

Fig. 5. Scheme of elongating a-CT.

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The catalytic reduction of carbon dioxide to carbon onion particles by platinum catalysts

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Since their discovery, carbon nanomaterials [1] have attracted a great deal of interest from many research groups worldwide, since they exhibit unique chemical and physical properties [2]. Both carbon nanotubes (CNT) and carbon onion particles (COP) have been prepared by a variety of methods that can be classified into

two broad categories: physical and chemical. These techniques have been reviewed recently [3], the physical methods include electric arc discharge synthesis, laser ablation, resistivity vaporization, electron or ion beam vaporization and sunlight vaporization. The chemical methods are, for example, the catalytic pyrolysis reactions of hydrocarbons, catalytic disproportionation of carbon monoxide (CO) [4,5], the reduction of perfluorinated hydrocarbons by an alkali metal amalgam, hydrothermal growth from amorphous carbon, catalytic reduction of CO and the thermal decomposition of Fe(CO)₅ in a

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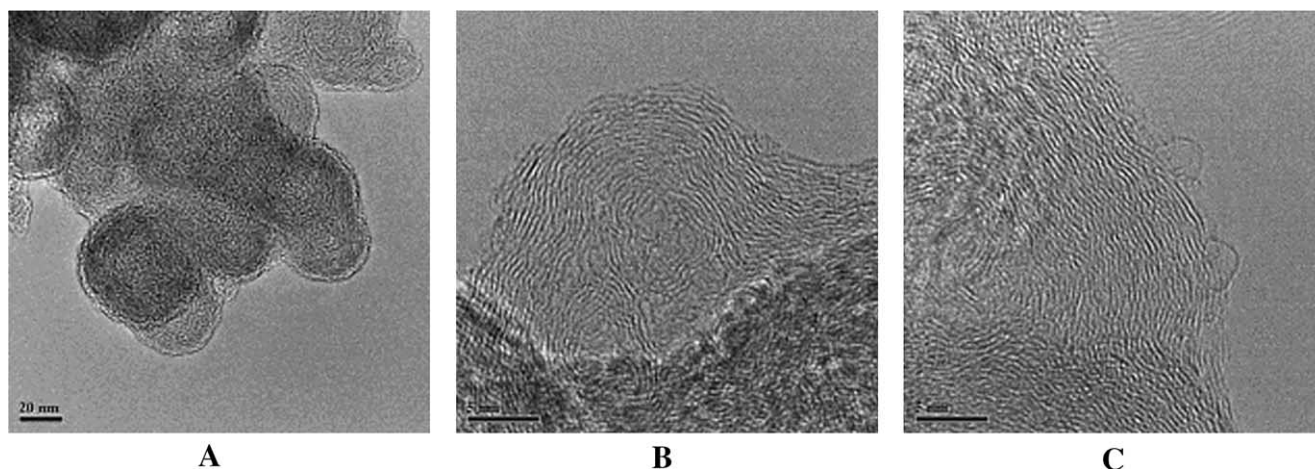


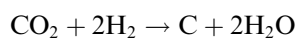
Fig. 1. HRTEM pictures of the product after workup. (A) Nanooxions, (B) twin spiral cores, (C) tilted multilayers.

flow of CO [6]. Among the chemical methods there are two reports for the use of carbon dioxide (CO₂) as a carbon source to produce carbon nanomaterials, both of which use supercritical CO₂, one with melted metallic magnesium at 1000 °C and a pressure of 10,000 atm [7] and the other one with melted metallic lithium at 550 °C and a pressure of 700 atm [8].

Because carbon dioxide is the final product of many industrial and biological processes, being the former a major source of global warming and since only plants can effectively regenerate useful products from it, the aim of the present report is to develop a low-temperature process for the preparation of carbon nanomaterials using CO₂ as the carbon source, based on the use of platinum thiaplatinacycles as catalyst precursors.

For a number of years we have been studying the synthesis of thiaplatinacycles of the type [Pt(η²-C,S-thiophene)(PEt₃)₂] and their role in homogeneous hydrodesulfurization (HDS) reactions [9,10] the former being useful intermediates for organic transformations [11] and active precursors in homogeneous HDS catalytic reactions [12].

In the present study, we investigated the reaction of CO₂ gas with thiaplatinacycles, such as [Pt(η²-C,S-C₁₂H₈)(PEt₃)₂], **1**, in a stainless-steel autoclave (300 mL), charged also with hydrogen and pressurized with argon up to 1380 psi at 40 °C (approx. 94 atm) to ensure the CO₂ being in a supercritical state (critical point, 31 °C, 73 atm), all used gases were in a high purity grade 99.998%. The main product identified was carbon, particularly in the form of carbon nanomaterials. Our interpretation of the chemical reaction that occurs is proposed as:



The HRTEM (high resolution transmission electron microscopy) micrographs depicted in Fig. 1 show the major structures obtained after workup. All the result-

ing structures turned out to be carbon nanomaterials, the main products (80%) being the well-known COP [13], Fig. 1A. Also, a few closely related particles with two spiral cores were observed [14], Fig. 1B, along with some tilted layers of carbon, characteristic of the ones present in multilayer CNT, Fig. 1C.

The X ray diffraction (XRD) pattern of the product shows a very small peak at $2\theta = 26.6^\circ$, regularly associated with the presence of graphite; the intensity of such peak is very small (Fig. 2), which is consistent with the low temperature used in the process [8].

Solid state ¹³C nuclear magnetic resonance (NMR) studies of the product obtained, were carried out by several groups to further characterize carbon nanomaterials (Fig. 3). The signal in a typical sample, located at δ 110.0, is considerably narrower (half-height line width is only about 10 ppm) than those observed previously [15], which may be attributed to uniform composition, packing or relatively few defects [16]. The absence of signals around 35 ppm and 179 ± 10 ppm implies that there are neither hydrocarbon nor graphite impurities, respectively.

The reaction, which was performed with thiaplatinacycles derived from dibenzothiophene [Pt(η²-C,S-

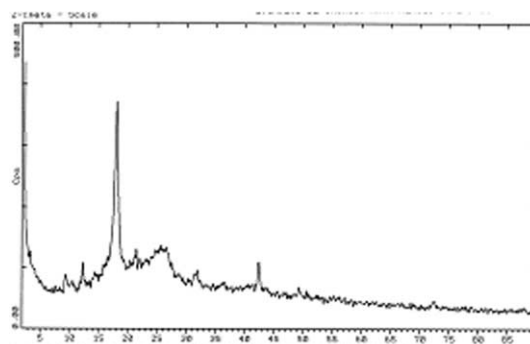


Fig. 2. XRD pattern for the sample.

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