GROWTH STUDIES AND PROPERTIES OF FERROMAGNETIC FILLED CARBON NANOTUBES

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Several authors have published papers about synthesis and properties of metal-filled carbon nanotubes [1 – 3]. Among many applications such structures can be used for data storage [4] or in biomedicine. Current applications mostly require a defined nanotube dimension and composition of filling material. To comlpy with these criteria a good understanding of the growth process is necessary.

In this work we discuss the context between different catalyst materials, the process conditions, the nanotube geometry, the structure of filling material (in cross section/ along the tube axis) and the magnetic properties.
The samples with metal-filled carbon nanotubes were prepared by pyrolysis of metallocenes (ferrocene, cobaltocene) or metallocene derivatives (e.g. cyclopentadienyliron dicarbonyl dimer) on oxidised silicon-wafers predeposited with 2 nm thin layers (Fe, Co).
Two methods were employed for the synthesis of filled carbon nanotubes. The first method, a classical hot wall reactor with a two zone furnace system was used to study the growth process in detail. The second method, a liquid source apparatus (LSCVD), which consists of a band evaporator and a hot wall reactor, was prefered to coat the substrates with high aligned nanotubes (Fig. 1). Both deposition processes were performed in the temperature range between 750°C and 925°C and in an argon flow.

At the beginning of our experiments we have determined the size of metal-particles (Fe, Co) on the surface of oxidised silicon wafers after thermal treatment by scanning electron microscopy (SEM). From these investigations the particle diameter distributions were obtained (Fig. 2). In comparison with cobalt the mean particle diameter of iron is shifted to larger values, this may be due to lower surface tension of liquid iron on the oxidised silicon substrate. It is well known that the nanotube diameter depends on the particle size. Therefore we have measured the outer diameters of nanotubes grown on oxidised silicon-wafers and observed, that the metal-particle diameter was in good approach with the tube diameter.
By using the chosen process conditions the growth rate of the nanotubes decreases with the deposition time. From short time experiments with duration of 30 s and 1 min we estimated starting growth rates between 30 and >100 nm/s. X-Ray diffraction (XRD), selected area electron diffraction (SAED) and transmission electron microscopy (TEM) investigations show that the iron phase inside the nanotubes comprise α-Fe, γ-Fe and Fe_3C, depending on the process conditions (Fig.3). At temperatures lower 825°C the major fraction is α-Fe in accordance with high measured saturation magnetization moments (up to 0,8 memu/mm²) and coercivities (up to 160 mT). We assume the distribution of the different iron phases is the following (in cross section): α-Fe in the centre, surrounded by γ-Fe and Fe_3C at the boundary to the graphite-shells of the tube. This result is in good agreement with Mössbauer studies performed by Ruskov et al. [5]. The filling degree decreases from the base to the top of the nanotubes. At the top particles are usually encapsulated. In conclusion to our results we propose a growth mechanism, finally.

In respect to the high potential for filled carbon nanotubes especially by filling with magnetic transition metals or alloys, we have started a research program for application of such filled nanotubes in the cancer diagnostic and therapy [6].

References:

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[4] S. Y. Chou et al., *J. Appl. Phys.* **76** (1994) 6073
[5] T. Ruskov et al., *J. Appl. Phys.* **96** (2004) 7514.
[6] I. Mönch et al., *J. MMM* (2005) (in press).

Figures:

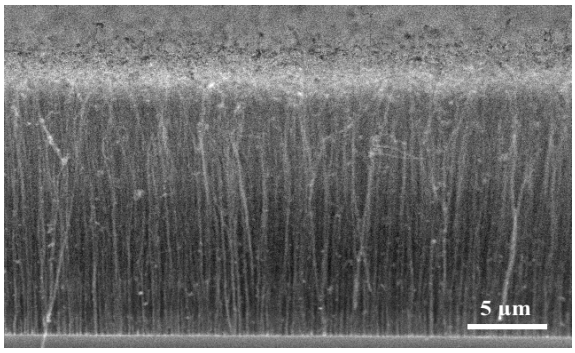

Fig. 1. SEM image showing aligned iron-filled MWCNT

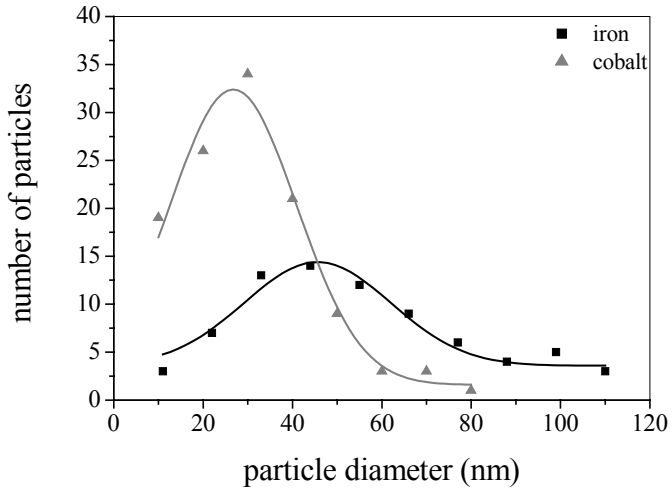

Fig. 2. diameter distribution of catalyst particles at 825°C (total particle-number per µm²)

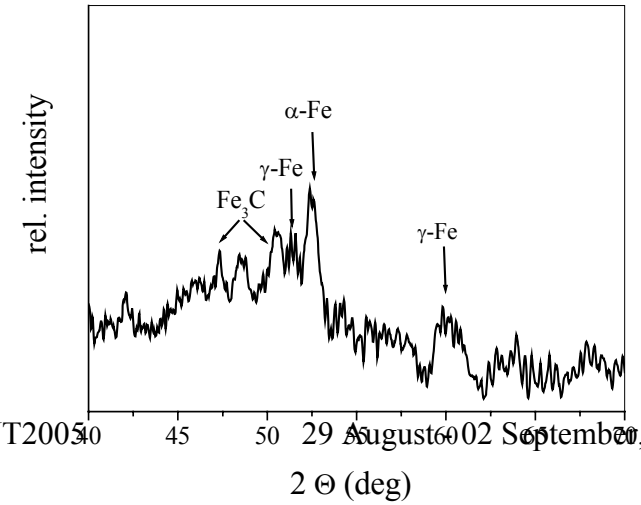

Fig. 3. XRD pattern of iron-filled MWCNT