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Porosity effects on the neutron total cross section of graphite

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ABSTRACT

For subthermal neutron energies, polycrystalline graphite shows a larger total cross section than predicted by existing theoretical models. In order to investigate the origin of this discrepancy we measured the total cross section of graphite samples of three different origins, in the energy range from 0.001 to 10 eV. Different experimental arrangements and sample treatments were explored, to identify the effect of various experimental parameters on the total cross section measurement. The experiments showed that the increase in total cross section is due to neutrons scattered around the forward direction. We associate these small angle scattered neutrons (SANS) to the porous structure of graphite, and formulate a very simple model to compute its contribution to the total cross section of the material. This results in an analytic expression that explicitly depends on the density and mean size of the pores, which can be easily incorporated in nuclear library codes.

1. Introduction

Graphite has been used in nuclear reactors since the birth of the nuclear industry due to its good performance as a neutron moderator material. Graphite is still an option as moderator for generation IV reactors due to its good mechanical and thermal properties at high operation temperatures [1]. So, there has been renewed interest in a revision of the computer libraries used to describe the neutron cross section of graphite [2–5].

A typical feature in the measured total cross sections of polycrystalline graphite found in the literature are the discrepancies between experimental total cross section in the low-energy region and the theoretical calculations commonly employed, as shown in Fig. 1. Such differences have been reported on nuclear-grade graphites of different origins by several groups [6–8,5]. Whilst the difference between the first Bragg edge (at 0.0018 eV) and 0.03 eV can be ascribed to the crystallographic texture of the material, there is yet no satisfactory explanation for the cross section measured at energies below the first Bragg edge. So, whilst the combined effect of nuclear absorption, incoherent and inelastic scattering predicts a value of nearly 0.2 b, the experimental values are typically over 4 b. The only exception is highly oriented pyrolityc graphite, which is polycrystalline but possesses near theoretical density and degree of orientation close to a single crystal, which shows a total cross section similar to the theoretical value [8–10]. Six decades after the first reports, there is still some controversy about the origin of the measured total cross section of graphite for low-energy neutrons.

In an early paper, Egelstaff [7] ascribed the origin of such an increase in the total cross section to small angle scattering of the neutrons due to refraction of the neutron beam as it passes through the many air/solid interfaces in the porous graphite. This explanation agreed with the experimental observation that an initially collimated neutron beam increases its divergence after passing through a graphite block [11]. In that work, Egelstaff briefly described two experimental arrangements aimed to subtract this small angle contribution, as he was mainly interested in the inelastic cross section and in the elastic cross section due to reflection on the crystallographic planes. By the same time, Antal et al. also recognized this 'porosity' scattering and performed similar corrective tests on experiments dedicated to study lattice defects in graphite, which also manifest as an increase in the total cross section for low-energy neutrons [12]. In a recent work, Bowman et al. [5] discarded the 'small angle' explanation and strongly criticized Egelstaff's experiments, reinterpreting Egelstaff account of the results, but without repeating the actual experiments.

In this work we study the origin of this discrepancy, by measuring the total cross section of room temperature graphite in the energy range from 0.001 to 10 eV, and compare it with standard calculation tools. To do so, we performed experiments on graphite samples from three different origins, studying the dependence of the total cross section with the geometry, orientation and dimensions of the actual sample. Besides this, we measured the total cross section of samples 'as received' and after an annealing treatment, to assess possible contributions due to scattering in lattice defects as discussed by Antal et al. [12]. Finally, in order to identify

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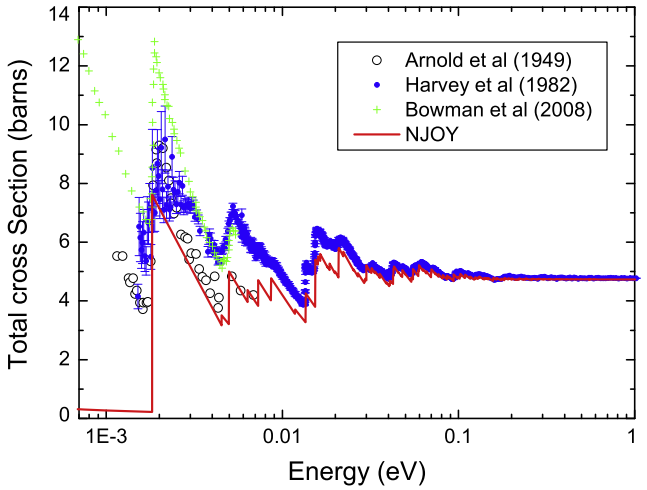

Fig. 1. Comparison of measured graphite total cross sections of different authors (cited in text [6,8,5]) with calculation performed with the NJOY code.

the contribution from small angle neutron scattering (SANS) we repeated Egelstaff work, by performing transmission experiments using two geometric configurations. The outcome of the experiments convinced us that the increase in the total cross section at subthermal neutron energies results indeed from small angle scattering in the porous graphite.

As mentioned, existing computer libraries of nuclear data provide evaluated values which differ from the actual measurements for polycrystalline graphite in the subthermal range. In particular, the very popular NJOY calculation code [13] includes a model for graphite with a detail of its crystal structure (elastic coherent term) as well as the vibrational modes of the lattice (inelastic term) [14]. However the code does not include any description of the 'mesoscopic' structure of the material such as porosity or voids, mainly because no theoretical model for the SANS total cross section has been developed so far. So, in the last section of this work we provide such a model of the total cross section, by assuming a very simple description about the distribution of the pores. We consider spherical air pores uniformly distributed in graphite, which results in an analytic expression for the total cross section that explicitly depends on the density and mean size of the pores. Finally, we show that the experimental total cross section of polycrystalline graphite can be effectively described by simply adding an analytic SANS component to the usual theoretical approach implemented in NJOY.

2. Experimental

2.1. Samples

Virtually all nuclear graphites are manufactured from petroleum or pitch cokes, binded and impregnated with different materials. By variation of these ingredients and the forming and final heat treatment, it is possible to produce a wide range of physical properties and behaviors under irradiation. In this work, samples of various geometries and sizes were produced from nuclear-grade graphites of three different types, namely 'needle-coke graphite', 'pitch coke graphite' and 'isostatically pressed graphite'.

2.1.1. Needle coke graphite

Needle coke graphite is widely used in the manufacture of electrodes in the metallurgical industry and corresponds to the most porous material. Because of its availability and purity it was used in the manufacture of the first moderator materials during the development of nuclear fission reactors [15]. This material has a large anisotropy, which caused some problems after long exposures to radiation. Samples were cut from a 24 cm cube provided by the RA-6 Nuclear Reactor, Centro Atómico Bariloche, Argentina. The density of the material we used is (1.681 ± 0.008) g/cm^3. An EXAFS analysis provided a composition (C 99.815%, S 0.102%, Ca 0.047%, Si 0.026%, Al 0.009% and K 0.001%). Pores of around 1 mm were visible to the naked eye. A micrograph of the surface of these materials is shown in Fig. 2.

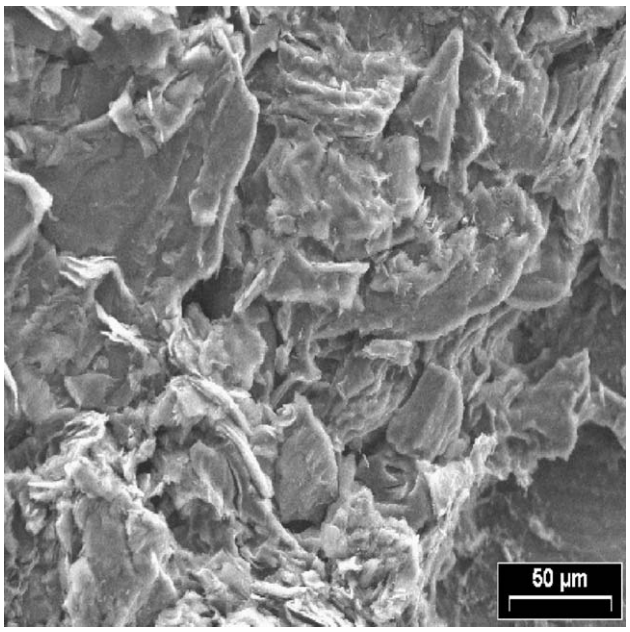

Fig. 2. Micrograph of the surface of the needle-coke nuclear-grade graphite cube studied in this work.

2.1.2. Pitch coke graphite

Pitch coke graphite is produced by heat treatment of the higher molecular weight products of pitch production, leading to more isometric particles than needle-coke graphite. The material used is more dense (1.725 ± 004) g/cm^3 than the needle-coke graphite. The sample presents a smoother surface than pitch coke graphite, but the pores are still visible to the naked eye. Spherical samples were machined out from a rod.

2.1.3. Isostatic-pressed graphite

Isostatic-pressed graphite uses finer coke particles and allows the manufacture of a wide range of isotropic graphites having very uniform and isotropic properties, which may be used as moderator materials. Spherical samples were moulded by isostatic pressing. These samples presented the largest density (1.740 ± 004) g/cm^3.

Table 1 describes the samples used on these experiments. The density of each sample was determined from its weight and volume. The porosity was calculated by comparison to the theoretical density for graphite $\rho_C = 2.266$ g/cm^3 and the measured densities ρ_a. The total volume of the pores is

$$V_P = M\left(\frac{1}{\rho_a} - \frac{1}{\rho_C}\right), \tag{1}$$

and the porosity

$$P = V_P/V_T, \tag{2}$$

where V_T is the total sample volume.

The values of the calculated porosities (shown in Table 1) are in the range of variation observed on graphites (between 7% and 30% [16]).