Proceedings of the Korean Nuclear Society Spring Meeting
Kwangju, Korea, May 1997

Pyrolytic Carbon Coating on A Simulated Fuel by Fluidized Bed Type Chemical Vapour Deposition

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Abstract

Pyrolytic carbon layer was coated on Al_2O_3 balls by fluidized bed type chemical vapour deposition unit to develop the coating technology for the preparation of coated nuclear fuel. The deposition was carried out at the temperature ranges between 1100℃ and 1300℃ with various gas contents and flow rates. Source and carrier gas were propane and argon, respectively. X-ray analysis shows that the deposition layer was typical carbon spectra. The growth rate of carbon layer depended on the amount of source gas and the deposition temperature. For the alumina balls with 2 mm in diameter, the deposition rate was 11 μm/hr in the flow gases containing 30% source gas at 1300℃ with a total flow rate of 2.0 *l*/min. Microstructural observation of the deposits with scanning electron microscope revealed that the deposits had relatively dense and isotropic structure. Chemical analysis by energy dispersive spectroscopy showed that the layer was pure carbon.

1. Introduction

Pyrolytic carbon coating on oxide fuel has attractive attention in nuclear field because of ifs superior performance of retaining the fission products[1, 2]. For the uniform coating on the nuclear fuel surface, a fluidized bed type chemical vapor deposition unit is generally used. Since the coated layer has two-dimensional crystal lattice the anisotropic orientation of each crystal result in determining the physical and mechanical properties such as density and strength.

The pyrolytic carbon layer is prepared by the thermal decomposition of various source gases including carbon atom such as methane, acetylene and propane. The microstructure of the deposits significantly depends on the coating condition[3]. The pyrolytic carbon is generally classified according to arrangement of crystallite into isotropic, laminar, columnar and granular structure[4]. From an engineering point of view, isotopic structure for the nuclear fuel and columnar structure for the rocket nozzle are recommended, respectively because of their structural orientation.

This study was initiated to develop coating technology and to prepare the coated nuclear fuel as an advanced concept for present power plant. Systematic study on the effect of processing parameters in coating technology together with the phase identification and observation of microstructure was carried out with X-ray diffractometer and scanning electron microscope.

2. Experimental method

Figure 1 shows the schematic diagram of the carbon deposition unit, which consists of a 1500 kW resistance furnace, temperature controller, gas controller system and alarm/monitoring systems for the unit. Inside of the furnace, a cone-shaped graphite coater with 50 mm in diameter, 300 mm in length and 60° in cone angle is installed. The spherical alumina balls with 2 mm in diameter were used for the simulated fuel particles. The total gas flow during each experiment maintained constant by gas flow meter. Propane(99.9%) and argon gas were used as a carbon source and a carrier gas, respectively. The total pressure in the coating chamber was 1 atm. The source gas and carrier gas were blended in a gas mixer with various compositions and preheated at 400℃ in pre-heater before flowing into the coater. The coating temperature was measured by Pt/Pt-5%-Re thermocouples contacted to the graphite coater in the furnace. Coating conditions employed in this study are shown in Table 1. The deposition rate was determined by measuring the thickness of the deposit layer on the reference graphite disk in the coater. The phase identification of the deposits was carried out with X-ray diffractometer(Mac Science model MXP3A-HF2000). The structure of each pyrolytic carbon deposit was observed by scanning electron microscope(Jeol 6400).

3. Results and discussion

Figure 2 shows the cross sectional view of the coated layer, which reveals that homogeneous and well deposited on the spherical alumina matrix. The thickness of the layer after 40 minutes coating was about 10 μm. Figure 3 shows the X-ray diffraction pattern of the deposits on the reference disk. It shows the typical graphite spectra without any detectable spectra of other elements.

Figure 4 shows the effects of the composition of flow gas and the coating temperature on the deposition rate with various total flow rates. Deposition rate increased as both the temperature and the flow rate increased. The temperature dependence of the deposit layer is related to the thermal decomposition of propane. The propane can be thermally decomposited above the temperature of about 900℃ [4]. Since the decomposition process is endothermic, the increased coating temperature resulted in enhancing the decomposition process.

The denser carbon content in the flow gas produced the thicker coated layer due

to the amount of carbon in the flow gas, which was thermally decomposed inside the coater. Although the density of the coated layer was not determined in this study, it is clear that the deposition rate was significantly dependent upon the thermal decomposition of carbon in the source gas.

There are four kinds of microstructure of pyrolytic carbon such as isotropic, laminar, columnar and granular structure. The structure was classified according to whether or not containing grains inside of the deposit layer: isotropic for the layer without cross pattern and grains, laminar for cross pattern without grains. granular for discrete grains and columnar for discrete elongated grains. Figure 4 shows the high magnification of the cross section of the carbon layer deposited in this study, which shows the typical isotropic structure. The formation mechanism of isotropic microstructure is not clear. It can be, however, explained as suggested by Bokros [3]: at the initial stage of coating, the deposition area is relatively small so that granular structure is formed immediately. As the deposition proceeds, the radius of the particle is increased so that the surface area becomes too large for granular structure to form and a transition into an isotropic structure occurs.

Figure 5 is the chemical analysis by the EDX on the coated surface, showing a typical carbon spectra. Since the isotropic microstructure of pure pyrolytic carbon was recommended for the nuclear fuel, the coating conditions selected in this study can be used for the preparation of the coated nuclear fuel.

4. Conclusions

To develop coating technology for the nuclear fuel as an advanced concept for existing power reactors, a systematic study on the effect of processing parameters on the microstructure of the deposit was carried out and from the study, can be driven the following conclusions :

(1) The homogeneous and pure isotropic carbon layer was well deposited on a spherical alumina ball.

(2) The deposition rate was 11 μm/hr in the flow gases containing Ar-30% C_3H_8 gas at 1300℃ with 2.0 *l*/min total flow rate.

(3) The deposition rate significantly depended on the thermal decomposition of carbon in the source gas.

References

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[3] J. C. Bokros, Carbon, vol. 3, 1965, pp. 17-29

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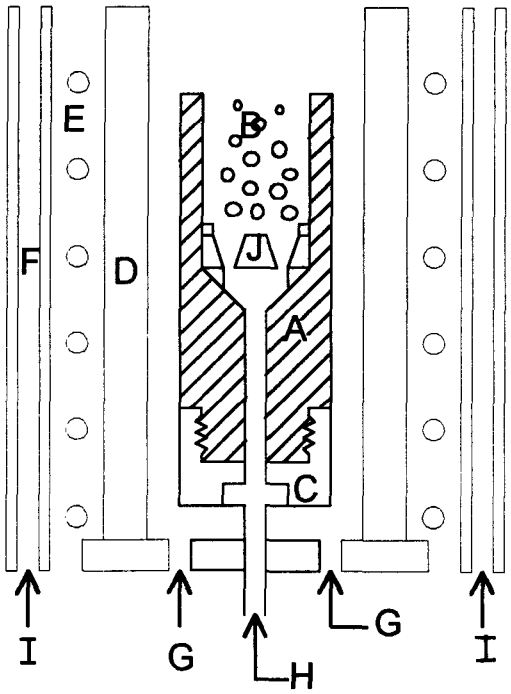
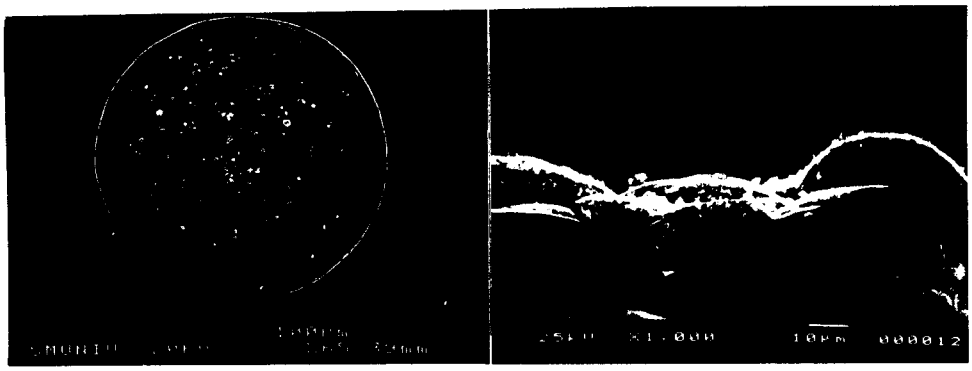
Table 1. Coating conditions of pyrolytic carbon in fluidized chemical vapor deposition unit

deposition temperature [℃]	1100~1300
total flow rate [l/min]	1.5~2.0
composition of source gas [%]	5~30
total weight of specimen installed [g]	80
size of each sample [mm]	dia. 2.0
preheating temperature [℃]	400
cone angle of coater [degree]	60
size of coater [mm]	I.D 50x300
cone angle of distributer [degree]	60
size of distributer [mm]	I.D 23x20
nozzle size of distributer [mm]	1.0



A : graphite coater
B : sample
C : stainless connecter
D : alumina tubing
E : heater
F : water jacket
G : argon gas in
H : coating gas in
I : cooling water in
J : separator

Fig. 1 Schematic diagram of the carbon deposition unit.



(a) (b)

Fig. 2 Scanning electron micrograph of the cross sectional view of the coated layer.

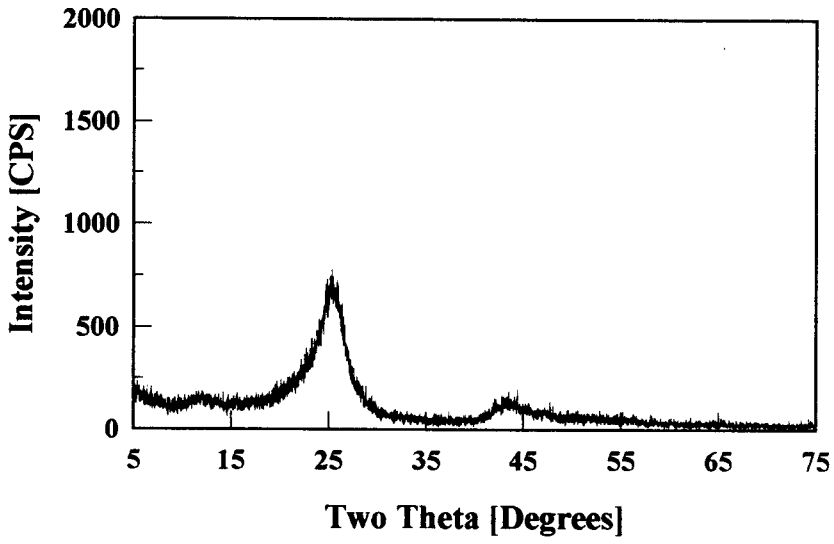


Fig. 3 X-ray diffraction spectra of the deposits on the reference disk.

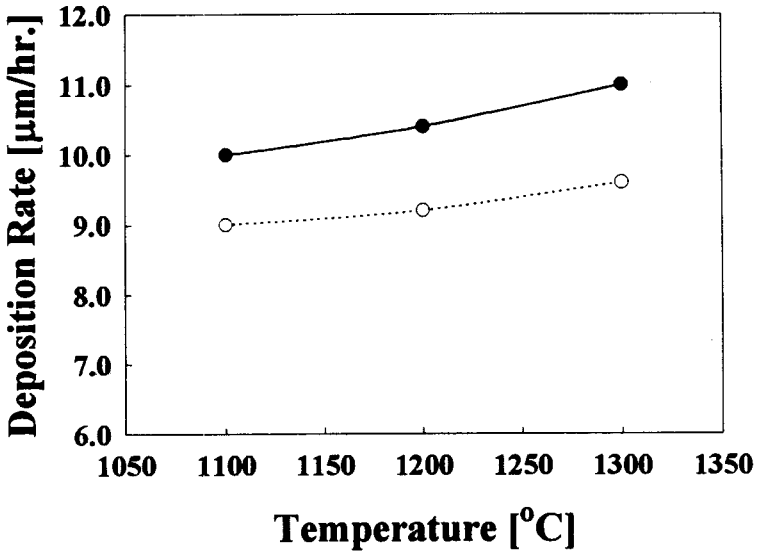


Fig. 4 Effects of the composition of flow gas and the coating temperature on the deposition rate. (● : 2.0 *l*/min ○ : 1.5 *l*/min)

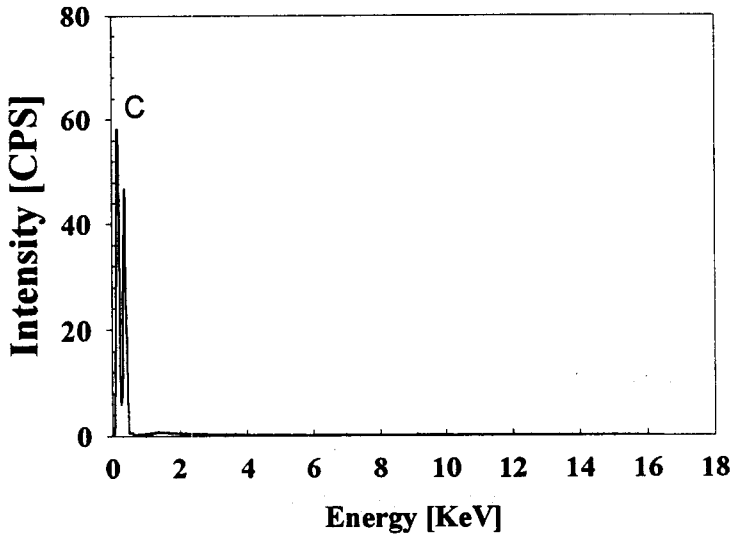


Fig. 5 EDX spectra measured on the coated surface.