Material Selection for the SPIRAL2 Neutron Converter

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INTRODUCTION

The SPIRAL2 project aims at delivering high intensities of rare isotopes beams by adopting the suitable method for each desired radioactive beam. The RIB's will be produced by the ISOL "Isotope Separation On-Line" method via a converter. The neutron converter has to produce an intense flux of fast neutrons, mainly in the forward direction respect to the incoming primary beam, enable to induce up to 10^{14} fission per second in the Uranium Carbide target located upstream the converter. The primary beam is constituted by deuterons of energy 40 MeV and current up to 5 mA (up to 200 kW). The neutron converter is conceived as a high speed rotating target, which limits the peak surface temperature of converter materials well below 2000 °C. Nuclear graphite made of natural carbon is a very suitable material as neutron converter. In fact, ^{nat}C(d,n) reaction is very prolific, especially in the forward direction where the neutron yield is comparable to that generated by other light material converters. The MPG-type and CGD-graphite types are analysed.

GRAPHITE SELECTION

Many materials can produce intense fluxes of fast neutron when bombarded with low energy deuteron beams. Among them only a few can withstand high power beam and work at temperatures around 2000 °C. Among several tested materials the graphite-brand results the most suitable one to produce intense neutron flux under rather severe operation conditions. Several types of commercial available graphite types have been investigated and experimentally tested. Two of them, the MPG fine-disperse carbon composites (NIIGRAFIT-Moscow-Russia Federation) and the CGD-brand graphite (HENSHKE-Germany), were selected. Both graphite types present very similar physical and mechanical characteristics, as shows in tables 1 and 2.

TABLE 1. *Lattice parameters and micro-structural characteristics of selected graphites.*

Sample	Lattice parameters		SCD, Å		Micro-distortion size ε *00l*
	a, Å	*c*, Å	*00l*	*hk0*	
MPG	2.464(1)	6.766(3)>	1500	250	0.0065
CGD	2.465(1)	6.764(4)	300	250	0.0030

TABLE 2. *Main physical and mechanical parameters of selected graphites (room temperature).*

Parameters	**unit**	**CGD**	**MPG**
Density	g/cm^3	1,80-1,84	1,72-1,87
Specific resistance	μΩm	9,4 -10,2	9,5 - 12
Bending ultimate stress	MPa	35	35 - 52
Compressive ultimate stress	MPa	55	65 - 119
Porosity	%	15	
Ashes content	%	< 0,01	0,1-0,25
Grain size	μm	<80	30-150
Young modulus	GPa	N/A	10-12
Heat conductivity	W/m*K	N/A	110-120

For reasons related to the commercial availability the CGD-brand graphite has been chosen as material for the neutron converter. To study the main physical and mechanical properties, and its serviceability under operating conditions, several samples of CGD graphite have been tested. The operation temperature and the heating time are the main parameters affecting the converter resources. Each graphite sample is properly shaped and clamped in the special holder where is heated by alternate electrical current passing through the sample. The active part of the sample has dimensions 60 x 5 x 1 mm^3. The heating of the samples are performed in a vacuum environment of the order of 10^{-2} mbar. Before and after long term heating (or sample breaking) the samples are analyzed by X-ray Diffraction (XRD), Transmission Electron Microscopy (TEM) and Accelerator Mass Spectroscopy (AMS) methods. Composite electro-physical and structural changes are investigated during the test as a parameter of material degradation in function of time.

MATERIAL ANALYSIS

The X-ray diffraction measurements of the CGD graphite samples were performed both for the plate and crumbled graphite. The measurements shown, figure 1, the presence of all reflexes typical for 2H graphite poly-type; however, their broadening rate is quite different. Narrow reflexes *00l* and *hk0* types indicate a rather large size of the Coherent Scattered Domain (CSD) both in the normal direction to the graphene layer and in the basis plane. At the same time, the essential broadening of *hkl* type peaks indicates the presence of large number of stacking faults (distortions of the stacking sequence).

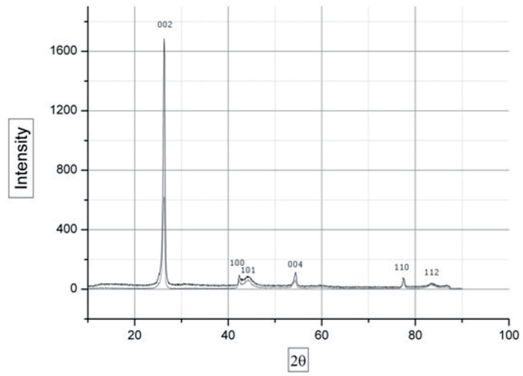

FIG. 1. *X-ray diffraction (XRD) measurements of the CGD graphite samples. The peak [002] is more intense for the plate because take place in the structure direction - [00l].*

The parameters and micro-structural characteristics of the samples are shown in Table 2.2. The uncertainty in the lattice parameters, which reaches 10% of CSD size, is enclosed in brackets. The lattice strain magnitude is known to characterize variations in interplanar spacing (interlayer spacing d_{002} in this study).

The electronic microscope photography of carbon samples were performed using a Transmission Electronic Microscope (TEM) with 200 kV accelerating voltage and 0.2 nm resolution. The samples to be investigated were prepared from alcohol suspension; a drop of it was placed onto the carbon substrate and the substrate set on a fixed copper grids. The observed defects in graphite could be divided into two types: the defects related to derangements between layers, and bond defects in grids. The first group is formed by layers packing defects which are attributed to packing derangements of parallel layers. The second group comprises the defects in carbon lattice bonds.

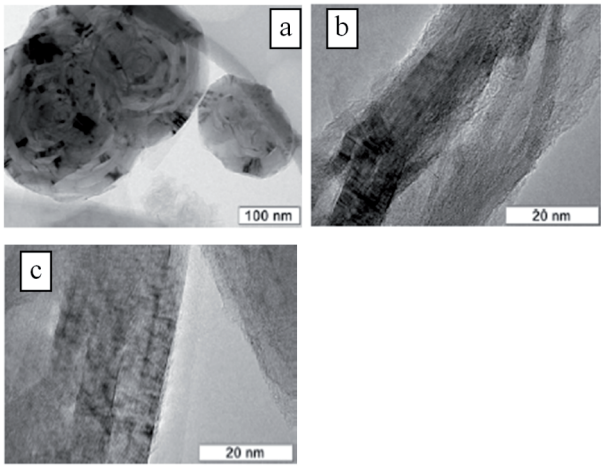

FIG. 2. *High resolution transmission electronic microscopy of CGD graphite samples. Interstitial impurity defects (a), split-defects (b) and twinning defects (c) are well visible.*

These are vacancies and their groups, impurity atoms implanted in hexagonal layer, isomeric bond defects, atoms *sp3* hybridization and boundary defects (see figure. 2).

In the pictures is possible to observe so-called Super-Molecular Structure (SMS). It consists on sites where the carbon layers have a parallel orientation. The site of un-basic twinning is well shown as inclined inter-crystalline border with an angle close to 48°. These edges are characterized by numerous breaks of connections and advanced system regional dislocation.

Besides, there are chemical defects (alien atoms implanted to a carbon grid) and defects related to atom displacements from their normal position in a lattice. Accelerator Mass Spectroscopy (AMS) measurements show the presence of these impurity particles. In particular, the potassium, sulfur and oxygen impurities are clearly seen in figure 3. Notice that copper is not a typical impurity of graphite; the presence of a copper peak is related to the holder.

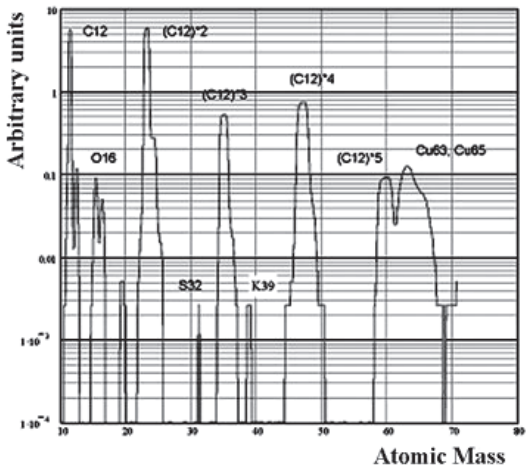

FIG. 3. *Impurity on CGD graphite detected by the Accelerator Mass Spectroscopy method. The presence of copper is related to the holder used to fix the sample.*

Electro-physical measurements for CGD graphite, in particular, measurements of conductivity, magneto-resistance and Hall's effect temperature dependence have shown full agreement of experimental results with those obtained for MPG graphite. It is necessary to note the absence of small region of negative magneto-resistance typical for MPG graphite, which is related to packing defects in graphite, as well as predominantly p-type conductivity.

CONCLUSIONS

Microscopic properties of MPG-type and CGD-type graphite were analyzed by XDR, TEM and AMS methods. The experimental results shows that both graphite types presents analog characteristics and are well suitable as material for the neutron converter.