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Effect of HF in the preparation of carbon-ceramic materials by sol-gel process

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Abstract – In this study, the influence on the morphology, porous structure, and mechanical properties of carbon-ceramic material with the use of HF in a two-step sol-gel process of tetraethylorthosilicate were evaluated. Micrographs obtained by Scanning Electron Microscopy (SEM) show the different morphologies of samples SGN, SGNF and SGF. SGNF is a mesoporous material with pores size of 110 Å, with good mechanical resistance and high dispersion of carbon graphite inside the matrix.

Carbon-ceramic materials, SiO_2/graphite (SG), prepared by sol-gel process has been used in the development of electrochemical sensors [1-2]. For these devices, application of these matrices should present mechanical rigidity, thermal and chemical stability provided by the silica framework allied with the graphite conductivity [2]. In this study, the influence on the morphology, porous structure and mechanical properties of carbon-ceramic material, with the use of HF in a two-step sol-gel process of TEOS, was evaluated. Three materials (SiO_2/graphite) were prepared using as catalyst: only HNO_3 (SGN), only HF (SGF) and also a mixture of both acids (SGNF) added at different steps of the synthesis. A graphite/SiO_2 ratio (w/w) of 1:1 was maintained for preparation of three samples. The preparation of the sample SGN were made in two-steps: the step of hydrolysis was achieved with the addition of an aqueous solution of HNO_3 (3.48 mol L^{-1}) to a solution of TEOS:ethanol (1:1 v/v) at reflux for a few hours (solution A). Then, the carbon graphite, a suitable amount of water was added to solution A, and subjected to ultrasonication until gelation of the material. For the sample SGF, water was added in the prehydrolysis step followed by HF as catalyst. Similarly, SGNF was prepared adding HNO_3 in the prehydrolysis step, followed by HF addition in the same quantities and conditions of preparation of SGN and SGF.

SEM images show different morphologies for the three materials studied (Fig. 1). Adsorption isotherms of nitrogen indicate that the SGN presents microporous structure and poor dispersion of carbon graphite in the amorphous silica matrix, as revealed by EDS mapping. Opposite behavior is observed with the use of HF (SGF) obtaining a material with larger pores and high carbon graphite dispersion in the amorphous matrix, but presenting lower mechanical resistance. By using a mixture of both catalysts (HNO_3 and HF) a mesoporous material of intermediate porosity (size pore = 110 Å), with good mechanical resistance and good dispersion of carbon graphite in the matrix of SiO_2 was obtained (Figure 2, and inset). Condensation kinetic of TEOS was considerably enhanced by using HF as catalyst, reducing the gelation time for SGF and SGNF. As Si is more susceptible to nucleophilic attack by the F^- ion, it reduces the electron density in the former atom, resulting in an increase of the condensation reactions of TEOS. With less time to gelation, a larger amount of carbon graphite could be entrapped in the silica matrix before deposition.

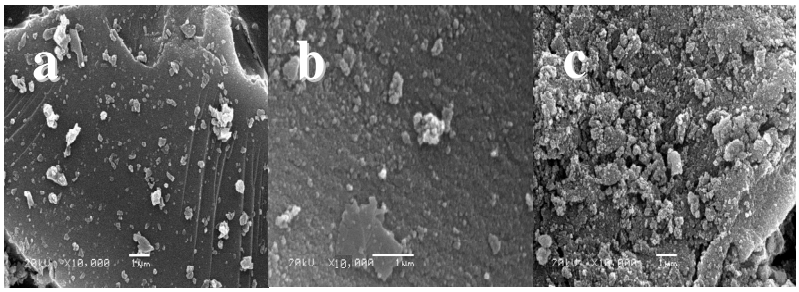

Figure 1: SEM images of the samples (a) SGN, (b) SGNF and (c) SGF.

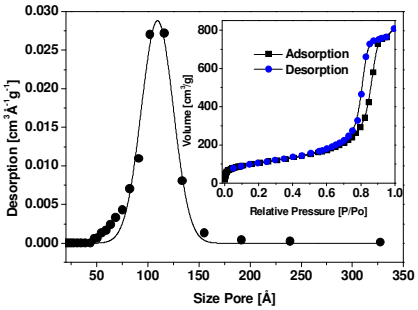

Figure 2: Curve of pore size distribution of the sample SGNF. Inset: Isotherm of adsorption-desorption of N_2.

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