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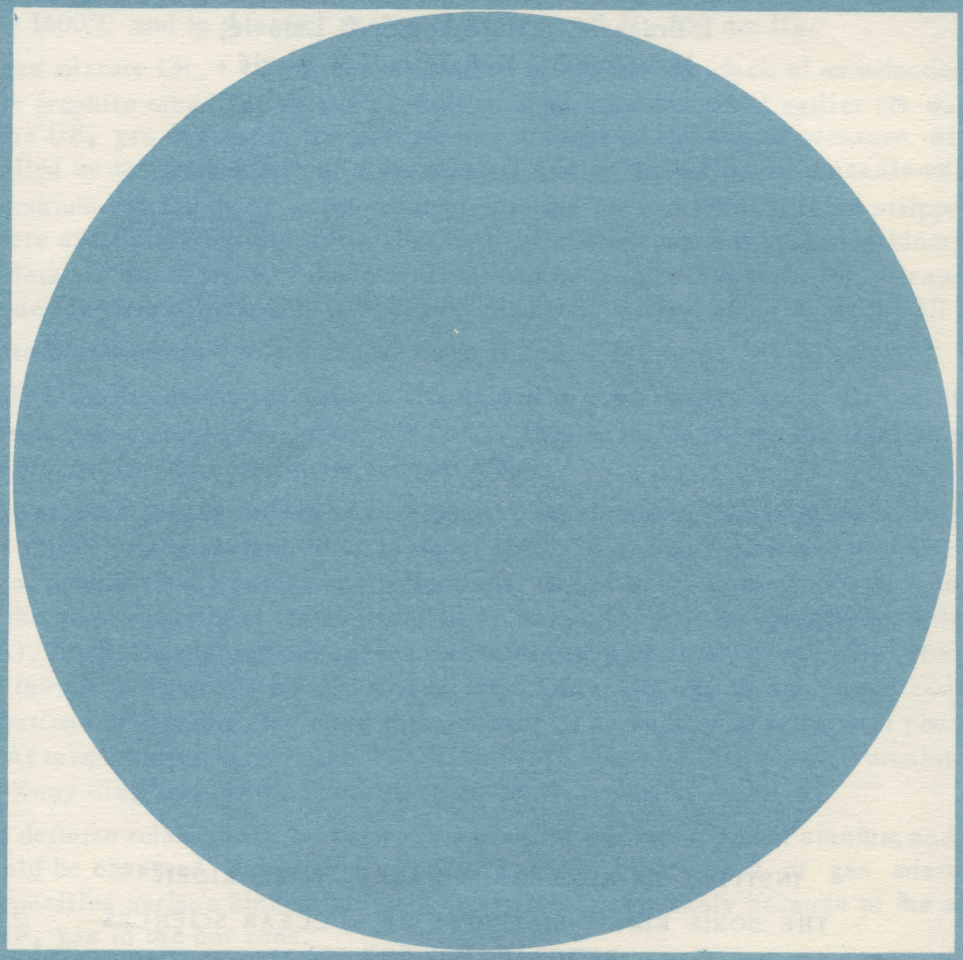

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SIMULTANEOUS VAPOUR DEPOSITION OF URANIUM AND CARBON

Research note by
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Laboratory of Reactor Materials

During recent years simultaneous vapour deposition of carbon and various metals has been the subject of a number of papers (c.f.ref. 1). However, there are apparently no published data concerning simultaneous deposition of uranium and carbon.

The purpose of this note is to present some recent data on the simultaneous pyrolytic deposition from a gas mixture with different proportions of UF_6 and CH_4 at temperatures ranging from 1400 to 1600°C and in the total gas pressure interval 50-150 mm Hg.

The gas mixture $CH_4 + UF_6$ was decomposed at the inner surface of an induction-heated polycrystalline graphite tube. The vapour deposition apparatus described earlier (2) was properly modified. The UF_6 gas content in the mixture was defined by the vapour pressure of the solid UF_6 (controlled by the temperature of a thermostat) and by the orifice of a needle-valve.

The uranium-containing pyrocarbon samples in the form of thin platelets stripped from the substrate were examined by X-ray diffraction, optical microscopy and oxidation kinetics measurements. A detailed description of these methods can be found elsewhere (3). Uranium content was determined in each specimen by a spectrophotometric method using Arsenazo III reagent (4).

Results presented in Table 1 can be summarized as follows:

1. Only a small quantity of uranium is retained in the deposits under the conditions studied, the highest uranium content being about 3% (w/o). This might be due to the relative stability of UF_6, which results in a low rate of its decomposition.

2. The highest concentrations of incorporated uranium were found in the samples deposited at about 1500°C. At temperatures up to about 1500°C the incorporation of uranium causes an appearance of double (00 ℓ) pyrocarbon reflections, one of which corresponds to a normal pyrocarbon and the other (usually of smaller intensity) to an almost ideal graphite (interlayer spacing 3.35-3.36 Å). In specimens, containing low concentrations of uranium, the only effect of uranium presence is the doubling of (00 ℓ) reflections. The X-ray diagrams of specimens containing higher concentrations of uranium also show the presence of an additional tetragonal phase identified as UC_2. At temperatures higher than 1500°C the only effect of incorporated uranium, as revealed by the X-ray diagrams, is the appearance of the UC_2 phase.

3. No definite relationship between the amount of the incorporated uranium and the total gas pressure could be observed. However, it appears that the high speed of gas mixture passage along the deposition surface hinders uranium deposition, presumably because of the short residence time of UF_6 gas in the hot zone.

4. The results of oxidation kinetics measurements, presented in Table 1, indicate two main points:

Table I

Deposition temperature °C	Total pressure mm Hg	Flow rates ml/hr		Uranium content in the deposit, wt%	X-ray results		Oxidation kinetics results			
		CH_4	UF_6		(00 ℓ) reflections*	UC_2 reflections	$k_1 \cdot 10^4$ min^{-1}	A_1, %	$k_3 \cdot 10^4$ min^{-1}	A_3, %
1420	100	1000	43	0.2	slightly deformed d = 3.410; L_c = 81	no				
1420	100	1000	230	0.3	double d_1 = 3.420; d_2 = 3.361	no				
1520	100	1000	30	2.7	double d_1 = 3.412; d_2 = 3.363	101+002; 110; 112, 103; 211+202	2.2	59	1840	41
1560	100	1000	13	0.2	regular d = 3.412; L_c = 140	no				
1580	100	1000	770	0.2	regular d = 3.413; L_c = 134	barely visible				
1600	100	1000	180	0.4	regular d = 3.412; L_c = 137	101+002 medium	1.4	86	1180	14
1640	100	1000	27	0.1	regular d = 3.408; L_c = 160	barely visible				
1520	100	2000	530	0.2	regular d = 3.411; L_c = 122	no				
1540	50	1000	75	0.2	regular d = 3.415; L_c = 103	weak				
1520	150	1000	25	0.1	double d = 3.408; d_2 = 3.35	no	1.1	79	1120	21
1520	150	1000	156	0.3	regular d = 3.410; L_c = 145	weak				

* d and L_c refer, respectively, to the interlayers spacings and crystallite heights as deduced from (00 ℓ) reflections (in Å).

– Samples with normal (00 ℓ) reflections, but containing the UC_2 phase, exhibit similar oxidation behaviour as pure pyrocarbon samples (3). They contain two constituents - a "nongraphitic" one and a small amount of a "graphitic" one - differing in their rate constants by a factor of about 1000.

– Samples exhibiting double (00 ℓ) reflections, where uranium apparently acts as a "catalyst" for graphitization, differ from pure pyrocarbon samples. The proportion of "graphitic" constituent A_3, and the corresponding oxidation rate constant k_3 are considerably higher in the former samples.

5. The microstructural examination of uranium-containing samples did not show any significant difference compared with pure pyrocarbon samples.

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