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# Usability of epoxy/ilmenite composite material as an attenuator for radiation and a restoration mortar for cracks

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ABSTRACT

In the study, the usability of epoxy/ilmenite (EP/IIm) composite material as a radiation shielding in many applications and a restoration/injection mortar for cracks developing in biological concrete shields was investigated. Radiation attenuation properties of EP/IIm composite material was searched out using $^{252}$Cf (100 μg) neutron source and gamma spectrometer with stilbene organic scintillator. Fast neutron fluence rates and gamma fluxes were measured and displayed as energy distributions and attenuation relations. Thermal neutron fluence rates were measured using BF3 detector and displayed as attenuation relations. The experimental attenuation parameters; macroscopic effective removal cross-section $\Sigma_R$ (cm$^{-1}$), total attenuation coefficient $\mu$ (cm$^{-1}$) and macroscopic cross-section $\Sigma$ (cm$^{-1}$) of fast neutrons, gamma rays and thermal neutron respectively have been determined. Theoretical calculations have been achieved using MCNP-4C2 code to evaluate the concerned parameters. Also, MERCSF – N program had been used to recalculate $\Sigma_R$ (cm$^{-1}$). The obtained experimental and theoretical results were compared, where a reasonable agreement was recognized.



## 1. Introduction

All nuclear installations acquire highly penetrating radiations which can adversely affect on receives whether they are personnel or sensitive equipments. For shielding against those radiations, neutrons and gamma rays are mainly considered since they have the most penetrating power, and give the main contribution to the dose outside the shield. Although, the processes by which both types of radiations interact with matter are considered fundamentals, unknown physical parameters called cross-section should be evaluated for each shielding situation (Jaeger, 1970; Goldstein, 1959).

Heavy concrete is by far the most widely used material for nuclear facilities biological shields because of its cheapness, its reasonable mechanical properties and its many physical attributes for an ideal shielding material for neutrons and gamma rays (Chilton, 1984). Any concrete structure may develop structural or non-structural cracks either by aging or any other cause. Cracks in reinforced concrete greater than approximately 1–2 mm require sealing/injection to prevent ingress of moisture, oxygen and other materials or for other reasons that could deteriorate the structure (Allen and Edwards, 1987). In addition, the nuclear facilities biological shield concrete structures deteriorate by the impact of increasing temperature and exposure to radiation. To restore those structures to their original strength, cured epoxy resin filled with crushed ilmenite aggregate is suggested as a repair mortar for In Situ Decommissioning (ISD) processes (El-Sayed Abdo et al., 2003a; Acevedo and Serrato, 2010).

This mortar will be satisfying the requirements of improved plastics such as high mechanical strength, adhesiveness and reasonable physical properties beside its main role in radiation shielding for this application. Since, epoxy resin consists of mainly light elements like C, O and H; it would perform good neutron moderation. In Addition, the Ilmenite filler, which is a heavy mineral, will play the key role in gamma ray and X-ray as well as fast neutron attenuation.

Recently, different researchers had contributed in providing works about different shielding materials such as heavy weight concrete properties by Sakr and El-Hakim (2005), high radiation shielding concretes by Sakr (2006), high-density concretes in radiotherapy by Facure and Silva (2007) and cracking effect on gamma-ray shielding by Lee et al. (2007).

In the present work, the composite physical properties; as specific heat, a.c. electrical conductivity, water absorption and porosity were investigated. Also, fast and thermal neutrons and gamma rays attenuation parameters were experimentally evaluated and theoretically calculated.

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## 2. Experimental and calculation methods

### 2.1. Materials and sample preparation

The standard Bisphenol-A based Epoxy resin (EP) with commercial name (DGEBA DER331 Product of DOW Chemical Company USA with technical purity 95% and epoxide weight: 182-192) hardened by polyoxyporopylendiamine (Cetepox 1465 H product of Chemical & Technologies for Polymers Co.) was used as the base polymeric formula. Ilmenite filler a product of (El-Nasr Phosphate Co. – Abu Galaka – Red Sea Mines – Egypt) was prepared by crushing the ore to mesh size −500 μm; certain weight of this filler was dispersed in the Epoxy formula (Resin 100 phr and Hardener 33 phr) to produce the epoxy/ilmenite (EP/Ilm) composite ($\rho$ = 2.716 g/cm$^3$) with 20 wt.%/80 wt.% loading fraction. The mixture was stirred at room temperature and degassed to allow the entrapped air to be released, then poured with great courtesy into the sample molds and left to cure. The sample molds were made of Teflon with according geometries. After 24 h, the samples were extruded from molds and left 7 days for ultimate cross-linking before they were shaped to the desired dimensions. Cylindrical samples of Ø = 10 cm and ≈5 cm in thickness were used for radiation attenuation measurements. The sample elemental analysis was obtained for both the Epoxy base formula and the Ilmenite filler. The "Elementar Analysensysteme GmbH – vario EL III Element Analyzer – Germany" was used to analyze the Epoxy at "Cairo University – Microanalytical Center – Giza, Egypt". Also, XRF was used to analyze Ilmenite at SGS, Birmingham, UK. The composite analysis and mechanical test results according to ASTM designations were presented elsewhere in (El-Sayed Abdo et al., 2003a).

Thermal properties was performed using dry fragments fragment samples ≈5 mg in weight and the "Parkin Elmer – Differential Scanning Calorimeter Dsc7-USA" to measure the specific heat in (J/g °C) versus to temperature rise in $T$ (°C). The average of 4 tests, which remained 1 h for each, was taken for the final plots (Speyer, 1994). The a.c. electrical conductivity "$\sigma$" was carried out on disc samples of ≈1 cm in diameter and ≈4 mm in thickness coated by silver paste using two – probe method with the applying of a complex impedance technique. Where, SR830 DSP Lock-in amplifier was used to measure the voltage difference $V_R$ between the two ends of known resistance $R$ which is connected in series with the sample in the frequency range ($10^2$–$10^5$ H$_Z$) and average recordings of 3–4 samples were used for the investigated plots (Ata-Allah, 2004). Bulk density, water absorption and total porosity were measured according to the ASTM Designation (D 570 – 81) and ASTM Designation (C 948-81)/re-approved 2001 (ASTM, 1981a,b).

### 2.2. Fast neutrons and gamma rays measurements

Fast neutron and total gamma ray spectra have been measured behind cylindrical samples of epoxy/ilmenite (EP/Ilm) composite with diameter Ø = 10 cm and ≈5 cm in thickness. Measurements have been carried out using a collimated beam emitted from the radioactive spontaneous fission ($^{252}$Cf 100 μg) neutron source and the neutron – gamma spectrometer with stilbene scintillator (4 × 4 cm) based on zero cross over method and pulse shape discrimination (P.S.D.) technique. The collimated beam was provided by a narrow beam experimental facility, which consisted of (radioactive source + collimator – samples holder – detector + collimator) as shown in Fig. 1.

The purpose of the beam and detector collimation is to provide a beam of specific intensity and geometry suitable for making measurements, as well as, to protect the detector against side scattered radiation and to reduce the incident thermal neutrons to enhance the discrimination capability. The pulse shape discrimination (P.S.D.) technique based on the zero-cross over method was used measure pulse amplitude distribution, as well as, to reject undesired pulses resulting from recoil protons and electrons due to neutrons and gamma rays respectively. The spectrometers set up, pulse amplitude distribution measurements and discrimination, in detail, and were given elsewhere in (Miller, 1968; McBeth et al., 1971).

Measured pulse amplitude distributions of recoil protons or electrons were converted into energy distribution of fast neutrons and gamma rays using the unfolding codes NSPEC and GSPEC based on double differentiation and matrix correction methods respectively (Toms, 1971; Kolevatov et al., 1969).

Spectrometer discrimination, linearity and energy scaling were checked before and through measurements by accumulating spectra of gamma rays emitted from $^{22}$Na, $^{137}$Cs, $^{60}$Co and Pu-α-Be neutron source. Checks indicated that the discrimination capability and pulse pile up rejection for recoil protons were good over the range of energy from 1.5 to 10 MeV. A block diagram of the spectrometer components was shown elsewhere in (Bashter et al., 1996).

### 2.3. Slow neutron measurements

The $BF_3$ (LND – 20354) and thermal neutron detection system were used to measure slow neutrons behind the concerned epoxy/ilmenite (EP/Ilm) composite with the same mentioned dimensions. The same experimental layout was used where; $BF_3$ tube was introduced into the detector collimator with an operating voltage of 1600 V. Its output was fed to the preamplifier, then, to the amplifier type ORTEC 572 A. The amplified pulses were fed to the PC incorporating a TRUMP 8 K/2 K data acquisition card. The slow neutron (0–1000 eV) flux was measured by integrating the net area under the peaks 2.31 and 2.7 MeV (Knoll, 1989). The experimental lay out and detection system diagram is found in Fig. 1 and elsewhere (El-Sayed Abdo et al., 2003b).

### 2.4. Theoretical calculations

#### 2.4.1. MCNP-4C2 technique

A typical and exact three dimensional model was designed using MCNP-4C2 computer code for the experiment geometry. In the model, built in $^{252}$Cf neutron source with spontaneous fission was used for the source term and variance reduction method DXTRAN sphere was used at the detector term to improve and increase the results accuracy. The program is designed to conduct the calculations using analytical methods starting from the radioactive source up to the DXTRAN outer sphere, hence, it starts using the MCNP technique reaching the DXTRAN inner sphere. The program is operated in the neutron – photon mode in order to accumulate the fast, slow neutrons and neutron induced gamma ray tallies. Then it was operated in the photon mode to obtain the gamma ray tallies which were added to the first run second term in order to get the total transmitted gamma ray component. A number of $10^8$ neutron and $10^9$ photon histories were performed to simulate the measurements and the tallies were scaled to the source strength using tally multiplying cards. The code was used to evaluate the attenuation parameters theoretically from the relevant attenuation relations by integrating the area under theoretical spectra for different thicknesses of the concerned composite (Briesmeister, 2000).

#### 2.4.2. MERCSF-N program

The MERCSF-N computer program had been constructed, verified and applied for calculating the macroscopic effective removal