, it can be corrected by employing irradiation during the curing process. [13]

We used as basic formulation for the trials an epoxy resin (diglycidyl ether of bisphenol-A) "DGEBA" Silaex SQ 2004, manufactured by Silaex Chemicals Ltd., viscosity 500 to 700cps, epoxy equivalent weight 195-215 EEW and average density around 1.13g/cm³. As curing agent, we used a modified polyamine catalyst base (diethylenetriamine) "DETA" SQ 3131 Silaex, the same manufacturer of the resin, with a viscosity around 3000cPs and density 1.10g/cm³.

2.1. Acceleration of Epoxy DGEBA-DETA Curing

The curing process acceleration of the mixture epoxy resin (DGEBA) and the modified polyamic catalyst (DETA) was conducted in the Multipurpose Irradiator of Cobalt-60, situated at the Institute of Nuclear and Energetic Researches (IPEN) at doses of 20.8, 60.8, 116.1 and 153.7 kGy.

2.2. Curing Ability of Water-Containing Mixture at Room Temperature

The studies for testing the ability of water solubilisation were carried out using the mixture of epoxy (DGEBA) resin and polyamic catalyst (DETA), respectively, in the ratio of 100:50 parts by weight, in which were added varying amounts of an aqueous solution 1.0 M in HCℓ prepared in Mill-Q water, and so make up 5, 10, 15 and 20% by weight of water in resin-catalyst mixture.

2.3. Tensile Test

The tensile strength tests according to ASTM D 680 were made using 5kg load and a separation speed of 50mm/min until rupture in an Instron Model 5567.

3. Results

The addition of determined amounts of water to the epoxy resin-catalyst mixture does not affect the curing at room temperature. This curing process takes between 24 up to 48 hours, considering that the amount of water cannot exceed more than 20% of the epoxy system weight. Amounts above 20% lead to partially cured regions causing discontinued polymerization.

The tensile strength tests were conducted on pieces prepared with fixed amount of 100:50 parts by weight of epoxy resin and modified polyamic catalyst. The amounts of water, in form of an acid solution 1.0M HCℓ, were added in portions ranging from 0 to 20% by weight of the resin-catalyst system, as shown in Fig. 3. The results revealed that there is a significant loss of the material's tensile strength.

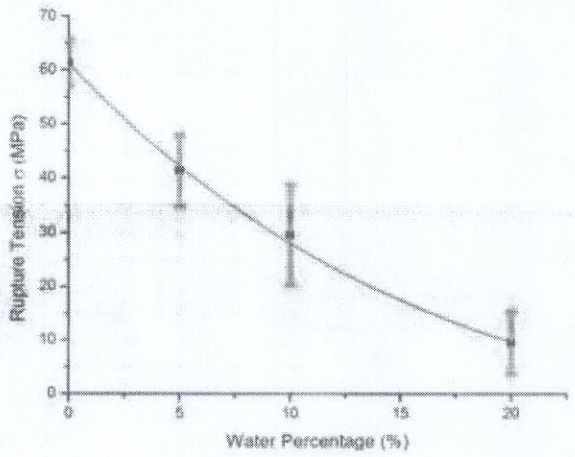

Figure 3: Rupture Tension *versus* Water Percentage.

The decrease of the material's resistance with water addition can be explained by the loss of its plastic properties. It is due to an increased crystallinity that occurs in this process. The elongation of the material becomes constant from 5% of water addition, as shown in Fig 4, in which it is represented the elongation of the pieces during the tensile test, depending on the percentage of water added to the catalysed epoxy mix.

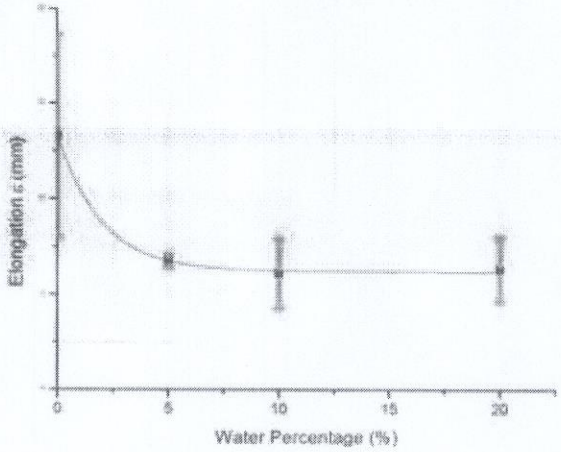

Figure 4: Elongation *versus* Water Percentage.

Tests to study how different doses of radiation can improve or not the material's properties were done. It was selected the same formulation using from the tensile test, but using the highest amount of water (1.0 M HCℓ) evaluated, 20% by weight. The dosages used were between 0 and 153.7 kGy.

After the tests, it was shown that irradiation enables increased strength in a dose around 37kGy, then it was observed an inverse process, in which a reduction of the tensile strength until it reaches a level close to 160kGy, where there seems to be a constant in tensile strength. However this tensile strength is higher than the initial state, when it was not used any dose, as shown in Fig.5.

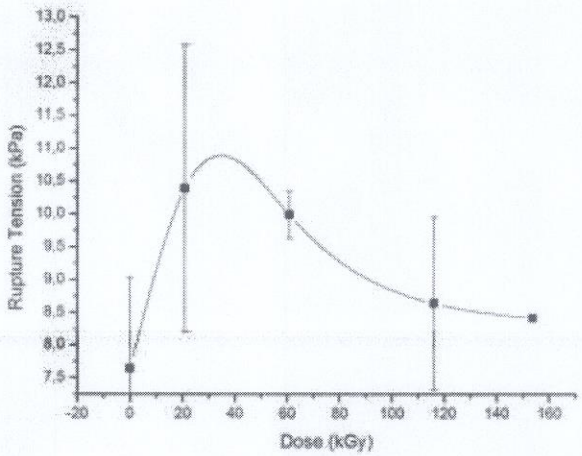

Figure 5: Rupture Tension *versus* Dose.

The increased strength up to a dose of 37kGy must be associated with the formation of additional cross-linking bonds in the polymerized material. On the other hand, the reduction of the resistance around 160kGy must be due to the disruption of some cross-linking bonds. From 160kGy, the threshold for the resistance, higher than the initial, but lower then doses around 37kGy, is therefore associated with the stabilization on the bonds rupture.

The water added to the composition of the epoxy matrix is fully incorporated into the polymer structure since there was no significant change in density during irradiation, as shown in Fig 6.

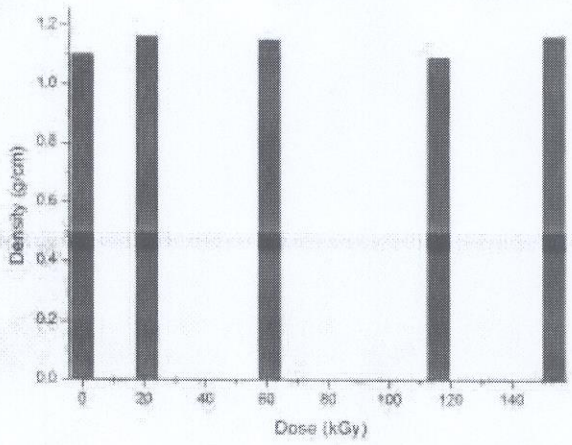

Figure 6: Density versus Dose.

4. Conclusions

Sealed sources can be made of DGEBA epoxy matrix and DETA polyamic modified catalyst, since the amount of radioactive material in the form of acidic solution does not exceed a content of 20% by weight.

The epoxy resin cure can be improved in relation to the cure at room temperature, with use of irradiation, since it is used a dose around 37kGy during curing.

The added amounts of water are, so far, completely incorporated to the cured resin system.

5. References

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