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New material for low-dose brachytherapy seeds: Xe-doped amorphous carbon films with post-growth neutron activated ^{125}I

R.G.F. Gonçalves [a,*], M.V.B. Pinheiro [a], R.G. Lacerda [a], A.S. Ferlauto [a], L.O. Ladeira [a], K. Krambrock [a], A.S. Leal [b], G.A. Viana [c], F.C. Marques [c]

[a] Departamento de Física, Instituto de Ciências Exatas (ICEx), Universidade Federal de Minas Gerais (UFMG), Campus da Pampulha, Av. Antônio Carlos, 6627, 31270-901 Belo Horizonte, MG, Brazil
[b] Centro de Desenvolvimento da Tecnologia Nuclear (CDTN), Comissão Nacional de Energia Nuclear (CNEN), Campus da Pampulha, Av. Antônio Carlos, 6627, 31270-901 Belo Horizonte, MG, Brazil
[c] Universidade Estadual de Campinas (UNICAMP), Instituto de Física "Gleb Wataghin", 13083-970 Campinas, SP, Brazil

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ABSTRACT

We report a novel material for use in ^{125}I brachytherapy that consists of amorphous carbon films grown by ion-beam-assisted deposition and doped with Xe (5 at%) by implantation. Samples of these films grown on Si substrates were irradiated with neutrons in a TRIGA-I nuclear reactor for the production ^{125}Xe, and latter characterized by gamma spectroscopy. The results indicate that the ^{124}Xe was efficiently converted into ^{125}Xe, the precursor of ^{125}I, and support the activity calculations for a model brachytherapy seed.

1. Introduction

In the last decades, several new forms of pure carbonaceous materials have been synthesized. Apart from the well known diamond and graphite polymorphs, the growing interest on novel carbon structures with mixed sp^2 and sp^3 carbon hybridizations is unquestionable. This is mainly because of their vast range of potential applications from electronics to biomedicine. Some examples are: the pyrolytic and glassy carbon, carbon fibers, fullerenes and carbon nanotubes. Among them, one particular class of carbonaceous materials, which is very attractive for several applications in materials sciences, is the amorphous carbon (a-C) (Popov et al., 2006; Krainyukova and Belan, 2007; Neuville and Matthews, 2007; Mélinon et al., 2007).

Noble-gas (NG) clusters present intriguing properties and have been the focus of intense experimental and theoretical investigations. Their physical properties are extremely sensitive to size, temperature and pressure (Federmann et al., 1994; Kakar et al., 1997; Knop et al., 1998). On the other hand, the a-C has a unique combination of macroscopic properties, such as high mechanical and chemical stability, high porosity, electrical conduction and biocompatibility (Belonoshko et al., 2001). However, for most applications, it is essential to control certain microscopic properties like pore size, internal stress, and doping during synthesis. One common way to dope hard a-C films is by implanting NG like Ar, Ne and Xe which can be accommodated even at high concentrations (up to 5%) in the a-C lattice (Lacerda et al., 2003). Depending on the deposition parameters (temperature, pressure, deposition rates, etc.), the controllable high internal stress present in a-C films (up to 10 GPa) can even stabilize NG clusters (Lacerda et al., 2003). Since electronic structure of the a-C trapped NG, and more specifically their core energies, depend drastically on the external pressure, these films can be used as pressure sensors when monitored by, e.g., photoelectron spectroscopy (Cynn et al., 2001).

Another application of the Xe-doped a-C materials is in the field of medicine. Radioisotopes have been used in nuclear medicine for invasive radiotherapy against cancer, in a treatment known as brachytherapy (Woolsey et al., 2003). In this treatment, seeds containing the radioisotope are implanted in tissues nearby the tumor. The effective dose applied to the tumor and the energy deposition rate, given by the seed activity, as well as their geometry around the tumor, must be such that are sufficient to kill the cancerous cells and minimize the damage in the adjacent healthy tissues (Woolsey et al., 2003). One of the drawbacks of the brachytherapy is the fabrication of the seeds that requires radioisotope enrichment and manipulation of hot materials. Even though these processes have been evaluated to minimize the human contact (Abdukayumov et al., 2004; Robertson, 1999), they are still costly. Among several radioisotopes used in brachytherapy,

* Corresponding author. Tel.: +55 31 37715918.
E-mail address: rgfurst@yahoo.com.br (R.G.F. Gonçalves).

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the ^{125}I is one of the most common (Robertson, 1999; Crook et al., 2007), due to its low X-ray energies (27.2–31.9 keV), low gamma energy (35.5 keV) and half-life (59.4 days) (Crook et al., 2007). It is mainly used in prostate tumors (Robertson, 1999; Crook et al., 2007). The low-dose ^{125}I brachytherapy seeds have usually diameters of 0.5–0.8 mm and length of 2–5 mm, a volume of about 1.5–10.0 mm^3, and activities of about 0.3–0.8 mCi (Robertson, 1999; Crook et al., 2007; Solberg et al., 2002).

In this work we study the neutron-activation of ^{124}Xe into ^{125}Xe in a-C films, and its transmutation into ^{125}I. The purpose of this work was to investigate the possibility of using Xe-doped amorphous-carbon as a novel matrix material for alternative ^{125}I brachytherapy seeds. Several Xe-doped a-C thin films over Si substrates were irradiated with neutrons and investigated with gamma spectroscopy over a broad energy range. The specific activities of the radioisotopes produced in these samples were determined and compared with calculations. The concentration of ^{124}Xe and xenon gas were determined. Finally, we demonstrate that with a suitable neutron irradiation parameter, a 100% ^{124}Xe-enriched bulk Xe-doped a-C seed will yield ^{125}I nuclides, under neutron activation, with activities comparable to commercial seeds of about 37.0 MBq (1.0 mCi).

2. Materials and methods

Samples of thin amorphous carbon (a-C) films were prepared by ion-beam-assisted deposition (IBAD), at 150 °C, over Si wafers (Lacerda et al., 2003). Two ion sources were used. The first one was employed to sputter a graphite target (99.99 at%) with a xenon (99.999%) ion beam (energy of 1.5 keV and current of 90 mA). The second ion gun was used to simultaneously bombard the film during the growth process with a xenon ion beam (energy varying in the 0–400 eV range and current of about 90 mA). The base pressure was under 10^{-5} Pa, and total pressure during growth was kept at 6×10^{-2} Pa. The a-C film thickness is between 80 and 100 nm. X-ray photoemission spectroscopy (XPS), ultraviolet photoemission spectroscopy (UPS), electron energy-loss spectroscopy (EELS), transmission electron microscopy and Raman scattering indicate that the material is composed of a compressed and dense sp^2 network ($\sim$90% by EELS). By carefully controlling the deposition conditions, sp^2-rich amorphous carbon films with different internal pressures (from 1 GPa to about 12 GPa) were prepared. Detailed information on the structural properties of these films can be found elsewhere (Lacerda et al., 2003). Six samples of 100 nm thick a-C film, doped with 5 at% of Xe over a Si-substrate with an area of $\sim$1 cm^2 each were prepared for neutron irradiation. The a-C film density is (2.2 ± 0.2) g m^{-3} and the total masses of the samples (Si, C and Xe) was about 19–31 mg. The a-C mass of the films was calculated from its density, thickness and the total mass of the sample. Four samples were irradiated in a TRIGA MARK-I reactor of the CDTN/CNEN nuclear research center in Belo Horizonte, operating at 100 kW. The average thermal and epithermal neutron flux were 6.4×10^{11} and 4.4×10^{11} n cm^{-2} s^{-1}, respectively (CDTN, 2000). The irradiation times were 2, 4 and 8 h. After irradiation the samples were characterized with gamma spectroscopy using an HP Ge-detector with 15% nominal efficiency and full line width at half maximum (FWHM) of 1.85 keV for the 1332 keV peak of ^{60}Co. The energy calibration and efficiency curve for several distances from the detector were fitted using suitable point sources (CDTN, 2000). The irradiated samples were measured at 5 and 10 cm from the detector. Table 1 shows several data concerning the samples investigated: the Si substrate mass (M_{Si}); the a-C film mass ($M_{a\text{-}C}$); the irradiation time (t_i) and integrated flux (Φ); and, for the samples characterized with gamma spectroscopy, the distance from the detector (d), as well as counting time (t_c).

The Canberra Genie 2000 software (Zhu et al., 2009) was used for the data acquisition and spectral analysis. In this software, the gamma spectrum peaks are crossed with library data to identify the isotopes. The area of the peaks was used to identify and calculate the activity of each isotope and, consequently, the initial ^{124}Xe and xenon gas concentration of a-C matrix using Eq. (1) (Parry, 2003). We calculated the number of isotopes in the sample (N) from: the peak area (A); the gamma detector efficiency (ε); the isotopic abundance (θ); the neutron capture cross-section (σ); the decay constant of the activated nuclide (λ); the neutron flux (Φ); and the irradiation (t_i), decay (t_d) and counting times (t_c) according to

$$N=\frac{A}{Ke^{-\lambda t_d}(1-e^{-\lambda t_i})(1-e^{-\lambda t_c})},\ \text{where}\ K=\varepsilon\theta\sigma\Phi\lambda^{-1} \quad (1)$$

After that, we calculated the sample and irradiation parameters for obtaining a ^{125}I activity comparable to low-dose commercial brachhytherapic seeds. The results show the activity of the seed after irradiation times of 44 h and neutron flux of 10^{14} n cm^{-2} s^{-1}. In this simulation, the seed has a cylindrical Si nucleus with 0.8 mm diameter and 5 mm length and the a-C film 2 μm thickness. The a-C film was doped with 5 at% of xenon noble gas, with an ^{124}Xe-abundance of 100%, instead of its natural abundance of 0.1%, through the enrichment of xenon gas. For these calculations, Eq. (2) was used (Shultis and Faw, 2002) and the ^{125}Xe and ^{125}I activities (A_0^{Xe} and A_0^{I}, respectively) were taken into account. We also considered the decay times (t) and the decay constants for ^{125}I (λ_I) and ^{125}Xe (λ_{Xe}).

$$A_I(t)=A_0^{Xe}\frac{\lambda_I}{\lambda_{Xe}-\lambda_I}e^{-\lambda_{Xe}t}(1-e^{-(\lambda_I-\lambda_{Xe})t})+A_0^{I}e^{-\lambda_I t} \quad (2)$$

The results were compared with the ChainSolver software (Romanov, 2003), which uses a numerical method.

3. Results and discussion

The xenon, carbon and silicon natural isotopes data were taken from National Nuclear Data Center, Brookhaven National Laboratory (Pritychenco et al., 2006). Their natural abundance,

Table 1
Sample parameters: Si substrate mass (M_{Si}); the a-C film mass ($M_{a\text{-}C}$); the irradiation time (t_i) and integrated flux (Φ); and, for the samples characterized with gamma spectroscopy, the distance from the detector (d), as well as counting time (t_c).

Sample	M_{Si} ($\pm$1 mg)	$M_{a\text{-}C}$ ($\pm$0.2 ng)	t_i (h)	Φ (n cm^{-2})	d (cm)	t_c (min)
1	23	4.3	8	1.47×10^{15}	5	30
2	22	4.1	–	–	–	–
3	23	4.3	2	3.69×10^{14}	-	–
4	31	5.8	–	–	–	–
5	25	4.7	4	7.37×10^{14}	10	120
6	19	3.6	8	1.47×10^{15}	10	120