CHARACTERISATION OF NANOPARTICLES PROCESSED BY ARC - DISCHARGE BETWEEN CARBON ELECTRODES CONTAINING Ni_2Y CATALYST

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INTRODUCTION

Processing of carbon nanotubes (CNT) is accompanied by formation of other by-products such as carbon rods, ribbons, encapsulates and onion-like structures. One of the most prospective catalysts for the synthesis of CNT appears to be Ni_2Y [1]. Proper control of the processing requires study of the amount, structure, composition and properties of the discharge products.

In this study the structure and magnetic properties of the products obtained by arch discharge between carbon electrodes containing Ni_2Y catalyst are investigated. Particular focus has been put on the ferromagnetic properties of carbon encapsulates.

EXPERIMENTAL

The cathode was made from bulk graphite. A hollow graphite rod, filled with the catalyst was used as an anode. Finely milled powder of Ni_2Y intermetallic phase was applied as the catalyst (specimen 1). The metallic powder was subsequently pressed into the hole in the graphite rod. The discharge was performed in helium gas atmosphere. The final carbon products, obtained in the course of discharge, were divided into four parts, depending on the collecting area. The first part, most plentiful, was collected from the chamber walls (*soot*). The second part grew up during the discharge around the cathode (*collar*). On the cathode grows also the third part - dense coating (*deposit*). At the bottom of the chamber accumulates material consisting of pieces of graphite, which ripped out of the electrodes and some soot. In these investigations only the soot (sample 2) and collar (sample 3) were studied. However, TEM studies were done for the collar and deposit. The magnetic properties were measured using a vibrating sample magnetometer EG&G PARC M4500. Curie temperature was measured using a Faraday magnetic balance. TEM studies were performed with the application of the HRTEM Jeol 3010. X-ray analysis was done with the application of the ADP-1 diffractometer using CrKα radiation. Assessment of the metallic particle size was done on a basis of the Sherrer formula.

RESULTS AND DISCUSSION

It is well established that nickel ions in the Ni_2Y compound do not contribute to the net magnetic moment [eg. 2]. However, in the course of milling, which follows the hydrogen decrepitation, partial decomposition of the Ni_2Y and formation of ferromagnetic and probably also superparamagnetic particles of metallic Ni may occur. The magnetic properties of this material are represented in Fig. 1 by loop 1. For the products of arch discharge (sot and collar, loops 2 and 3, respectively) the magnetisation values close to saturation can be achieved in much lower field of 0.5 kOe (400 kA/m). Assessment of the Ni content on the basis of magnetisation gives values 2 and 8 wt%. in the soot and collar, respectively. If this assessment is made on a basis of absorption spectroscopy the respective contents are 11 and 14 wt.% of Ni. Thus, we suppose that not all Ni particles existing in the product are ferromagnetic. Some of them can be either superparamagnetic or in a form of a diamagnetic compound.

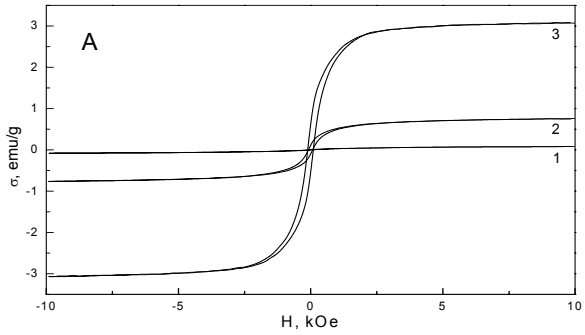

Fig. 1. Hysteresis loops for the specimens investigated The loop number represents the specimen number.

X-ray patterns for the discharge products are shown in Fig. 2. Two diffraction peaks having 2Θ=68.4 and 80.9° represent (111) and (200) crystallographic planes for the FCC Ni lattice, respectively. For the collar (specimen 3) the 68.4° peak represents superposition of two reflections: a narrow one and a wide one (Fig. 3). The presence of the narrow and wide peaks gives an evidence

for the existence in the products of broad distribution of particle size. For the collar, the calculations of the particle size, D_{111}, give values 42.4 nm and 5.4 nm for the large and small crystallites, respectively. In the soot the particle size is more homogeneous with the mean size about 5 nm.

Studies of the lattice constants for the plane (111), for all the specimens investigated, exceeds those for pure nickel [3]. This difference can be explained by the dissolution of some carbon in Ni. Assuming that the increase of the lattice parameters is fully related to the carbon dissolution, we can estimate the carbon content to be 2 wt.% for the peak d_{111}=2.058Å. The idea of carbon dissolution can further be confirmed by the fact that the increase of the coercivity for the particles follows change of the lattice parameters. Moreover, the measurements of the Curie temperature, for the specimen 3, gives a value of 350 °C, which is somewhat lower than those for the pure Ni (354.4 °C).

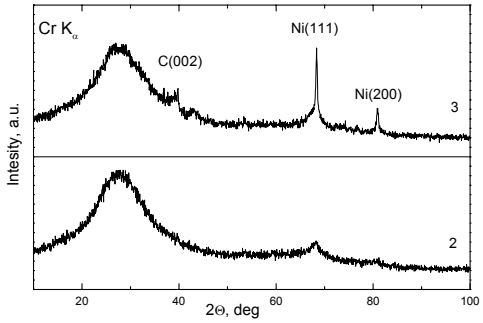

Fig. 2. Diffraction patterns for the soot (specimen 2) and collar (specimen 3). Number of pattern represents specimen number.

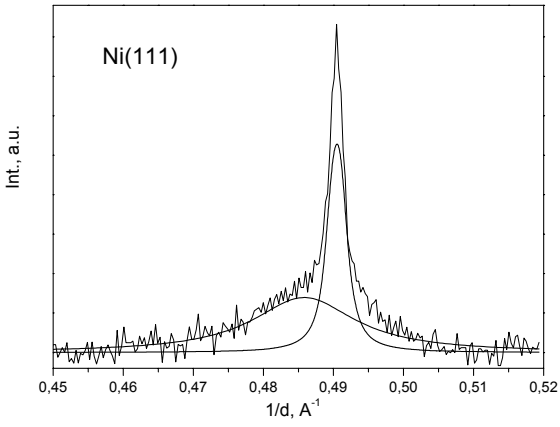

Fig.3. Partition of the Ni (111) diffraction line for the contributions from small and large particles.

Carbon encapsulates are clearly visible in the discharge products (Fig. 4). The electron diffraction analysis proved that the encapsulates contain yttrium oxide and nickel in the deposit and collar, respectively. This means that the starting catalyst Ni_2Y decomposes in the course of arc-discharge and the yttrium locates in the deposit and Ni goes mainly to the collar and in smaller amount to the soot (from magnetic measurements). The encapsulates, which are particularly of interest in these investigations, have size within a range of 5 to 40 nm, which generally agrees with the calculations from the x-ray studies.

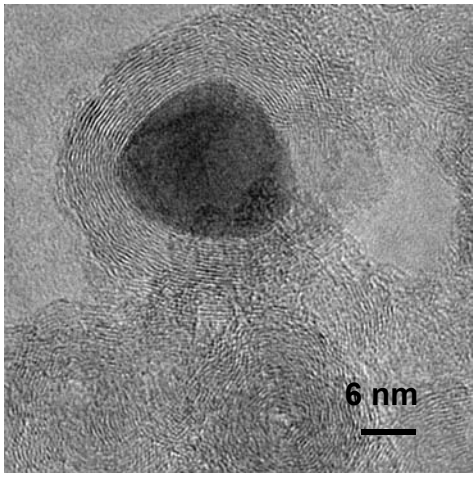

Fig. 4. Ni encapsulate in the collar.

Summarising, it has been found that the arch-discharge between the composite electrodes graphite - Ni_2Y results in a decomposition of the intermetallic Ni_2Y phase and formation of Ni ferromagnetic nanoparticles, encapsulated in the carbon shells, in the collar and soot, and yttrium oxide in the deposit. The Ni particles, due to the carbon dissolution, exhibit increased lattice parameter, when compared with the pure metal, which grows with decreasing their size. The carbon concentration in smaller crystallites exceeds those in the larger ones.

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