Direct Synthesis of Nanodiamonds by Ar-H_2-CH_4 Microwave Discharges

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A experimental study on microwave plasma based assembly of nanodiamonds at atmospheric pressure conditions is presented. The synthesis method is based on introducing a carbon containing precursor (methane), through a microwave (2.45 GHz) argon plasma environment, where decomposition of methane molecules takes place and carbon atoms and molecules are created and then converted into solid carbon nuclei in the post-discharge zone. The influence of additional hydrogen gas injected into the background gas mixture on the carbon species production and on the structural qualities of fabricated nanodiamonds has been investigated. Optical emission, Fourier transform infrared spectroscopy, Raman spectroscopy, scanning electron microscopy (SEM), and X-ray diffraction techniques (XRD) have been applied to study plasma emissions, the output gas stream composition, and the material and chemical analyses of synthesized nanostructures.

1. Introduction

Nanodiamonds have a 3D structure formed by sp^3 bounded carbon atoms arranged in a tetrahedral symmetry and their dimensions are in the range 2-5 nm. Nanodiamonds have a distinct combination of outstanding unique mechanical, chemical, biological, magneto-optical and electronic properties, which can be improved by adding functional groups. Moreover, they are nontoxic and can be used in biomedical applications, as drug carriers and delivery vehicles. Nanodiamonds are not destroyed by the human immune systems, and can be associated with a multitude of molecules and give targeted drug release. Usually extreme environment are necessary to synthesize nanodiamonds. Nanodiamonds can be found with extremely low concentrations on Earth in crude oil and in certain sediment layers, or in space, in meteorites, interstellar dust or in protoplanetary nebulae. The frequently used method is detonation of high explosive materials.

2. Experimental Setup

Nanodiamonds have been produced by a surface wave induced microwave plasma, using a waveguide surfatron-based setup. The microwave power is provided by a 2.45 GHz generator (Sairem), with maximum power of 2 kW. The generator is connected to a waveguide (WR-340) system, which includes an isolator, directional couplers, a three-stub tuner, a moveable short-circuit and a waveguide surfatron as the field applicator. The discharge ignites in a quartz tube, placed vertically and perpendicularly to the waveguide wider wall. A methane (CH_4) and hydrogen (H_2) gas mixture is introduced into the "hot" microwave argon plasma environment, where decomposition of methane molecules takes place and carbon atoms and molecules are created. Afterwards, carbon atoms and molecules are converted into solid carbon nuclei in the "colder" nucleation zones where nanometer-size particles are generated. The addition of H_2 is important since the surface of sp^3 clusters must be either stabilized through termination with functional groups or reconstructed into sp^2 carbon.

3. Results and Conclusions

XRD, Raman and SEM results showed that atomic hydrogen can kinetically etch the non-diamond sp^2-C and allow diamond-phase sp^3-C to grow. The stability of the nanodiamonds may also depend on the surface terminations, i.e. hydrogen bonds on the nanodiamonds boundaries, therefore, this method can be tuned from non-diamond to diamond phase growth, by controlling the C:H ratio in the gas mixture.

Acknowledgements

This work was funded by Portuguese FCT—Fundação para a Ciência e a Tecnologia, under Project UID/FIS/50010/2013, Project INCENTIVO/FIS/LA0010/2014, and grant SFRH/BD/52413/2013 (PD-F APPLAuSE)