PREPARATION OF C60 THIN FILM BY THERMAL VACUUM EVAPORATION

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Abstract: Results of thermal vacuum evaporation method used for C60 thin films preparation are presented. Fullerene molecules were obtained by arc discharge. The deposition was made on glass, quartz, KBr and mica substrates. The influence of the sublimation temperature and substrate temperature is investigated in order to optimize the deposition, in terms of deposition rate and uniformity.

Keywords: fullerene, nanomaterials, thin films, vacuum deposition

1. INTRODUCTION

Fullerenes, a new form of carbon, were discovered in 1985 [1] in graphite vaporization under inert gas at low pressure. Fullerenes have many properties different from either diamond or graphite. Potential applications include superconductors, sensors, catalysts, optical and electronic devices, polymer composites, high-energy fuels and biological and medical materials. But wide application of fullerenes is undetermined by an inability to produce large and inexpensively quantities. The arc discharge method developed by Kratschmer's group [2] in 1990 remains until now the major tool for synthesizing fullerenes. Several works has been done for the macroscopic production of the fullerenes by using other different techniques, laser ablation [1], electron beam evaporation [3], heat resistive method [4], diffusion flame [5] and ion beam sputtering [6].

Experimental studies of C60 layers deposited on different substrates have been carried on by many groups with the aim to understand the mechanism of growth of ordered layers in various conditions. The fundamental problem was the qualification of the layer-substrate interaction responsible for the introduction of crystalline order into the grown layer. The problem of influence of the lattice mismatch and deposition temperature on the orientation and size of obtained crystallites was studied. For this purpose, different kinds of substrates were used, e.g. GaAs [110], Si[100], Si[111], CaF, MoS, mica and many other metallic substrates such as Au, Ag and Cu [7]. C60 mono- and multilayered structures have been obtained in the ultra-high vacuum system as molecular beam epitaxy, although layers were also grown at the higher pressure by traditional vacuum evaporation.

It was originally assumed that the solid aggregate of C60 fullerenes had a hexagonal close-packed structure. However, recent x-ray diffraction studies have shown unambiguously that it adopts the face-centered cubic (fcc) structure (providing that all solvent molecules are eliminated). Synchroton x-ray powder profile gives a lattice constant a = 1.417 nm. This value implies the close packing of pseudospheres having a diameter of 1.002 nm. This is consistent with the fitted radius of the C60 skeleton of 0.353 nm and a carbon van der Waals diameter of 0.294 nm, which is slightly smaller than that of graphite (0.335 nm). The intermolecular bonding is dominated by van der Waals forces, as confirmed by measurements of the isothermal compressibility. The C60 aggregates, grown from solution, are shiny and black and reach 300 pm in size [8].

This paper presents the results obtained in C60 thin films deposition using thermal vacuum sublimation. The influence of the sublimation and substrate temperatures are investigated.

2. EXPERIMENTAL SETUP

The fullerenes were prepared by evaporation of carbon electrodes in an electric arc discharge process in helium atmosphere. The experimental setup is presented in [9]. In order to obtain fullerene thin films, a special deposition installation was realized. In figure 1 the sketch of the deposition installation is presented. The installation is mounted on a VUP-4 vacuum system. The main part consists in a pure graphite cylindrical evaporator (4). In order to eliminate the volatile impurities, the evaporator was heated for two hours to 1173 K in vacuum.

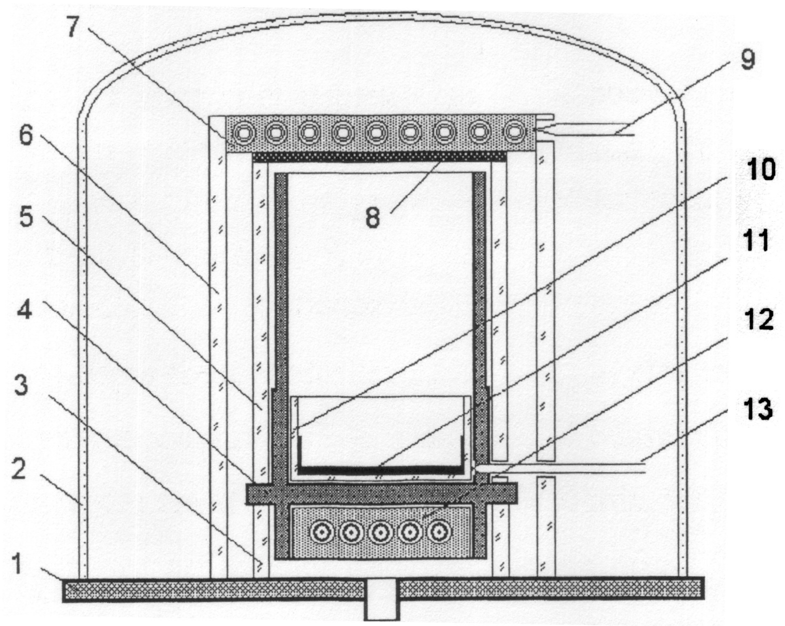

Fig.1. Schematic diagram of thin film deposition system. 1) vacuum installation support; 2) deposition chamber; 3) quartz support; 4) graphite cylinder; 5), 6) quartz cylinders; 7) substrate heater; 8) substrate; 9), 13) thermocouple; 10) quartz container; 11) C60 material; 12) heater.

The reactor walls maintain the temperature gradient along the evaporation direction. The temperature is maintained with the heater 12. Inside of the graphite reactor is placed a quartz container, filled with the C60 material, in molecular form. The substrate 8 is fixed on the quartz cylinder 5 and is temperature controlled with the heater 7. In order to make furthers optical and electrical measurements, the films were deposited on glass, quartz, KBr and mica. Prior deposition the substrate plates were cleaned in $H_2O_2/H_2SO_4/H_2O$ solution and with distillated water. After that, the substrates were dried in vacuum.

3. RESULTS AND DISCUSSION

It is not easy to growth fullerene thin films because of the low vapor pressure and high condensation speed. The results obtained using various deposition methods are presented in Table 1.

In our case, the deposition was made on glass, quartz, KBr and mica substrates, heated to 413 – 493 K, with various sublimation temperatures, in the range 650 – 830 K. On glass substrate, uniform films with 2 – 2,5 µm are obtained. On other substrates, the obtained thickness was 1 – 1,5 µm.

The dependence between deposition rate and sublimation temperature for several substrate temperature is presented in figure 2.

Table 1.

Deposition method	Process parameters			Film parameters	
	Pressure (Torr)	$t_{sustrate}$ (°C)	$v_{deposition}$ (Å/min)	substrate	Crystallite dimensions (μm)
Vacuum sublimation	10^{-6}		5-10	glass	0,01
		20	15	Si	0,03
				SiO_2	amorphous
	10^{-6}	20-200	10	mica	0,1
	10^{-6}	100	15	NaCl	0,01
	10^{-6}	240	5	KI(001)	0,3
	10^{-6}	20	20	Si(100)	0,02
Molecular epitaxy	10^{-9}	200	0,1	mica	0,4
				NaCl	0,2
				MoS_2	1
	10^{-9}	20-200	0,3-1	Ge(001)	
	10^{-9}	200	0,5	Si(111)	0,1
Sublimation in furnace with temperature gradient	6.10^{-7}	200	7	glass	1
Heated wall	10^{-7}	140	1,3	mica	0,05-0,1

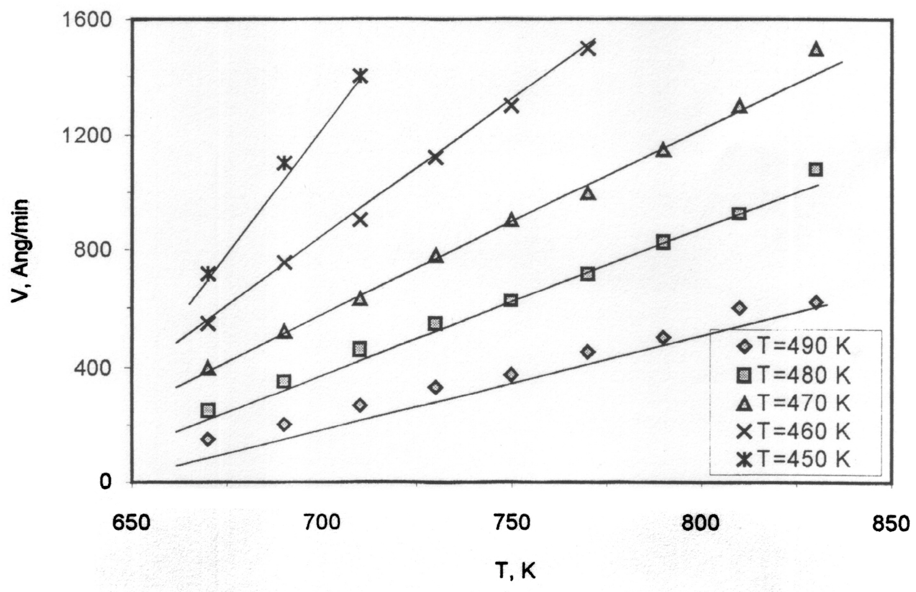

Fig.2. Dependence between deposition rate and sublimation temperature for several substrate temperature

When the sublimation temperature increases from 670 K to 840 K, the deposition rate increases rapidly. The increase is almost linear. When the substrate temperature increases, the deposition rate has an important reduction, because of the re-evaporation of C60 molecules on substrate.

In order to optimize the deposition conditions, the distance between vapor source and substrate was also modified using cylinders with various lengths. For all cases, the films exhibit very good thickness uniformity indicating an almost zero temperature gradient on the substrate surface. The efficiency of the deposition, in terms of films thickness, is presented in figure 3. In this figure the dependence between source-substrate distance and film thickness is shown, for two substrate temperatures. For both cases, the maximum efficiency appears when the ratio between length and diameter of the graphite cylinder is around unity.

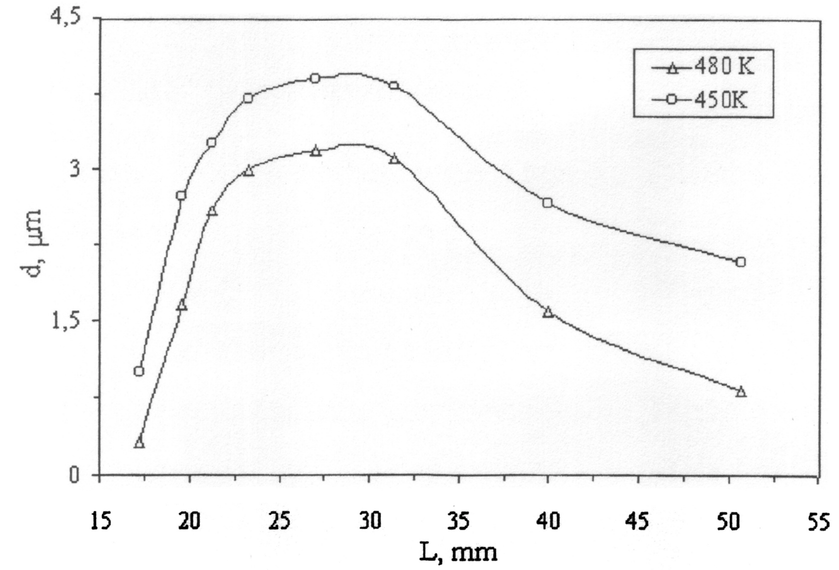

Fig.3. Dependence between source-substrate distance and film thickness for two substrate temperatures when the sublimation temperature is 790 K

4. CONCLUSIONS

This paper describes a fullerene thin films deposition system, based on vacuum evaporation (sublimation). When the sublimation temperature increases, the deposition rate increases rapidly. When the substrate temperature increases, the deposition rate has an important reduction. The films exhibit very good thickness uniformity indicating an almost zero temperature gradient on the substrate surface. The maximum efficiency, in terms of films thickness, appears when the ratio between length and diameter of the graphite cylinder is around unity.

5. AKNOWLEGMENTS

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