Characterization Studies of Cyclotron CS-30 Carbon Puller Material Using Powder X-Ray Diffraction and SEM, EDX Cross Section Method

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Abstract. The carbon puller material of Cyclotron CS-30 has been characterized to investigate the structure and composition of the puller. Two samples of pullers have been prepared in the form of powder from locally made carbon and the original cyclotron puller. X-ray diffraction (XRD) analysis of both samples powder gives identification of the structure and phase type of carbon, meanwhile the morphology and the elemental composition of both samples can be shown by Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Analysis (EDX) technique. XRD samples are prepared by grinding the puller surface meanwhile the samples for SEM analysis are prepared by cross section method. XRD result shows amorphous carbon structure and a typical broad XRD profile’s peaks and the diffused nature of the XRD profile indicate the presence of disorder in the samples. The first peak of the profile corresponds to the (002) peak of the hexagonal graphite structure, while the second peak corresponds to the (100) and (101) for both samples. Results of SEM-EDX showed that the carbon material from the original default puller of the CS-30 cyclotron contains mass percentage of lead significantly by 9.15%.

1. Introduction

Many of the important attributes for an effective process in cyclotron are very early in the acceleration process-in the central region. The performance of the cyclotron central region is essentially determined by first few turns of the accelerated ion beams[1]. For this reason, the central region has accordingly been the subject of a great deal for intensive study over a long period at our cyclotron CS-30 facility in national nuclear energy agency of Indonesia (Batan). Unfortunately, an in depth study and information correlating the structural characteristics of the material using in the central region is unavailable. Understanding the relationship between material properties, chemistry and atomic scale structural arrangement is critical for discovery and optimization of new functional materials[2]. In the present study, we report the characteristic material for one of the component in the central region, using one from the original manufacturer and one that have been locally made as a sample.

The sample component being used was puller, made of carbon. Since more than a quarter century, progresses in the development of carbon-based materials have been reported due to the importance of carbon in modern science, in high technology and also in a large domain of human activities. It is
chemically inert and also resistant to corrosion and shock[3]. Carbon as a material possesses an excellent properties e.g. it can withstand very high temperature up to 3000°C, it exhibits low friction, low thermal expansion, good electrical conductivity and high mechanical strength[4].

Irradiation induced displacement of carbon atoms cause excess energy associated with vacancy/interstitial pairs to be stored up in the graphite crystallites[5]. The stored energy is released when the carbon atoms and the vacancies recombine and the release curves are characterized by a peak that becomes broader with increasing radiation dose whereas the maximum release rate of energy is reduced. So, the necessary and binding conditions are that the material carbon puller should be highly dense, amorphous and isotropic to achieve dimensional stability under irradiation.

The principal techniques that widely used for characterization of the material such as analysis of the microstructure and composition of a material are powder x-ray diffraction (XRD), scanning electron microscopy (SEM), and energy dispersive x-ray (EDX) spectroscopy. X-ray diffraction has been in use in two main areas, for the fingerprint characterization and the determination of the structure. Each solid material has its unique characteristic X-ray powder pattern which may be used as a "fingerprint" for its identification. Once the material has been identified, X-ray crystallography may be used to determine its structure, i.e. how the atoms pack together and what the interatomic distance and angle are, etc. X-ray diffraction is one of the most important characterization tools used in solid state chemistry and materials science[6]. Meanwhile, the EDX technique has been widely used to identify metals and/or contaminants while SEM is used to investigate the morphology and using the SEM technique, it is possible to observe the surface defects of the material[7,8]. Cross sectional method for powder sample in SEM measurement is rarely used by scientist, but in this paper we describe how to prepared powder sample with this method. The method contain metallurgical technique and can obtain to view the grains and particles clearly. The research findings would provide an important test basis for further studying of puller material under irradiation.

2. Experimental method

2.1. Chemicals and instrumentation

The carbon material used for the analysis is derived from the component called puller from the central region part of National Nuclear Energy Agency of Indonesia/Batan’s cyclotron CS-30. Two sample are locally made carbon puller and original puller from the manufacturer with specific surface area 1.124cm$^2$/g and 1.060cm$^2$/g respectively. Local carbon puller made previously by the researcher in the cyclotron department using local graphite rod turn into the same shape of the original puller.

The structure of the sample was analyzed with X-ray powder diffraction (XRD Philip PW 1710 16) using Cu K$_\alpha$ radiation at room temperature. Scanning Electron Microscopy (SEM); Electron Dispersive X-ray (EDX) measurement. Experiments and analyses involving electron microscopy were performed in the Science and Technology Center for Advanced Materials of National Nuclear Energy Agency (BATAN), Indonesia. Secondary electron and backscattering electron images with qualitative and semi-quantitative chemical analyses in the EDX mode were obtained using a JEOL Type JSM - 6510 LA equipment.

2.2. Procedure reaction

Material for powder x-ray diffraction is finely ground to less than ~10 μm (or 200-mesh) in size. The resulted pattern was then matched using MATCH program to search the pattern structure approach.

SEM-EDX sample was prepared by cross sectional method to see through the cross sectional in each grain in the sample. The detailed sample preparation process of carbon for SEM;EDX measurement is as following :

1. Mount the powder by suspending the powder in resin+hardener media to consolidate the sectioning.
2. To expose a fresh surface the next step is grinding with the abrasive paper of 400, 600, 800, 1200 and 1500 in sequence.
3. Polishing by alumina paste will removes the damages imparted of grinding operation and
followed by adding the sample into the water in ultrasonic cleaner for half an hour.

(4) Finally, sputtering technique (GL-1110X SPC-12-Compact Plasma Sputtering Coater) was performed to coat the sample with Au.

3. Result and discussion

Figure 1 shows the pattern of the xrd measurement for carbon puller which was locally made and the original carbon puller from manufacturer. As expected, the carbon material pattern shows some sharp diffraction peaks and also a broad features characteristics of an amorphous material. The diffused halos appearing at 2θ values i.e., 25 to 55 degrees are indicative of the amorphous nature of the puller material having some degree of arrangement (crystallinity) in the molecular chain of both samples. Both pattern fit well each other despite the fact that the local carbon has higher intensity and broader peak. The triangular peaks observed in the diffraction patterns result point to the presence of crystallinity.

![XRD Pattern of the carbon puller material](image)

Figure 1. XRD Pattern of the carbon puller material

Figure 2 and 3 Show the identification of the peak correspond to the hexagonal structure of graphite in (002), (100) and (110) reflection using Match program. The diffraction profiles show the presence of a asymmetric (002) band around 26°, which suggests some graphite-like structure (crystalline carbon and a weak band at 43°. It is suggested that carbon has an intermediate turbostratic or random layer lattice structure between graphite and amorphous[9]. This means that the carbon puller, amorphous carbon, contains significant amount of highly disordered material, which is responsible for the back ground intensity of the diffraction. Position of this peak is found to shift to higher 2θ value with increase in elemental content of carbon, this can be caused by the mechanical grinding of the sample without any heat treatment.

Refinement on the pattern data was not performed because of the lack of crystallinity and therefore next calculation on the percentage of crystallinity and amorphous part needs to be done. One of the basic assumptions of meaningful XRD is the homogeneous spatial and angular, distribution of the crystallite orientations in the different directions. Any deviation from homogenity makes sampling of intensities difficult (the reproducibility of the measurement).

In general, the comprehensive characterization of amorphous phases necessitates the use of multiple analytical techniques. Pair Distribution Function (PDF) analysis of total elastic scattering (Bragg and diffuse) perhaps will be the most powerful method to access the Medium-Range Order (MRO) as seen in amorphous material. PDF can be used for exploring the effects of processing or
storage on amorphous materials and may be employed as a tool for comparing different processing methods and also to discern small structural differences[10].

Figure 2. XRD Pattern of locally made carbon puller matched using Match program

Figure 3. XRD Pattern of original carbon puller matched using Match program

Figure 4. SEM image of the original manufacturer carbon puller with EDX measurement respectively

The morphology and the EDX measurement are shown respective in the Figure 4 from the original manufacturer carbon puller. As a comparison in Figure 5, it also shown the microscopy characteristics of the locally made carbon puller. Three point have been made in each image of SEM for best contrast that shows the grain cross section. The light and the darker color in contrast show the difference in resin media, grain and cross sectional inside the grain. Visually the grain of the local carbon is bigger than the original and in Figure 6 for higher magnification of 2500x, the agglomeration of the particle occurs. Adding a very small amount of powder into the resin media help reduce the agglomeration.
The spectrum of energy dispersive x-ray from the original pulser show some amount of lead/Pb element which is not found in the locally made carbon puller. The detail of the composition is in Table.1. This element is suggested came from the irradiation in the cyclotron’s central region during the process or it can naturally exist in puller. The Irradiation changes the crystal properties and indirectly the structure factors that can to be related the macroscopic to the microscopic properties. This can cause the displacement of the atoms and increase the disorder (amorphous) in the sample which correlated to the diffraction pattern result.
Table 1. Elemental composition (%) correspond to the EDX measurement

| % Mass         | Point | C   | O   | Cl | Au  | Pb | Total |
|---------------|-------|-----|-----|----|-----|----|-------|
| Original      | 007   | 36.33 | 10.78 | 2.35 | 41.97 | 8.58 | 100.00 |
| Manufacturer  | 008   | 36.00 | 1.97 | 1.97 | 43.00 | 9.24 | 100.00 |
| Carbon Puller | 009   | 39.85 | 1.87 | 1.87 | 41.02 | 8.46 | 100.00 |
| Locally Made  | 004   | 63.84 | -    | 1.02 | 35.14 | -   | 100.00 |
| Carbon Puller | 005   | 63.55 | -    | 0.81 | 35.63 | -   | 100.00 |
|               | 006   | 64.76 | -    | 0.36 | 34.88 | -   | 100.00 |

4. Conclusions
The result of the structural investigation of the two sample of carbon puller showed that the diffraction pattern of such a material consists of two types of reflection. Crystalline type reflection which is graphite-like structure with diffraction peaks and two amorphous carbon which indicate the presence of disorder in the samples. SEM-EDX measurement of the original carbon material from Cyclotron CS-30 puller show 9.24% mass of lead.

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