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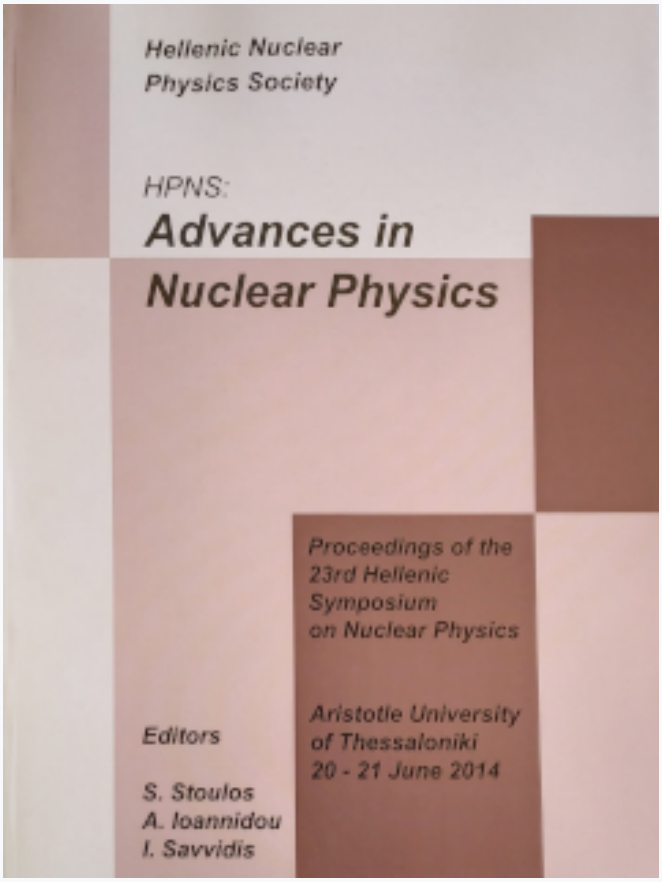

Large Sample Neutron Activation Analysis of Ceramic Matrix Composites

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Large Sample Neutron Activation Analysis of Ceramic Matrix Composites

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Abstract

Carbon fiber reinforced SiC ceramic matrix composites (C_f/SiC) are promising structural materials for a variety of high-temperature aerospace and energy applications. Since the matrix elements are carbon and silicon, they present low activation after neutron irradiation and therefore are materials of particular interest for fusion energy technology applications. Large Sample Neutron Activation Analysis (LSNAA) was applied to determine the elemental composition of C_f/SiC specimens joined together using a high temperature graphite based adhesive. The neutron irradiations and gamma ray measurements were performed at the BISNIS facility of the Hoger Onderwijs Reactor, TU Delft. The results of this study demonstrate the feasibility of application of LSNAA as a cost-effective method for non-destructive elemental composition analysis of whole ceramic specimens enabling the calculation of induced activity and dose rate after irradiation in different neutron spectra.

Keywords: Neutron Activation Analysis, Large Sample, Ceramic Matrix Composites

Introduction

Neutron activation of structural materials used for future fusion plants may result in radiation doses to personnel, which in the medium-term could complicate the handling of components in maintenance operations. Furthermore, it may produce long-term activation products, which could require special treatment, storage or disposal as waste at end of life of the station. Therefore, advanced prediction of the induced activity and the resulting radiation dose rate levels after plant shut-down is an essential precondition for the choice of materials for fusion. The calculation of activity and dose rate levels is based on detailed evaluation of the activating neutron fluence and knowledge of the actual elemental composition of the materials [1]. Carbon fiber reinforced SiC (C_f/SiC) ceramic matrix composites (CMC) are promising structural materials for a variety of high-temperature aerospace and energy applications [2]. As the manufacturing processes of CMC materials have reached a high maturity level, their limited use arises from the fact that they are not effectively integrated to compound structures. Thus, joining of CMC materials themselves or with other materials consistently, with reproducible microstructure, strength and in-service integrity, is of paramount importance for the envisaged applications.

Since the matrix elements are Carbon and Silicon, C_f/SiC present low activation after neutron irradiation and therefore are materials of particular interest for fusion technology applications [3]. In this work, Large Sample Neutron Activation Analysis (LSNAA) [4] was used to determine the elemental composition in C_f/SiC samples joined together using high temperature graphite based adhesive. The analysis assisted in the evaluation of induced activity and dose rate in the specimens as a function of time after the end of irradiation in different neutron spectra.

Experimental

The Cf/SiC (SICARBON^{TM}) ceramic composites were supplied by Airbus Group Innovation [5]. The bonding of the individual CMC specimens was carried out using a commercial high temperature graphite based adhesive from Cotronics with a service temperature of 2980 °C. Before the bonding, perforation of the specimens was performed in order to enhance the shear strength of the joints, as it has been proved in a study of CMC to Titanium alloy joining [6]. Table 1 shows the physical characteristics of the samples. Each sample was composed of two CMC specimens of 50 x 50 x 2.5 mm^3. The X-ray images of the samples are shown in Fig. 1.

Table 1 Sample Description

Sample	Dimensions (mm^3)	Mass (g)
A	50 x 50 x 5	24.377
B	50 x 50 x 5	22.848

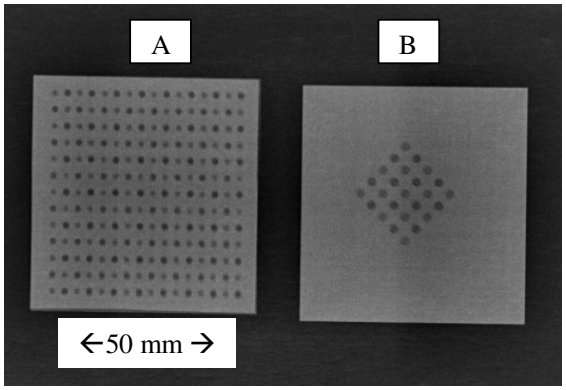

Fig. 1 X-ray image of C_f/SiC samples analyzed in this work

The LSNAA experimental procedure was performed at the facilities of Reactor Institute Delft (The Netherlands). Neutron irradiation was performed at the Big Sample Neutron Irradiation System (BISNIS) installed at Hoger Onderwijs Reactor graphite thermal neutron column [7]. BISNIS provides a moderated neutron flux of 5×10^8 $cm^{-2}\cdot s^{-1}$. The samples were irradiated for 26 h. Zinc flux monitors were positioned on the surface of the samples and at predetermined positions within the graphite moderator. Measurements were performed using a germanium detector based gamma spectrometry system [8]. The detector consisted of a high-purity germanium crystal of 96% relative efficiency, FWHM of 1.82 keV at the 1332.5 keV peak and peak to Compton ratio of 97:1. Spectrum analysis was performed using Gamma Vision™ software.

Simulations

Monte Carlo code MCNP-5 [9] was used to simulate the irradiation and gamma detection systems using cross section data from the Evaluated Nuclear Data File system [10]. Neutron and photon self-shielding correction factors for the samples were calculated. The germanium detector Full Energy Peak Efficiency (FEPE) for the volume source geometry configuration was calculated using the efficiency transfer method on the basis of the FEPEs calculated for the sample and measured for a reference point source [11]. The radionuclide inventory and gamma dose rate was predicted using FISPACT-2007 code and the European Activation File [12].

Results and Discussion

Elemental analysis

The measured isotopes, their half-lives, photon peak energies used in analysis, MCNP calculated gamma correction factors, $f\gamma$, and the elemental concentration results for the two samples studied are shown in Table 2.

Induced activity and dose rate predictions

The FISPACT predicted activity and contact dose rate as a function of time post-irradiation for 3 MeV and 14 MeV neutrons irradiation, are shown in Figs 2a and 2b, respectively. Calculations were performed for a 2.5 year irradiation period at a neutron fluence rate of 1×10^{15} $cm^{-2}\cdot s^{-1}$. It is noted that these results are indicative since SiC composition is taken from reference [13]. For the cooling time period studied, ^3H was the dominant radionuclide as far as specific activity is concerned for both 3 MeV and 14 MeV neutrons. For 3 MeV neutrons, the dominant nuclides contributing in gamma dose were ^{54}Mn and ^{108m}Ag, at 1 y and 100 y, respectively. On the other hand, for 14 MeV neutrons the dominant nuclides for gamma dose were ^{22}Na and ^{26}Al, at 1 y and 100 y, respectively.

Table 2. Isotopes, half-lives, photon energies, gamma correction factors and analysis results for the two samples

Isotope	Half-life	Peak (keV)	fγ	Sample A Concentration (mg/kg)	Sample B Concentration (mg/kg)
Cr-51	27.70 d	320.1	0.6801 ± 0.0002	10323 ± 1713	8267 ± 1304
Au-198	2.70 d	411.8	0.6831 ± 0.0002	1.87 ± 0.73	96.5 ± 5.1
W-187	23.90 h	479.5	0.6855 ± 0.0002	1333 ± 94	1749 ± 131
Br-82	35.30 h	554.3	0.6882 ± 0.0003	6529 ± 796	4660 ± 607
Na-24	15.02 h	1368.5	0.7099 ± 0.0005	27079 ± 1457	29229 ± 1585
La-140	40.28 h	1596.5	0.7136 ± 0.0005	175 ± 12	193 ± 13

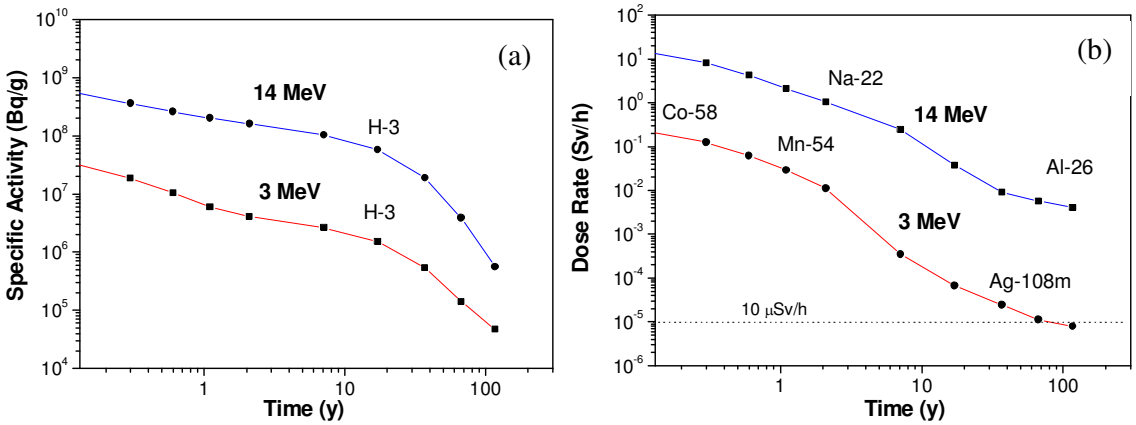

Fig. 2 Specific activity (a) and contact gamma dose rate (b) as a function of time after irradiation for 3 MeV and 14 MeV neutron irradiation for typical samples

Conclusions

LSNAA technique was applied to determine the elemental composition of joined ceramic matrix composite specimens. Moreover, the induced activity and gamma dose rate as a function of time after sample irradiation in different neutron spectra were evaluated. The results of the present study provide important information and therefore may assist the radiation protection of personnel in maintenance and radioactive waste management operations, which involve handling of activated structural materials, in future fusion plants.

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