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Journal of Nuclear Materials 224 (1995) 12–16

journal of nuclear materials

Effect of deposition temperature on the properties of pyrolytic SiC

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Received 12 January 1995; accepted 20 February 1995

Abstract

TRISO-coated fuel particles have been chosen for the fuel of our 10 MW HTR experimental reactor. SiC is the key component of the coatings for TRISO coated particles. In the present work, the study of the effect of the deposition temperature on the density, surface morphology, fracture surface structure, strength and Young's modulus of the deposited SiC is reported.

1. Introduction

The release of radioactive fission products causes safety and environmental problems. It is one of the major issues which have to be solved in the development of nuclear energy technology. Radioactive fission products have to be confined at the places where they are produced. In the HTR, the basic unit which confines the radioactive fission products is the coated fuel particles (cp) [1]. There are two kinds of cp, TRISO and BISO, which are fully developed at the moment. TRISO cp (with a SiC coating layer) have a much higher ability in retaining solid fission products and have a higher irradiation stability than BISO cp (without a SiC coating layer), and are regarded as the standard fuel for the future HTR [2]. Extensive R&D programs of this type of cp have been carried out [3–6] for more than 30 years.

An R&D program of TRISO cp has been established for the fuel of the HTR-10 experimental reactor which is to be built at the site of our institute [7,8]. SiC is the key component of the coatings, the quality of SiC is vital to the performance of TRISO cp in service. The investigation of the effect of deposition temperature on the density, surface morphology, fracture surface structure, strength and Young's modulus was carried out, and the results are reported in the present paper.

2. Deposition of SiC coatings

The deposition of SiC coatings was carried out in a 2″ fluidized bed, the schematic diagram of the coating is shown in Fig. 1. The coating procedure of the SiC layer was the following: the fluidized bed was heated under flowing Ar up to the temperature set for the experiment. Particles which had already been coated with a buffer layer and an inner dense PyC layer were loaded into the fluidized bed. The Ar flow rate was adjusted to optimize the fluidizing state. Methyltrichlorsilane (MTS) was introduced into the fluidized bed, which decomposed and formed SiC. The formed SiC deposited onto the substrate for the time preset. After deposition, the fluidized bed was cooled down under flowing Ar to a temperature lower than the threshold of the oxidation temperature of graphite. Ar was switched off, the fluidized bed was cooled further down to room temperature, then the coated particles were unloaded.

The temperature range studied was from 1450 to 1650°C, the temperature step was 50°C. MTS was metered in by heating the MTS evaporator located in a thermostat to the temperature preset, H_2 was used to sweep the MTS vapour into the main stream of the coating gas. Fig. 2 is the schematic diagram of the MTS supply system used. The concentration of MTS was adjusted by changing the temperature of the thermo-

SSDI 0022-3115(95)00039-9

stat and/or the flow rate of the sweeping gas, the amount of MTS metered in was first predicted by the MTS vapour pressure at the temperature set and the sweeping gas flow rate, and later checked by weighing the MTS evaporator before and after the coating process. In present work, the concentration of MTS was kept at about 1 vol%. Other parameters of the deposition were:

- particle load: 80 g,
- Ar/H_2 ratio: 1/1,
- total gas flow rate: 500 l/h.

3. Effect of deposition temperature on the density of SiC

The effect of the deposition temperature on the density of SiC is shown in Fig. 3. It can be seen that the density of SiC deposited at 1450 and 1550°C reaches the theoretical value. However, the drop of density at 1500°C is odd. SEM observations (Fig. 4 and Fig. 5) show that the density of SiC deposited at 1450°C should be lower than that of SiC deposited at 1500°C. The measurements of strength and Young's modulus show that SiC deposited at lower temperature has lower values than SiC deposited at higher temperature. Density measurements were repeated three times, the result remained unchanged. The reason of this abnormal phenomenon is not clearly known yet. One explanation could be that the density was measured by the titration method, the SiC deposited at lower temperature has a porous structure, the pores are interconnected and open to outside, the titration liquid penetrates into the pores, therefore, instead of the apparent density, the measured density is the true density. The apparent density of SiC deposited at 1450°C may actually be lower than that deposited at 1500°C.

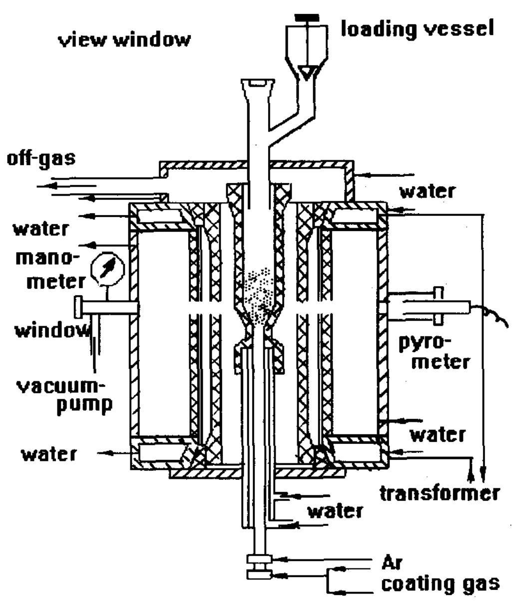

Fig. 1. Schematic diagram of the coating.

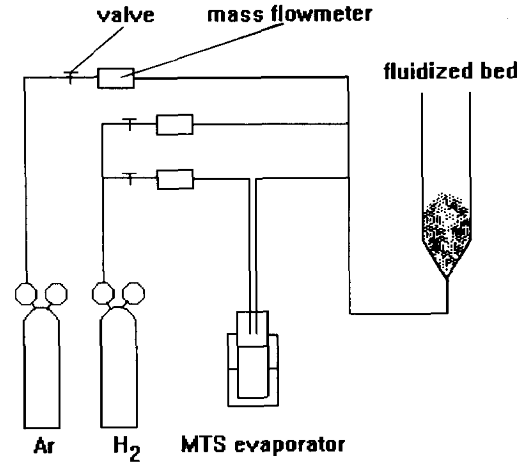

Fig. 2. Schematic diagram of the MTS supply system.

4. Effect of deposition temperature on the surface morphology

The surface morphology of deposited SiC is shown in Fig. 4. The outer surface morphology changes remarkably with increasing deposition temperature. SiC has a cauliflower structure when it was deposited lower than 1550°C, and the lower the temperature, the rougher the cauliflower structure. A granular morphology dominates the surface structure when the deposition temperature was higher than 1600°C, the higher the deposition temperature, the coarser the granular morphology.

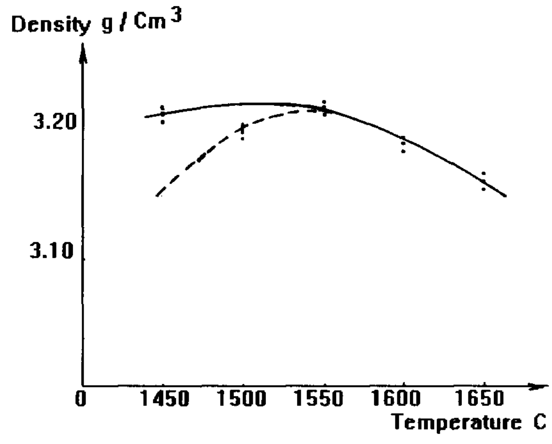

Fig. 3. The effect of deposition temperature on the density of SiC.

5. Effect of deposition temperature on the fracture surface structure

Fig. 5 illustrates the fracture surface structure of SiC deposited between 1450 and 1650°C. The fracture surface of SiC deposited at 1450°C is characterized by an inhomogeneous and small grain size, by highly developed micropores with a small amount of large pores scattered on the fracture surface and by intergranular fracture together with some transgranular fracture. At 1500°C, the micropores disappear, there are some isolated holes on the fracture surface and transgranular fracture occupies quite a large area. At 1550°C, the structure becomes homogeneous, the pores disappear, grain coarsening starts and the transgranular fracture dominates the fracture surface. At 1600°C, the structure becomes inhomogeneous, the grains become coarser and there are some pores on the fracture surface, intergranular fracture can occasionally be seen at some places. At 1650°C, the structure becomes very

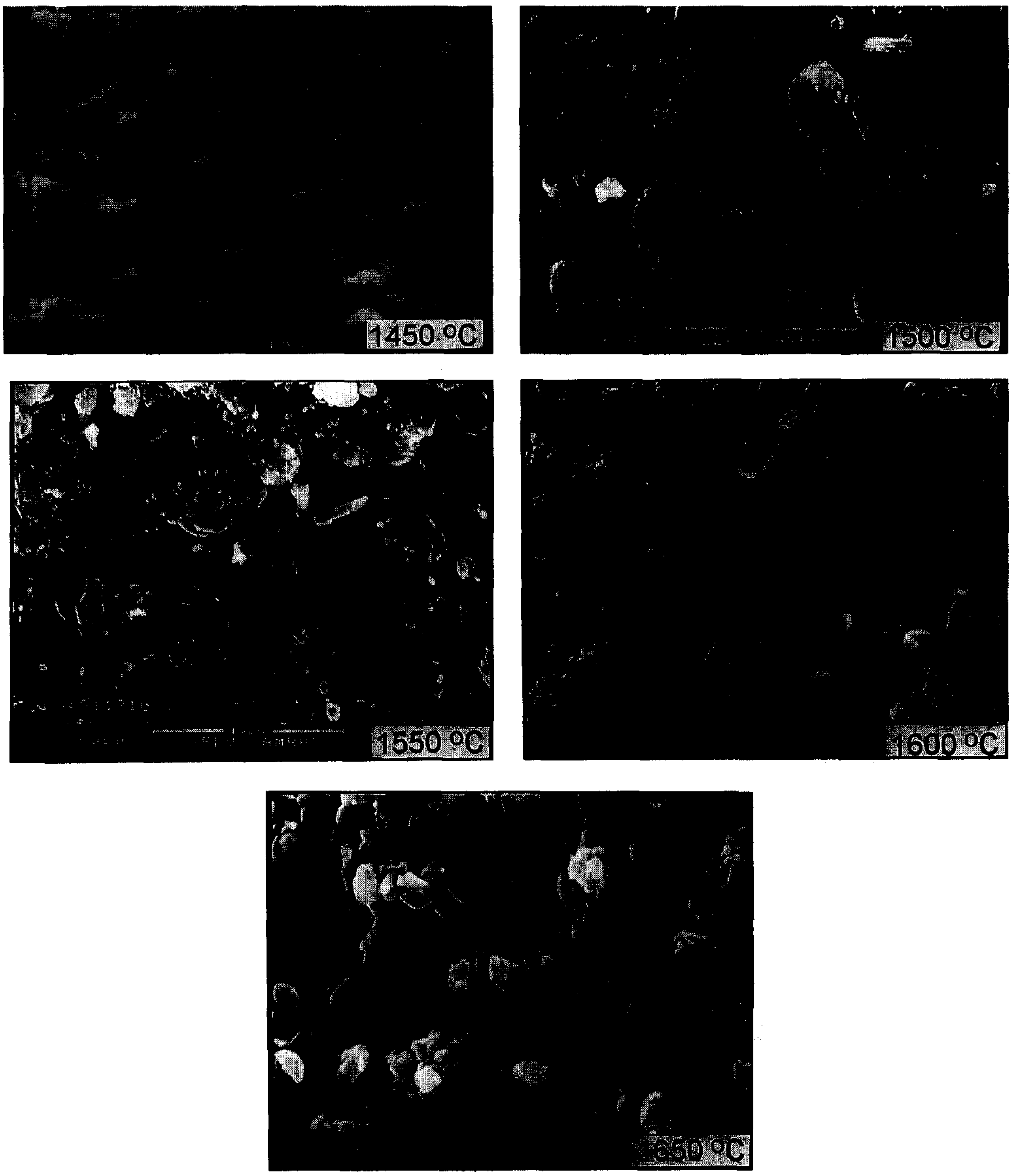

Fig. 4. Effect of temperature on the surface morphology of SiC.

porous, large pores spread all over the fracture surface can be seen.

6. Effect of deposition temperature on the strength and Young's modulus of SiC

Strength and Young's modulus are very important parameters of SiC, especially in TRISO cp. It is assumed that the SiC coating layer is the only layer which confines the fission products and the fuel [9], therefore the mechanical performance of TRISO cp will be dependent on these parameters. They are indispensable input data for cp design and also serve as an important characterization parameter in quality control during production of cp. An apparatus for determining these parameters has been developed and manufactured [10] according to the principle of brittle ring test developed at KFA Jüulich, Germany. Samples were prepared manually by metallographic methods, the sampling size

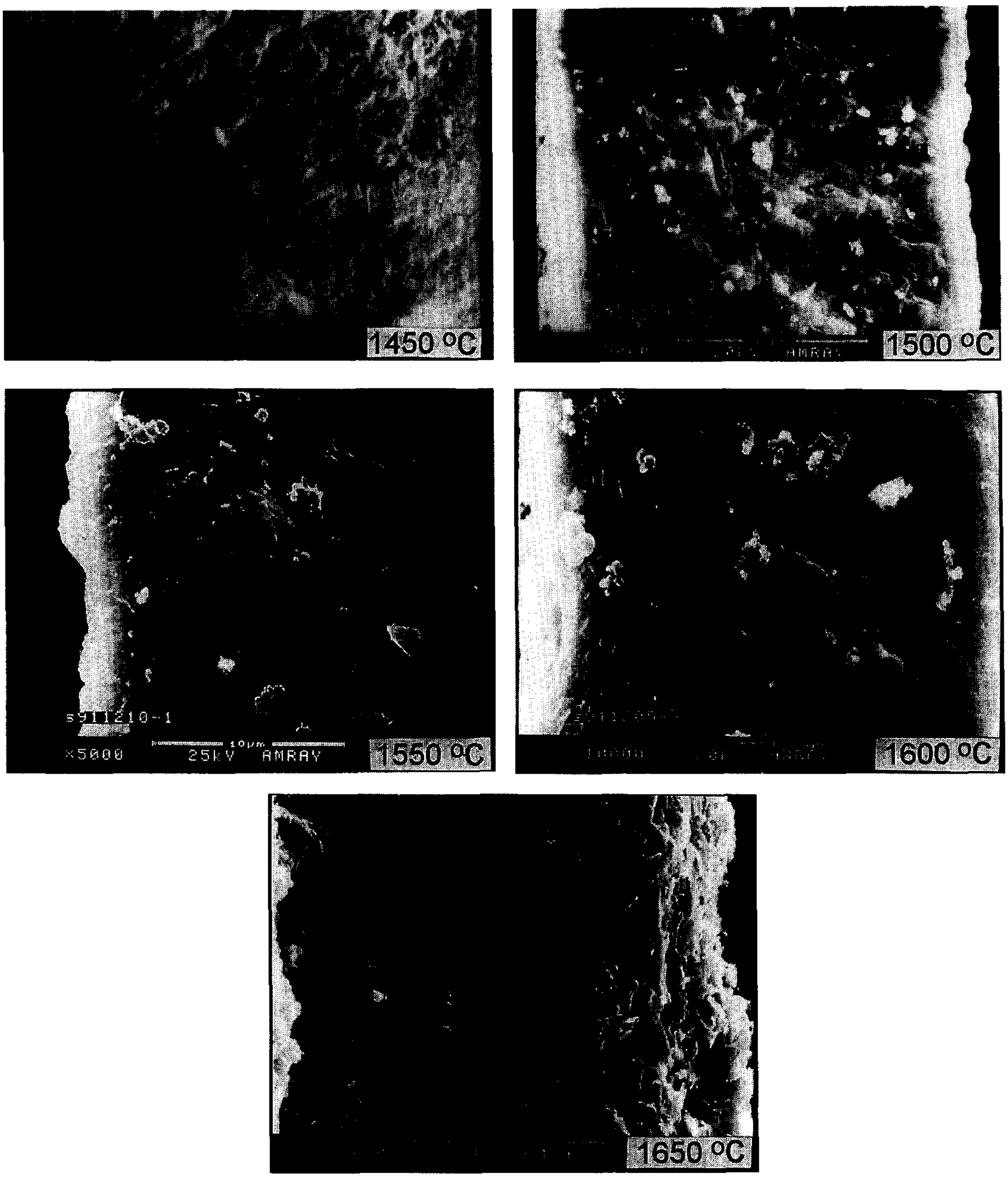

Fig. 5. Effect of temperature on the fracture surface structure of SiC.

Table 1
The effect of deposition temperature on strength and Young's modulus of SiC

	Temperature (°C)				
	1450	1500	1550	1600	1650
Strength (MPa)					
average σ	670	1285	1447	783	693
standard deviation s	146	223	269	153	214
s/σ (%)	22	17	19	21	31
median strength	659	1282	1412	780	680
Weibull parameter m	5.4	6.4	6.6	5.8	4.1
Young's modulus (GPa)					
average E	392	455	422	330	330
standard deviation s	188	107	111	172	118
s/E (%)	48	24	26	52	36

was equal to or higher than 30 rings for each deposition temperature. The results are given in Table 1.

7. Discussion and conclusions

(1) The deposition temperature has a remarkable effect on the properties studied. To secure the quality of TRISO cp, the temperature should be strictly controlled. From the results obtained up to now, the deposition temperature of SiC for TRISO cp should be kept in the range 1500 to 1550°C.

(2) The measured data of strength and Young's modulus conform quite well with SEM observations of the surface morphology and the fracture surface structure of SiC. The rougher the surface, the lower the strength and Young's modulus; the more homogeneous the fracture surface structure, the higher the strength and Young's modulus. The measured results are comparable with the data published in the literature [11–13].

(3) The reason for the disconformity of the measured density with the measured strength, Young's modulus and SEM observations is yet unclear, an explanation could be that the measured density is the true density instead of the apparent density. Should this be the case, the density value measured by the titration method has to be very carefully evaluated, for it is the key property of the SiC coating layer.

Acknowledgments

Prof. Nickel of IWE/KFA, Germany is sincerely appreciated for his kindness of presenting the 2″ fluidized bed and the kernels for this study to our institute. The International Atomic Energy Agency is very much acknowledged for financial support. The authors would like to take the opportunity to thank Ms. Jiang Xinli of Scientific Instrument Corporation, Chinese Academy of Science, for her help in SEM observation. We also would like to thank colleagues of the New Materials Division, INET, who helped in measuring the properties.

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