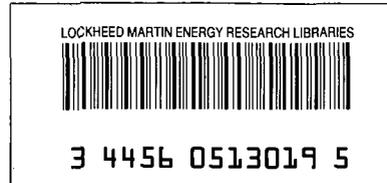


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ORNL-TM-2199

DATE - April 10, 1968

## OXIDATION OF PYROLYTIC-CARBON-COATED FUEL PARTICLES BY LOW CONCENTRATIONS OF WATER VAPOR

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### ABSTRACT

The oxidation of various batches of pyrolytic-carbon-coated fuel particles by water vapor was studied at 1100 to 1400°C using flowing helium-water vapor mixtures having water vapor concentrations of 250 to 1000 ppm and a total pressure of 1 atm. Rates of reaction of water vapor with pyrolytic carbon coatings were determined from continuously recorded weight changes and from analyses of effluent gases for reaction products. Percentage failure of coatings was determined from the quantity of uranium and/or thorium leached by acid from the oxidized coated fuel particles.

Differences in oxidation rates were observed for the various batches of coated particles which could not be correlated satisfactorily with properties of the coatings such as density, anisotropy, and crystallite size. Percentages of failed coatings at constant burnoff also varied from batch to batch. Limited data obtained from exposure of silicon carbide coatings and fuel sticks to water vapor are included.

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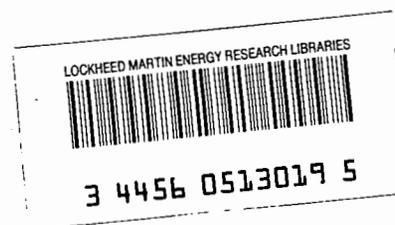
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## CONTENTS

	Page
Abstract . . . . .	1
Introduction . . . . .	4
Experimental . . . . .	9
Materials . . . . .	9
Procedures . . . . .	13
Results and Discussion . . . . .	14
Preliminary Experiments Using a Mullite Reaction Tube . . . . .	14
Rates of Reaction of Coatings with Water Vapor . . . . .	15
Failure of Coatings on Fuel Particles . . . . .	21
Oxidation of SiC-Coated Fuel Particles . . . . .	22
Reaction of Fuel Sticks with Water Vapor . . . . .	23
Acknowledgments . . . . .	24
Bibliography . . . . .	25
Figures . . . . .	28
Appendices . . . . .	
Appendix A. Additional Oxidation Data . . . . .	
Appendix B. Plots of Reaction Rates as a Function of Burnoff . . . . .	



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## INTRODUCTION

The past several years have witnessed rapid progress in the design and development of high temperature gas-cooled reactors which largely has been due to the application of all-ceramic cores. These reactor systems, in which graphite is employed as moderator as well as for the fuel element structure, yield higher thermal efficiencies than can be achieved with metal clad fuel elements.

Recent developments in the science and technology of coated particle fuels have assisted materially in the successful development of all-ceramic reactor cores. For instance, low permeability graphite and special purge systems are not included in the more recent designs for reactors because of satisfactory retention of fission products by pyrolytic carbon coated fuel particles at high burnup. Coated particle fuels currently are so attractive that all of operating and projected high-temperature gas-cooled reactors either use coated particle fuel or plan to do so. These reactors, which are all graphite-moderated and helium-cooled have or expect to have coated particles dispersed in a graphite matrix or as bonded beds of coated particle fuel in graphite structures. The physical characteristics of the coated particles as well as the geometry of the graphite fuel elements being used or planned for use in the various reactors differ considerably. Some of the fuel characteristics for a number of reactors are described briefly.

Dragon Reactor.<sup>1,2</sup> The annular fuel compacts are prepared by warm pressing a mixture containing graphitized coke, coated particles, and phenol formaldehyde resin followed by heat treatment to 1800°C. The fissile particles originally had (Zr,U)C cores coated with interrupted laminar pyrolytic carbon ~ 100 μ thick. Subsequent fuel charges contain coated particles with cores of UC plus excess carbon and have a three layer coating consisting of a silicon carbide layer between two layers of pyrolytic carbon giving a total thickness of ~ 175 μ. The fertile particles have (Th,U)C<sub>2</sub> cores with three layer coatings of the type mentioned above. Since this is an experimental reactor various types of cores and coatings are to be used in future fuel loadings.

Peach Bottom HTGR.<sup>3,4</sup> The annular fuel compacts are prepared by hot pressing at 750°C a mixture of coated particles, graphite, and pitch binder and then heat treating at 1400°C. The cores contain (Th,U)C<sub>2</sub> having a Th to U ratio of 5 for the fissile particles and 21 for the fertile particles. Monolithic pyrolytic carbon coatings of ~ 55 μ thickness are present on all coated particles.

Public Service of Colorado (PSC) HTGR.<sup>5,6</sup> Current plans indicate that coated fuel particles will be present in channels. The coated fuel particles are to be bonded by a mixture of resin and charcoal to keep them immobile. The fertile particles containing ThC<sub>2</sub> cores and the fissile particles having (Th,U)C<sub>2</sub> cores (Th: U of 4:1) are to be coated with a BISO type of coating. This consists of an inner layer of low density pyrolytic carbon and an outer layer of high density isotropic pyrolytic carbon giving a total coating thickness of ~ 100 μ in the case of the fissile particles and ~ 125 μ for the fertile particles.

Arbeitsgemeinschaft Versuchs Reaktor (AVR).<sup>7,8,9</sup> The spherical fuel bodies (60 mm O.D.) are prepared by injecting a mixture of coated fuel particles, binder, and graphite

filler into a hollow graphite sphere and finally baking at  $1450^{\circ}\text{C}$ . The  $(\text{Th},\text{U})\text{C}_2$  cores (Th:U of 5:1) have a duplex type of coating  $\sim 100 \mu$  thick consisting of an inner layer of laminar pyrolytic carbon and an outer layer of columnar pyrolytic carbon.

Closely related to the AVR is the Thorium Hochtemperatur Reaktor (THTR).<sup>10</sup> Present designs call for a spherical fuel body similar to that used in the AVR, but it is expected that the Th to U ratio will be different and that changes in the coatings, including use of silicon carbide, will occur.

Ultra High Temperature Reactor Experiment (UHTREX).<sup>3,11,12</sup> The hollow cylindrical fuel bodies are prepared by extruding a mixture of coated fuel particles, carbon black, graphite flour, and binder, curing, and baking at  $1700^{\circ}\text{C}$ . The  $\text{UC}_2$  cores have a triplex type of coating  $\sim 100\mu$  thick. This consists of an inner buffer layer of pyrolytic carbon, a middle layer of isotropic pyrolytic carbon and an outer layer of granular pyrolytic carbon.

During normal operation, the concentration of contaminants in the helium are maintained at very low levels by passing some small fraction of the coolant through a purification system. The oxidation of the graphite structures and fuel elements by the low concentrations of oxidants ( $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{O}_2$ ) present in the helium, as well as carbon deposition from carbon monoxide, is expected to be tolerable during the life time of the core under such conditions. The fuel and most of the fission products are retained within the coated particles although a portion of some fission products (barium and strontium for examples) will diffuse through the pyrolytic carbon coating during prolonged operation at high fuel temperatures. Failure of the coatings will cause a rapid release of volatile fission products such as xenon and krypton which have been contained in the particles and slower release of

other less volatile fission products. An ingress of steam into the coolant from a damaged heat exchanger probably is the most likely event which would cause failure of a large number of coatings. It is anticipated that if the coated particles are supported by a carbonaceous matrix or contained within graphite that these materials would afford some degree of protection to the coatings against oxidation by steam. In an extreme case, where an appreciable steam pressure remains in the hot core for a prolonged period of time, extensive damage would occur to the graphite structures and fuel bodies and probably less severe but significant damage to the coatings on the fuel particles.

In an early study<sup>13</sup> of the oxidation of pyrolytic-carbon coated fuel particles by steam, weight losses were determined in the temperature range of 700 to 1100°C using partial pressures of steam ranging from 20 to 635 torr. All of the coated fuel particles studied had a laminar type of pyrolytic-carbon coating which cracked and peeled during oxidation by steam. The coatings also appeared to be oxidized by different mechanisms, one type involved a pitting attack of a local nature, the other a more general erosion type involving larger areas of the coatings. In another study,<sup>14</sup> spherical fuel bodies with a graphite matrix supporting pyrolytic-carbon-coated fuel particles were oxidized at temperatures of 800 to 1000°C by a steam pressure of 730 torr. Although the fuel bodies were severely oxidized, in some instances, no evidence of damage to the coatings of the fuel particles was observed. Unsupported fuel particles were severely damaged at comparable exposure to steam indicating that the graphite matrix afforded protection to the coated fuel particles.

During the period in which these earlier studies<sup>13,14</sup> were performed, the types of coated fuel particles available were quite limited and as a consequence coated fuel particles

with a laminar type of coating were mainly employed because of their availability. Intensive efforts of a large number of investigators<sup>15-20</sup> has resulted in remarkable progress in the science and technology of coated particle fuels during the past few years. As a result, numerous types of coated fuel particles have been made available and properties of the coatings such as structure, density, anisotropy, and crystallite size have been correlated with experimental conditions employed during deposition of the pyrolytic-carbon.

Blood and Overholser<sup>21</sup> determined rates of oxidation of a number of different types of pyrolytic carbon coatings at 1000°C using steam pressures of 5, 45, and 520 torr. Some marked differences in reaction rates were found for the various coated fuel particles. Incidence of failure of the coatings at various burnoffs also was examined. The effects of various containers, used to hold the loose coated fuel particles during oxidation, on the reaction rates were studied. Alumina and platinum containers gave comparable reaction rates and as anticipated graphite containers caused a decrease in the oxidation rate of the coatings compared to the other containers. The degree of protection for the coatings provided by graphite was highly dependent on the configuration of the graphite container, however.

Burnette et al.<sup>22</sup> studied the oxidation of pyrolytic-carbon-coated fuel particles at temperatures of 1000 to 1400°C using partial pressures of steam of 0.025 and 0.22 atm. They employed irradiated and unirradiated coated fuel particles having a single type of coating structure (BISO). Oxidation rates and particle failure times were determined for unsupported particles and for particles enclosed in a sealed graphite capsule. Results of this study show that unsupported coated fuel particles failed rapidly when exposed to steam at temperatures of 1300 to 1400°C, but were

resistant to steam attack at temperatures of 900 to 1000°C. Coated particles contained in graphite capsules were protected from oxidation by steam at temperatures above 1000°C, whereas no protective effect was noted at 1000°C and at lower temperatures. The protective action of the graphite is attributed to depletion of steam as it diffuses through the graphite container and a buildup of reaction products (H<sub>2</sub> and CO) which retard the steam-carbon reaction.

None of these earlier studies employed very low partial pressures of water vapor. If the fuel element design provides for containment of the coated fuel particles by a graphite structure and/or support of the particles by a carbonaceous matrix, the concentration of water vapor reaching the coatings on the particles would be expected to be low due to depletion during passage through a portion of the fuel element before reaching the coated particles. Studies presented in this report were performed at low concentrations of water vapor to establish reaction rates for the pyrolytic carbon coatings with water vapor at temperatures comparable to those that might exist in an operating fuel element. A number of batches of coated fuel particles prepared under various coating conditions were examined in an attempt to correlate reaction rates of the coated particles with water vapor with physical properties of the coatings.

## EXPERIMENTAL

### Materials

Various batches of pyrolytic-carbon-coated fuel particles prepared under different coating conditions, and as a result having different physical properties, were employed. Relevant information for the various batches of coated particles is given in Tables 1 and 2 and additional structural detail may be seen in the photomicrographs (Figs. 6-13).

Table 1. Deposition Conditions Used for Preparation  
of Various Batches of Coated Fuel Particles

Batch Designation	Deposition Temperature <sup>a</sup> (°C)	Hydrocarbon Decomposed <sup>a</sup>	Average Deposition Rate <sup>a</sup> (μ/min)	Fuel Particle	
				Composition	Diameter (μ)
Granular 4 <sup>b</sup>	1730	Methane	0.2	(Th,U)C <sub>2</sub>	450
Isotropic 5 <sup>b</sup>	1650	Methane	1.5	(Th,U)C <sub>2</sub>	450
Isotropic 6 <sup>b</sup>	2000	Methane	0.4	(Th,U)C <sub>2</sub>	385
Isotropic 7 <sup>b</sup>	1800	Methane	0.4	(Th,U)C <sub>2</sub>	385
OR-688 <sup>c</sup>	1250	Propylene	6.7	ThO <sub>2</sub>	230
OR-689 <sup>c</sup>	1250	Propylene	7.0	ThO <sub>2</sub>	230
YZ-134 <sup>c</sup>	2000	Methane	1.5	ThO <sub>2</sub>	275
YZ-135 <sup>c</sup>	2000	Methane	2.9	ThO <sub>2</sub>	275
YZ-136 <sup>c</sup>	2000	Methane	0.7	ThO <sub>2</sub>	275
OR-788 <sup>c</sup>	2000	Methane	1.7	UC <sub>2</sub>	210
OR-789 <sup>c</sup>	1250	Propylene	7.0	UC <sub>2</sub>	210
OR-790 <sup>c</sup>	1600	Methane	4.4	UC <sub>2</sub>	210

Table 1 (continued)

Batch Designation	Deposition Temperature <sup>a</sup> (°C)	Hydrocarbon Decomposed <sup>a</sup>	Average Deposition Rate <sup>a</sup> (μ/min)	Fuel Particle	
				Composition	Diameter (μ)
OR-813-R <sup>c</sup>	2000	Methane	1.8	UO <sub>2</sub>	235
OR-814-R <sup>c</sup>	1250	Propylene	8.0	UO <sub>2</sub>	235
OR-815 <sup>c</sup>	1600	Methane	5.0	UO <sub>2</sub>	235
OR-818 <sup>c</sup>	1250	Propylene	9.0	UO <sub>2</sub>	235

a. Deposition conditions for outer coating; most batches have inner layer 25-40 μ thick of low density carbon deposited from acetylene at 1050°C. Batch OR-813-R has intermediate coating deposited from propylene at 1250°C and Batch OR-818 has SiC layer next to outer layer.

b. Supplied by General Atomic Division, General Dynamics Corporation. Further details given in Ref. 20.

c. Supplied by Metals and Ceramics Division, Oak Ridge National Laboratory. Further details given in Ref. 23 and 24.

Table 2. Properties of Various Batches of  
Pyrolytic-Carbon Coated Fuel Particles

Batch Designation	Total Diameter ( $\mu$ )	Total Coating Thickness ( $\mu$ )	Pyrolytic Carbon Coating (wt %)	Coating Density <sup>a</sup> (g/cm <sup>3</sup> )	Surface Area <sup>b</sup> (m <sup>2</sup> /g)	Bacon Anisotropy Factor	Apparent Crystallite Size, L <sub>c</sub> (Å)
Granular 4	825	190	53.9	2.01	0.028	1.06	139
Isotropic 5	825	190	49.0	1.55	0.024	1.00	49
Isotropic 6	760	190	65.2	2.00	0.028	1.30	145
Isotropic 7	830	225	73.3	1.82	0.062	1.05	105
OR-688	435	105	48.4	1.9	0.070	~1	34
OR-689	435	105	50.0	1.9	-	~1	34
YZ-134	500	115	46.6	1.94	0.029	~1	109
YZ-135	500	115	41.8	1.89	0.064	~1	124
YZ-136	500	115	46.2	2.05	0.022	1.2	122
OR-788	445	115	56.8	2.0	0.072		
OR-789-C	440	115	59.3	2.0	0.130		
OR-790-C	440	110	51.0	1.6	0.224		
OR-813-R	470	115	52.5	1.75	0.152		
OR-814-R	450	110	53.2	1.98	0.110		
OR-815	470	115	47.3	1.54	0.154		
OR-818	490	125	-	1.97	0.131		

<sup>a</sup>Outer coating (see note a Table 1); <sup>b</sup>Based on weight of coated fuel particle.

Some of the properties were obtained from pyrolytic carbon deposited on flat substrates present in the deposition furnace during coating of the fuel particles. It was assumed that the structure of the pyrolytic carbon formed in this manner was representative of that deposited on the fuel particles. Most of the batches of coated particles were used without any pretreatment but in a few runs the coated particles were leached with acid prior to the oxidation tests.

### Procedures

The experimental studies were performed at temperatures of 1100 to 1400°C using helium-water vapor mixtures having water vapor concentrations ranging from 250 to 1000 ppm (parts per million by volume) and a total pressure of 1 atm. The experimental equipment employed is similar to that used in earlier studies.<sup>25</sup> An Ainsworth semimicro recording balance sensitive to 0.02 mg was used to obtain a continuous record of weight changes. A sensitive gas chromatograph (Burrell K-7 with thermionic detector) was used to analyze effluent gases thereby providing a second method of measuring reaction rates. This instrument was particularly useful for detecting very low concentrations of oxygen and nitrogen that might have leaked into the system. A rhodium wound resistance furnace equipped with a programmed power supply was employed along with a high purity alumina reaction tube.

The apparatus is depicted in Fig. 1. Purified helium was mixed with helium, which had passed over saturated calcium chloride at °C, in the proper proportion to give the desired water vapor concentration as measured by a moisture detector. After establishing that the combined helium stream was essentially free of oxygen and with the furnace at the desired temperature, the helium-water vapor mixture was introduced into the reaction chamber, flowing downward past

the coated particles (flow rate of 200 cm<sup>3</sup>/min STP in most cases). Approximately 100 mg of coated particles was contained in a basket made of platinum gauze and this in turn was suspended by a platinum wire from the balance. The effluent gases were analyzed for reaction products by the gas chromatograph and reaction rates obtained from these data and the recorded weight changes.

Incidence of failure of coatings was obtained from the quantity of uranium and/or thorium leached by nitric acid or a mixture of nitric and hydrofluoric acids following exposure of the coated particles to water vapor. Microscopic and metallographic examinations also were made of many of the oxidized coated fuel particles.

## RESULTS AND DISCUSSION

### Preliminary Experiments Using a Mullite Reaction Tube

The first studies of the oxidation of pyrolytic-carbon-coated fuel particles by low concentrations of water vapor were performed in a mullite reaction tube while awaiting delivery of high purity alumina tubes. These runs were made with Isotropic 6 particles at 1100 and 1200°C using water vapor concentrations of 250, 500 and 1000 ppm at flow rates of 200 and 400 cm<sup>3</sup>/min STP. The experimental data obtained were so variable that they have no significance. In virtually all cases the oxidation rates decreased with time and in some instances the final rates were an order of magnitude less than the initial rates. Although runs were made at different temperatures, flow rates, and water vapor concentrations, the rates were so inconsistent that it was impossible to determine the effects of these parameters on the reaction rates.

Geary and Littlewood<sup>26</sup> observed, during studies of the graphite-steam reaction in a closed mullite tube that specimens degassed at 1100°C and higher temperatures subsequently

gave reaction rates which decreased with time. Subsequent studies in an alumina reaction chamber led them to postulate that the silica in the mullite catalyzed the steam-graphite reaction but that this catalytic effect decreased with time. The rates found in mullite, although they decreased with time, were higher than those found in the alumina system at comparable temperatures and water vapor pressures. Our results seem to be in general agreement with these observations. No decreases in reaction rates were found when a high purity alumina reaction tube was used in place of mullite. Furthermore, the reaction rates obtained in alumina tubes are lower than the lowest rates found in mullite under comparable exposure conditions. All available evidence suggests that the silica present in mullite was responsible for the erratic data obtained in these early runs.

This conclusion is a bit disturbing since there is contradictory evidence available. For instance, Burnette et al.<sup>22</sup> used mullite in this temperature range with no difficulty and report that containment of coated particles in a silica boat gave lower reaction rates than obtained with a platinum holder. The principal difference between their experimental conditions and ours is the much lower water vapor pressure used in our studies.

#### Rates of Reaction of Coatings with Water Vapor

There is no entirely satisfactory way of expressing the rates of reaction for the various coatings. Roughness factors for various coatings were found to vary from about 8 to 20 in earlier work<sup>21</sup> casting doubt on the validity of using geometric surface areas as a basis for expressing reaction rates. The weight of coating remaining at any particular time was more or less arbitrarily chosen as the basis for calculating reaction rates recognizing the fact that the

coatings in some cases consist of two or more layers of pyrolytic carbon having different structures. For sake of comparison, reaction rates obtained at 5% or less burnoff were selected. Rates, so obtained, do not vary a great deal from those calculated on the basis of the original weight of pyrocarbon except at high burnoffs where the deviation becomes important.

The effects of temperature on the oxidation rates of various coatings obtained at a water vapor concentration of 1000 ppm are shown in Fig. 2. Rates given for various coatings at the same temperature and temperature coefficients found for different coatings vary considerably. Apparent activation energies of about 30 to 65 kcal/mole were found which is a fairly wide range. Burnette et al.<sup>22</sup> report a value of 63 kcal/mole in the temperature range of 1000 to 1400°C at a partial pressure of steam of 0.025 atm. Values ranging from 40 to 70 kcal/mole have been reported by various investigators<sup>25,27,28,29</sup> for the reaction of graphite with low partial pressures of water vapor. Differences in geometry of graphite specimens probably are responsible for part of the variations.

Reaction rates measured for two batches of coated particles at 1300°C using various concentrations of water vapor are shown in Fig. 3. The apparent order of the reaction with respect to water vapor concentration ranges from about 0.5 to 1 with an increase with increasing water vapor concentration evident. A similar behavior which was observed previously<sup>21</sup> at higher water vapor concentrations was thought to be due to coating failures and subsequent catalysis of the reaction by the exposed fuel particles. This does not explain the behavior of Isotropic 6 particles because the fraction failing was essentially the same for all water vapor concentrations. Burnette et al.<sup>22</sup> reported an apparent order of 0.7 but used only two concentrations. Various investigators<sup>25,28,29,30</sup>

have reported reaction orders in the range 0.5 to 1 for the reaction of graphite with water vapor using low and relatively high ranges of concentrations. No satisfactory explanation can be given for the differences in either the apparent orders of reaction or activation energies observed in these studies.

Data obtained for the various batches of coated particles are presented in Table 3, Figs. 4 and 5, and in Appendices A and B. Reaction rates presented in Table 3 and weight losses shown in Figs. 4 and 5 demonstrate the variability of the various batches when exposed to water vapor. In the early studies it was believed that the higher rates found for Isotropic 5 than for Isotropic 6 coated particles were due to the lower coating density of the former. Subsequent studies with YZ-134 and OR-689 coated particles showed that variables other than density must be important. Data from other batches tend to confirm this belief. At most, one can say that there is some tendency for reaction rates to increase with decreasing coating densities but some notable exceptions are evident. Other coating properties such as anisotropy and crystallite size also have no consistent effect on reaction rates.

Oxidation rates found for coatings prepared from methane and propylene indicate that the hydrocarbon used is not a controlling factor; both high and low rates were found for propylene as well as for methane coatings. Fuel particles used in preparing the coated particles conceivably could have some effect on the oxidation rate of the coating if the degree of contamination varies with the type of fuel particle and the contaminants in the coating catalyze the reaction with water vapor. Limited data available suggest that rates of oxidation of coatings (high density) on  $\text{UO}_2$  particles may be lower than those of coatings on  $\text{UC}_2$  cores. It is not easy to establish, however, that higher concentrations of contaminants

Table 3. Oxidation Rates and Coating Failures  
for Various Batches of Pyrolytic-Carbon Coated  
Fuel Particles (100 mg sample used in all runs)

Batch Designation	Run No.	Temp. (°C)	H <sub>2</sub> O Concentration (ppm)	Flow Rate (cm <sup>3</sup> STP/min)	Reaction Rate <sup>a</sup> (mg g <sup>-1</sup> hr <sup>-1</sup> )	Coating Oxidized (wt %)	Coatings Failed <sup>b</sup> (%)
Granular 4	22 <sup>c</sup>	1100	1000	200	0.3	3.4	1.6
	21 <sup>c</sup>	1200	1000	200	4.4	12	51
	19 <sup>c</sup>	1300	1000	400	16	46	72
Isotropic 5	8 <sup>c</sup>	1100	1000	200	5.0	15	d
	9 <sup>c</sup>	1200	1000	200	15	37	44
	10 <sup>c</sup>	1300	1000	200	21	52	52
	11 <sup>c,e</sup>	1400	1000	200	28	53	87
Isotropic 6	45	1100	1000	200	0.3	0.8	<0.1
	13	1200	1000	200	0.8	3.6	0.2
	29 <sup>c</sup>	1200	1000	200	0.8	3.7	<0.1
	53	1300	250	400	2.9	6.0	<0.1
	17	1300	500	400	3.7	10	<0.1
	16	1300	1000	400	6.5	17	0.1
	14	1300	1000	200	4.9	14	0.1
	15	1400	1000	200	17	45	33
	Isotropic 7	20 <sup>c</sup>	1300	1000	400	7.9	19
YZ-134	50	1100	1000	200	0.9	1.6	0.8
	25	1200	1000	200	18	38	4.4
	28 <sup>c</sup>	1200	1000	200	4.0	12	2.9
YZ-135	23	1200	1000	200	19	48	17
YZ-136	24	1200	1000	200	16	31	16
OR-688	26	1200	1000	200	7.2	21	6.7
OR-689	49	1100	1000	200	3.6	7.0	0.8
	31	1200	250	200	1.1	6.4	<0.1
	30	1200	500	200	1.9	8.3	0.1
	27	1200	1000	200	7.2	19	0.8
	52 <sup>c</sup>	1200	1000	200	10	27	2.4
	34	1300	250	200	5.2	13	0.2
	33	1300	500	200	8.5	24	0.4
	35	1300	1000	200	18	56	3.4

Table 3 (continued)

Batch Designation	Run No.	Temp. (°C)	H <sub>2</sub> O Concentration (ppm)	Flow Rate (cm <sup>3</sup> STP/min)	Reaction Rate <sup>a</sup> (mg g <sup>-1</sup> hr <sup>-1</sup> )	Coating Oxidized (wt %)	Coatings Failed <sup>b</sup> (%)
OR-788	40	1100	1000	200	1.6	4.9	2.1
OR-789-C	37	1100	1000	200	12	40	21
	36	1200	1000	200	23	64	31
OR-790-C	42	1100	1000	200	6.2	16	0.3
OR-813-R	48	1100	1000	200	1.8	3.2	4.0
	44	1200	1000	200	9.0	15	13
	46	1200	1000	200	5.2	10	6.7
OR-814-R	41	1100	1000	200	2.9	5.1	5.4
	43	1200	1000	200	5.5	13	8.6
OR-815	39	1100	1000	200	6.0	15	3.1
	51	1200	1000	200	19	43	17
OR-818 <sup>f</sup>	54	1100	1000	200	4.0	8.0	d
	54	1200	1000	200	14	100	d

<sup>a</sup>Reaction rate based on weight of pyrolytic-carbon coating; rates given are for 5 wt % or less burnoff.

<sup>b</sup>Calculated from quantity of uranium (thorium) in acid leach solution and total quantity of uranium (thorium) originally present in 100 mg of coated fuel particles.

<sup>c</sup>Leached with acid solution prior to exposure to water vapor.

<sup>d</sup>Not determined.

<sup>e</sup>15 hr exposure to water vapor.

<sup>f</sup>Layer of silicon carbide beneath outer layer of pyrolytic-carbon.

occur in coatings laid down on  $UC_2$  cores than on  $UO_2$  cores. A higher bed temperature employed in the coating process might be expected to increase the degree of contamination of the coating and in turn enhance the rate of reaction with water vapor. The oxidation rate given for OR-788 coated particles is significantly lower than that reported for OR-789-C coated particles although the bed temperature used in preparing the latter was  $1250^\circ C$  compared to  $2000^\circ C$  for the former.

Limited studies of the effects of leaching coated particles by acid prior to oxidation were made since it was believed that contamination of the coatings contributed to the variable reaction rates observed. Reaction rates found for unleached and leached Isotropic 6 and OR-689 coated particles show that acid leaching had no important effect on the oxidation rates. Data obtained for YZ-134 coated particles, however, indicate that such leaching caused a significant decrease of the reaction rate. Surface contamination by U reported for a number of the batches of coated particles<sup>24</sup> could not be correlated with reaction rates measured for these coated particles.

Any further study of the effects of coating contamination does not appear attractive since such contamination would be expected to vary from batch to batch and, in addition, a reliable determination of the type and degree of contamination is not a simple matter. Furthermore, it appears unlikely that acid leach of large-scale batches of coated particles would be feasible because of costs and possible damage to coatings. The oxidation rates reported have been obtained for materials which are believed to be representative of current coating technology and include data for types of coated fuel particles that are suitable for commercial application.

### Failure of Coatings on Fuel Particles

Data are included in Table 3 which show the percentage of coatings that failed during exposure to water vapor for the various batches of coated fuel particles. These values are based on the percentage of uranium and/or thorium leached by acid following such exposure. In some instances, the long exposure time combined with high oxidation rates caused extensive oxidation of a large portion of the pyrolytic carbon and, as might be expected, high percentages of coating failures were observed. Substantial percentages of failures also were found at fairly low burnoffs in a number of cases. Data obtained for Isotropic 6 and OR-689 coated particles show low percentages of failures at substantial burnoffs, and suggest that burnoffs up to 10 wt % may occur with few coating failures. Limited data given for OR-790-C coated particles indicate that this batch also is resistant to coating failure. Granular 4, OR-813-R and OR-814-R coated particles, on the other hand, show substantial percentages of failures at burnoffs of approximately 5 wt %.

The tendency of failure of the various coatings may be associated with the type of oxidation that the coatings undergoes, which in turn, could be related to the type and degree of contamination of the coatings. A generalized type of attack could remove a substantial portion of the coating without penetration to the fuel particle, whereas, a localized or pitting type of attack could develop a path to the fuel particle but cause burnoff of only a small portion of the pyrolytic carbon. Photographs and photomicrographs of oxidized coated particles from several batches are presented in Figs. 6-13. Random samples were used for microscopic examinations. The general appearance indicates that oxidation of the coatings occurred through a localized or pitting type of attack. This was the case for essentially all of the coated particles examined.

## Oxidation of SiC-Coated Fuel Particles

Coated fuel particles having combinations of SiC and pyrolytic carbon coatings are being prepared for possible use in fuel elements. The SiC layer will stop movement of certain fission products that ordinarily would migrate to the surface if the coating consists of only pyrolytic carbon. An outer layer of SiC is expected to be stable in the presence of water vapor at high temperatures and if it is would protect the pyrolytic carbon present between the SiC layer and the fuel particle.

Two batches of coated particles having layers of both pyrolytic carbon and SiC were exposed to a water vapor concentration of 1000 ppm in helium at high temperatures to examine the behavior of SiC coatings. OR-818 coated particles have a 58  $\mu$  thick outer layer of pyrolytic carbon deposited over a SiC layer 22  $\mu$  in thickness. Two additional layers of pyrolytic carbon separate the SiC from the UO<sub>2</sub> fuel particle. The other batch (GA-330) has a layer of SiC 34  $\mu$  thick deposited on a 72  $\mu$  thick layer of pyrolytic carbon which covers the UO<sub>2</sub> fuel particle.

The OR-818 coated particles were exposed to water vapor at 1100°C for 22 hr and then at 1200°C for 100 hr. The reaction rates obtained for the outer layer, which are given in Table 3, are similar to rates measured for other coatings indicating that SiC had no important effect on the reaction of water vapor with pyrolytic carbon. After the outer layer had been oxidized, the reaction rate became essentially zero as evidenced by both the weight changes and gas analyses. This indicates that the SiC coating was stable.

The GA-330 coated particles were exposed to water vapor at 1200°C for 22 hr, at 1300°C for 42 hr, and at 1400°C for 22 hr. No detectable weight changes occurred and the gas analyses also showed that there was no measurable reaction

of carbon with water vapor. Thus, it was shown that the SiC coating did not fail during prolonged exposure to water vapor at temperatures up to  $1400^{\circ}\text{C}$ . (An alumina container was used for the coated particles in these studies because of interaction of Pt with SiC at temperatures above  $\sim 1250^{\circ}\text{C}$ .)

#### Reaction of Fuel Sticks with Water Vapor

Data obtained from studies of the reaction of fuel compacts,<sup>31</sup> containing bonded beds of coated fuel particles, with water vapor suggested that the bonding material protected the pyrolytic carbon coatings to some degree; probably because the residual carbon from the binder was more reactive with water vapor than the coatings. In view of these findings, limited studies of fuel sticks were made at  $990^{\circ}\text{C}$  using a water vapor concentration of 1000 ppm in flowing helium in an attempt to establish relative reaction rates for the coatings and residual binder material.

The experimental equipment and techniques used are essentially the same as used for loose coated fuel particles. The fuel sticks were prepared by Metals and Ceramics Division of ORNL using 1002SB  $\text{ThO}_2$  coated particles, prepolymerized furfuryl alcohol resin, and graphite filler. The mixture was molded, cured at  $80^{\circ}\text{C}$ , baked at  $1000^{\circ}\text{C}$ , and finally heat treated at  $1500^{\circ}\text{C}$  in helium. A typical fuel stick is 0.15-in. in diameter, 0.28-in. long, and weighs about 170 mg. It contains about 6 mg of graphite, 13 mg of residual carbon from the resin, and 75 mg of pyrolytic carbon as coatings on the fuel particles.

Rates of total carbon removal as measured by continuously recorded weights and by occasional analysis of effluent gases are given in Fig. 14 for two fuel sticks. These curves show that the rate of oxidation of carbon decreased rapidly during the early part of each run and became nearly constant toward the end. The latter part corresponds to the oxidation

of pyrolytic carbon coatings on the fuel particles, whereas the rates observed during the early part of the run represent, to a large degree, those due to oxidation of residual carbon from the resin. Intermediate rates probably are those of graphite filler with some contributions from the coating and bonding carbons. It appears that the rate of oxidation of the binder carbon is at least an order of magnitude greater than that of the pyrolytic carbon coatings.

Examination of the materials at the end of the runs showed that essentially all of the graphite filler and bonding carbon had been oxidized and approximately 20 wt % of the pyrolytic carbon originally present in the coatings was removed. Acid leach of the coated particles showed that about 3% of the coatings failed during these runs. Evidence of rather severe attack of the coatings is shown in Fig. 15.

#### ACKNOWLEDGMENTS

The authors gratefully acknowledge the assistance of the Analytical Service Group of the Analytical Division by providing metal analyses and of the Metallography Group of Metals and Ceramics Division by performing metallographic examinations of the coated fuel particles.

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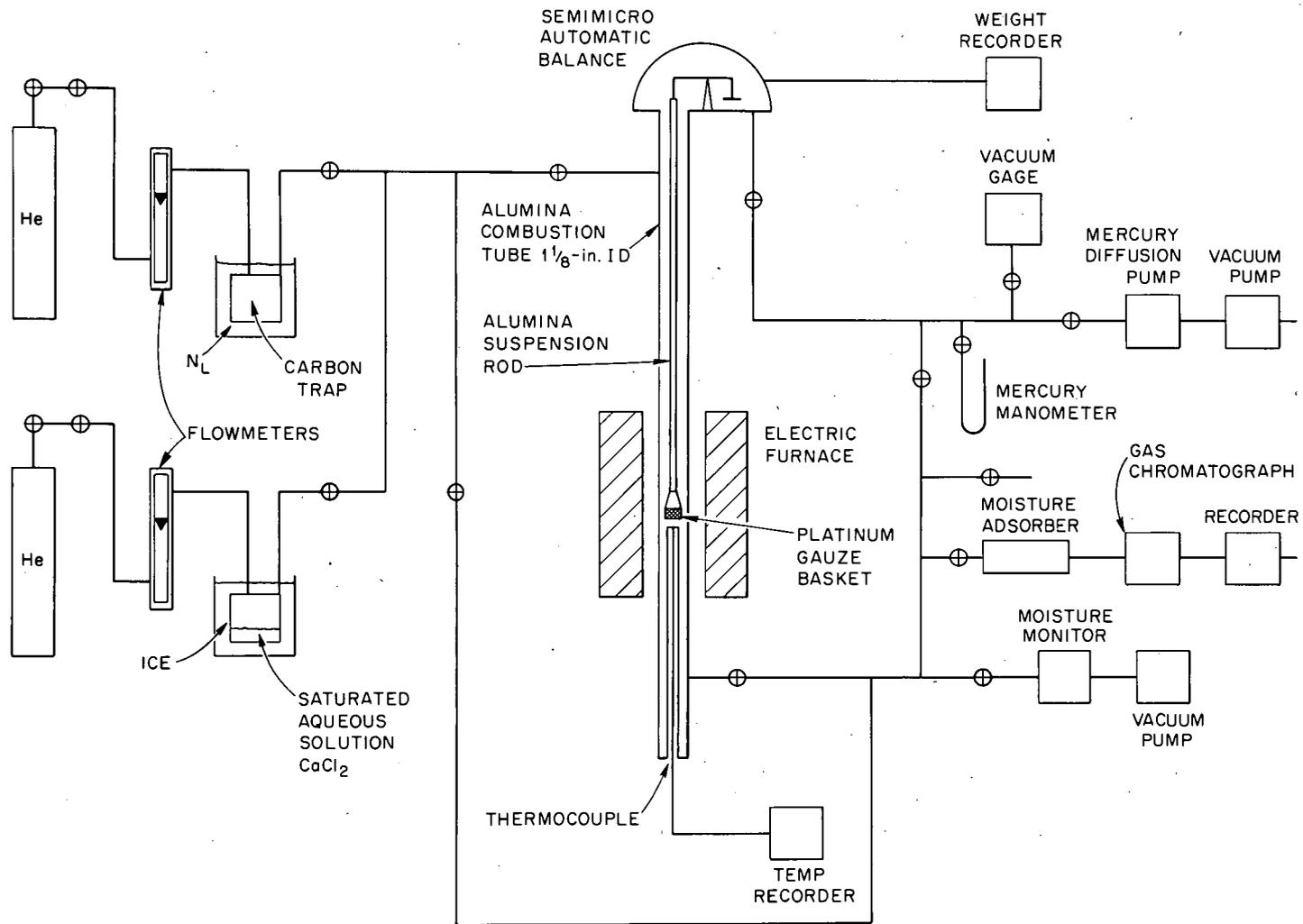


Fig. 1. Diagram of Experimental Apparatus.

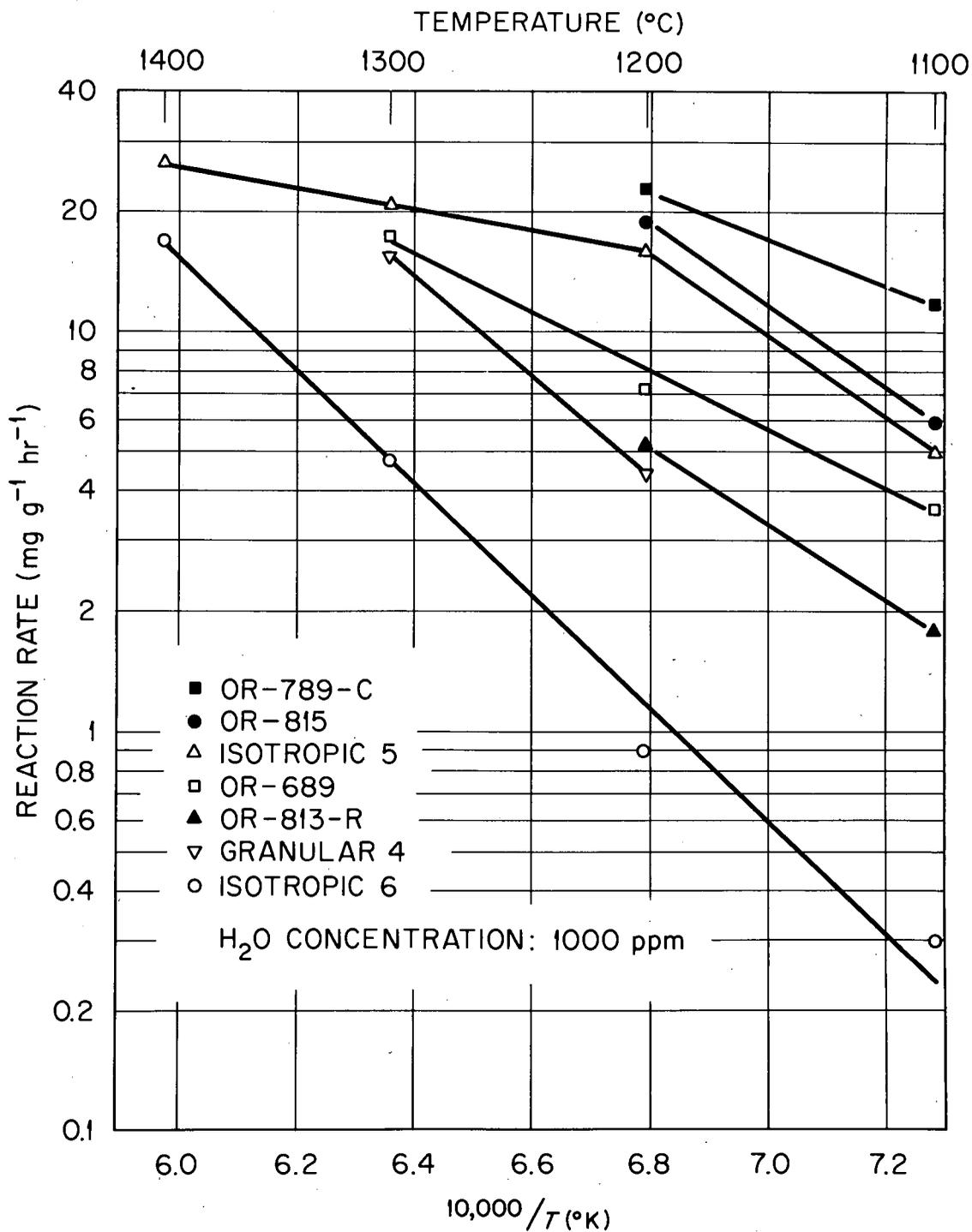


Fig. 2. Effects of Temperature on Reaction Rates of Various Batches of Coated Particles with Water Vapor.

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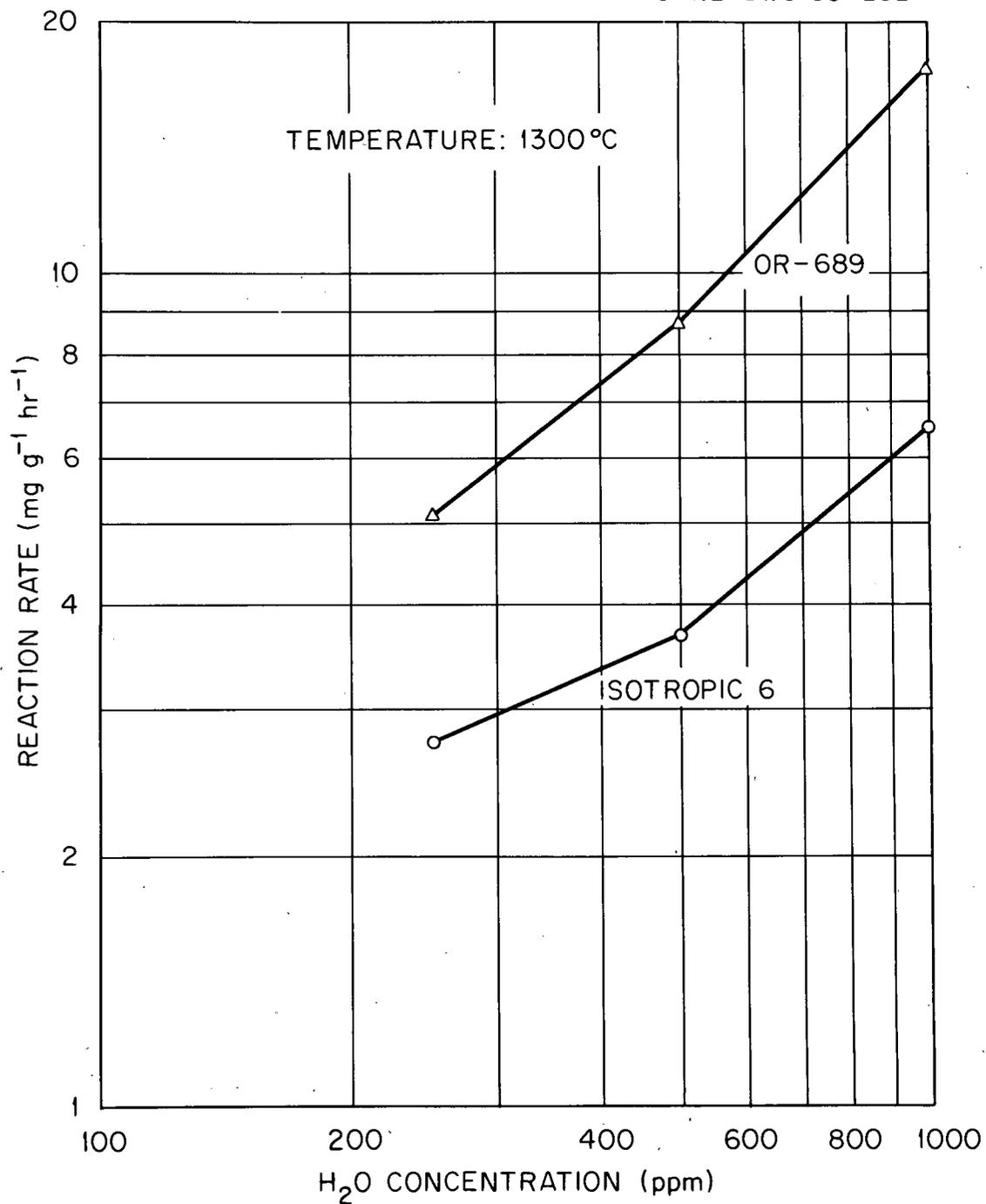


Fig. 3. Reaction Rates of Isotropic 6 and OR-689 Coated Particles at Various Water Vapor Concentrations.

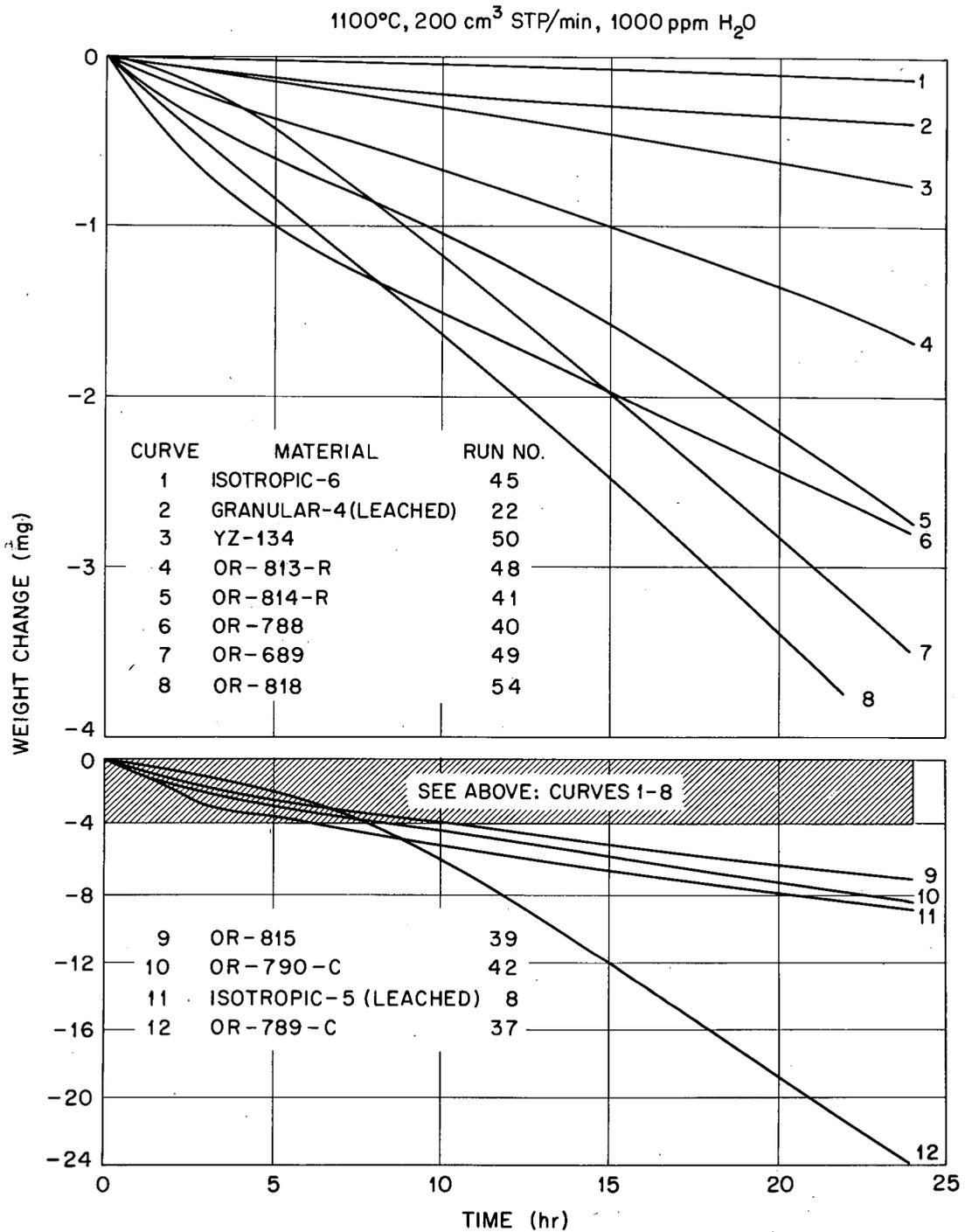


Fig. 4. Weight Losses of Various Batches of Coated Particles upon Exposure to Water Vapor at 1100°C.

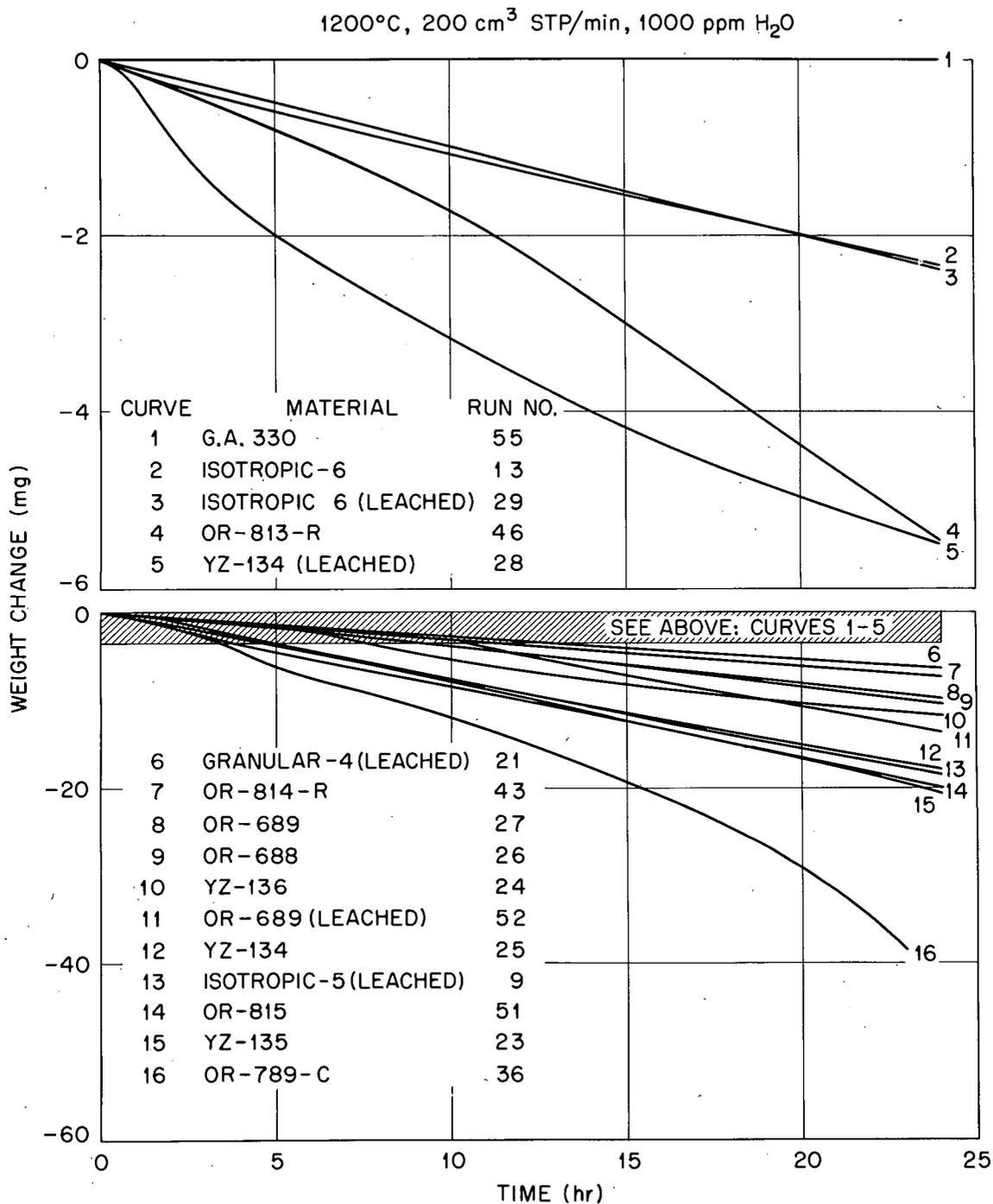


Fig. 5. Weight Losses of Various Batches of Coated Particles upon Exposure to Water Vapor at 1200°C.

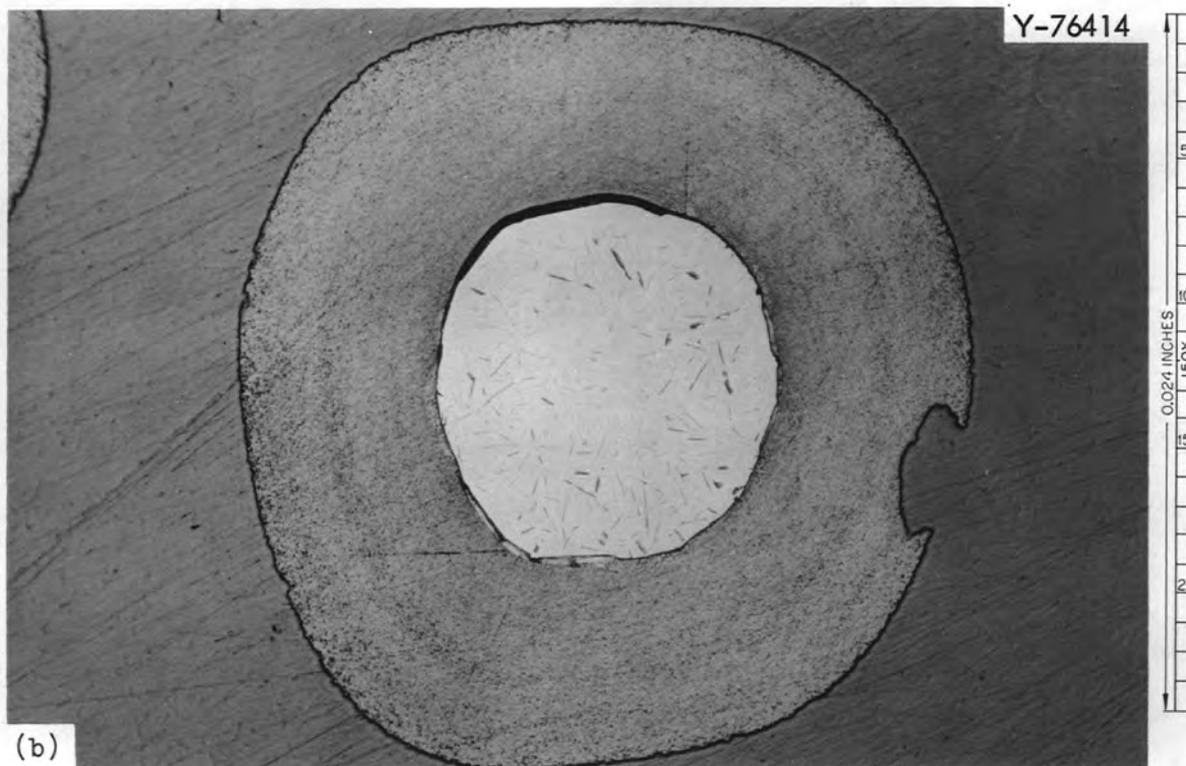
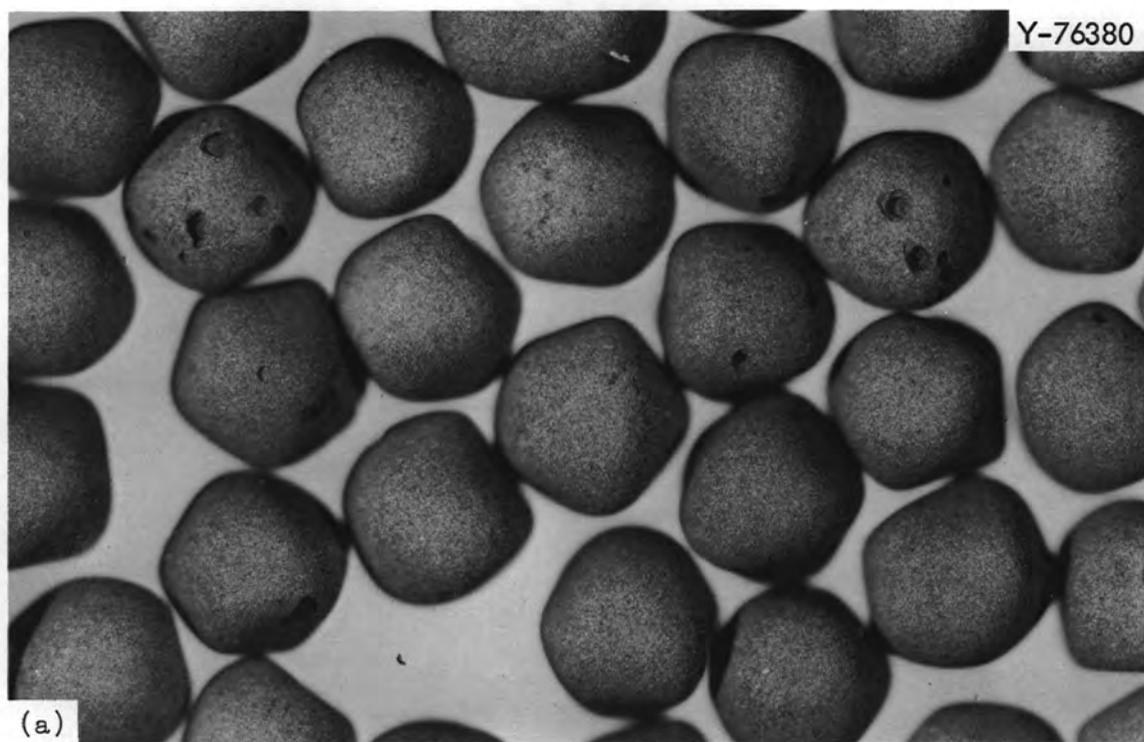


Fig. 6. Leached Isotropic 6 Coated Particles After 24 hr Exposure at  $1200^{\circ}\text{C}$  to Water Vapor Concentration in Helium of 1000 ppm. (a) 33X. (b) Sectioned particle. 150X.

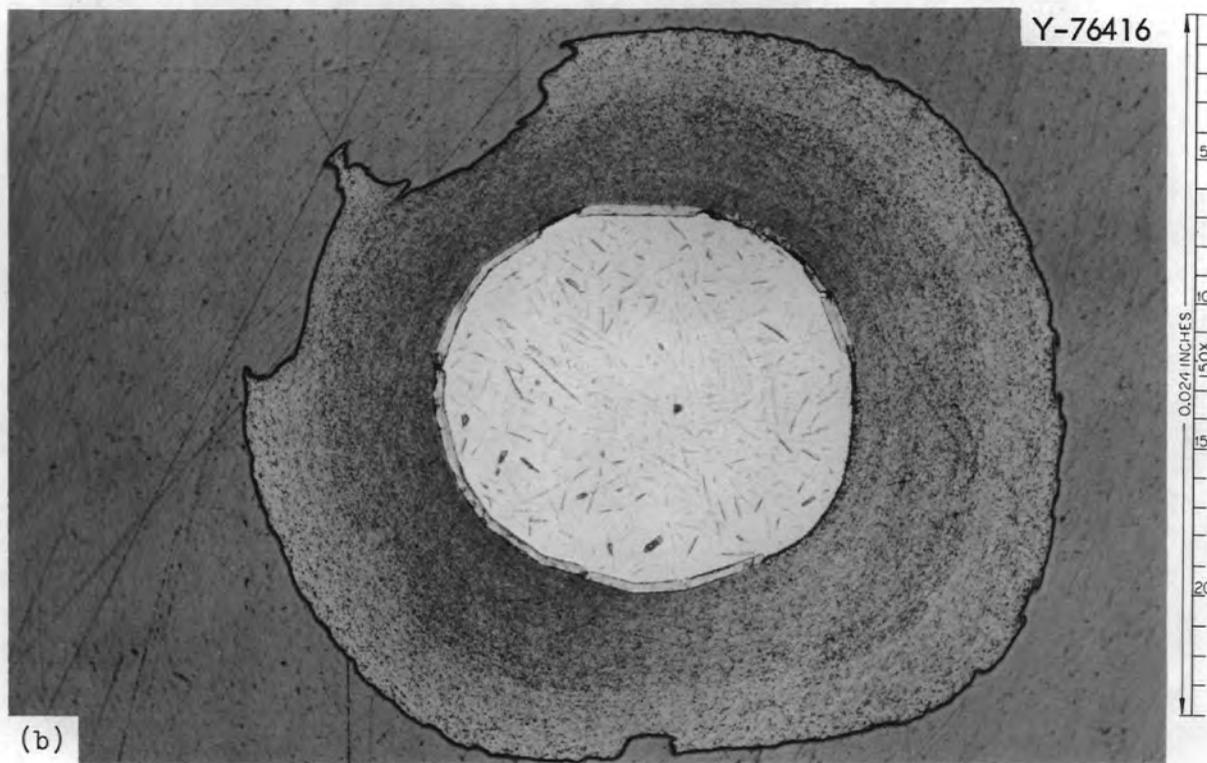
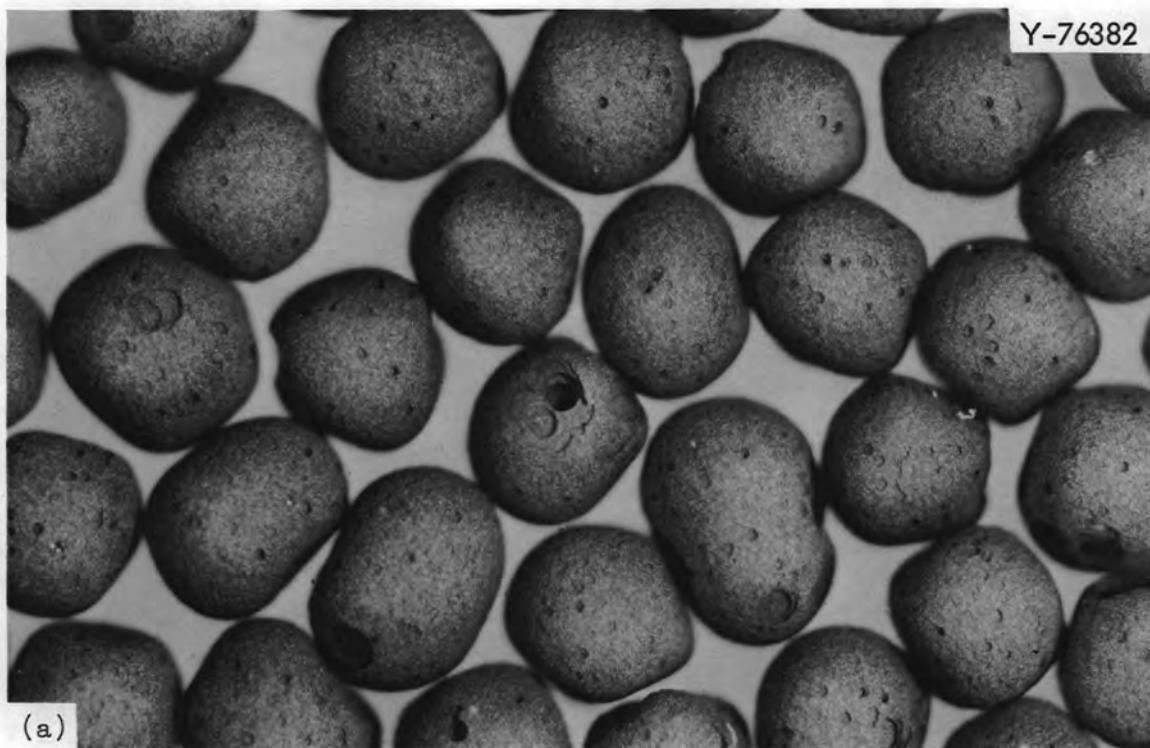


Fig. 7. Leached Isotropic 6 Coated Particles After 24 hr Exposure at  $1300^{\circ}\text{C}$  to Water Vapor Concentration in Helium of 1000 ppm. (a) 33X. (b) Sectioned particle. 150X.

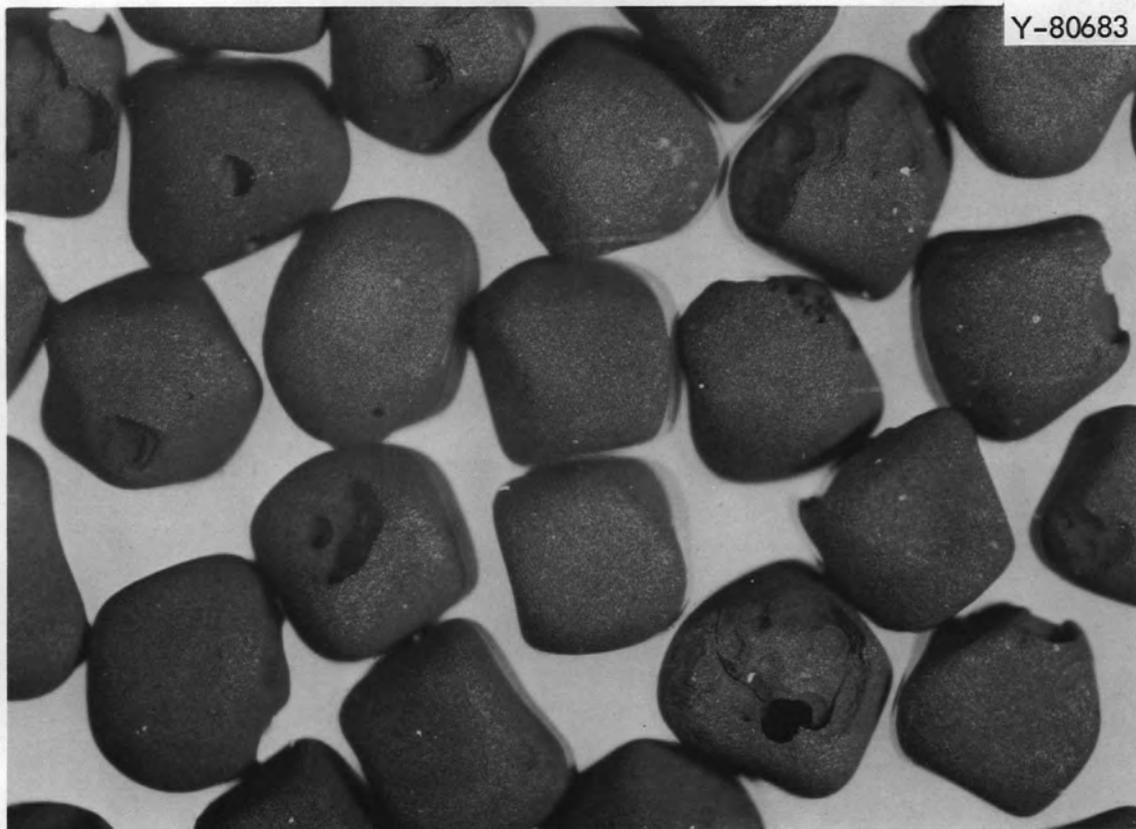


Fig. 8. Leached Isotropic 7 Coated Particles After 24 hr Exposure at 1300°C to Helium with Water Vapor Concentration of 1000 ppm. 33X.

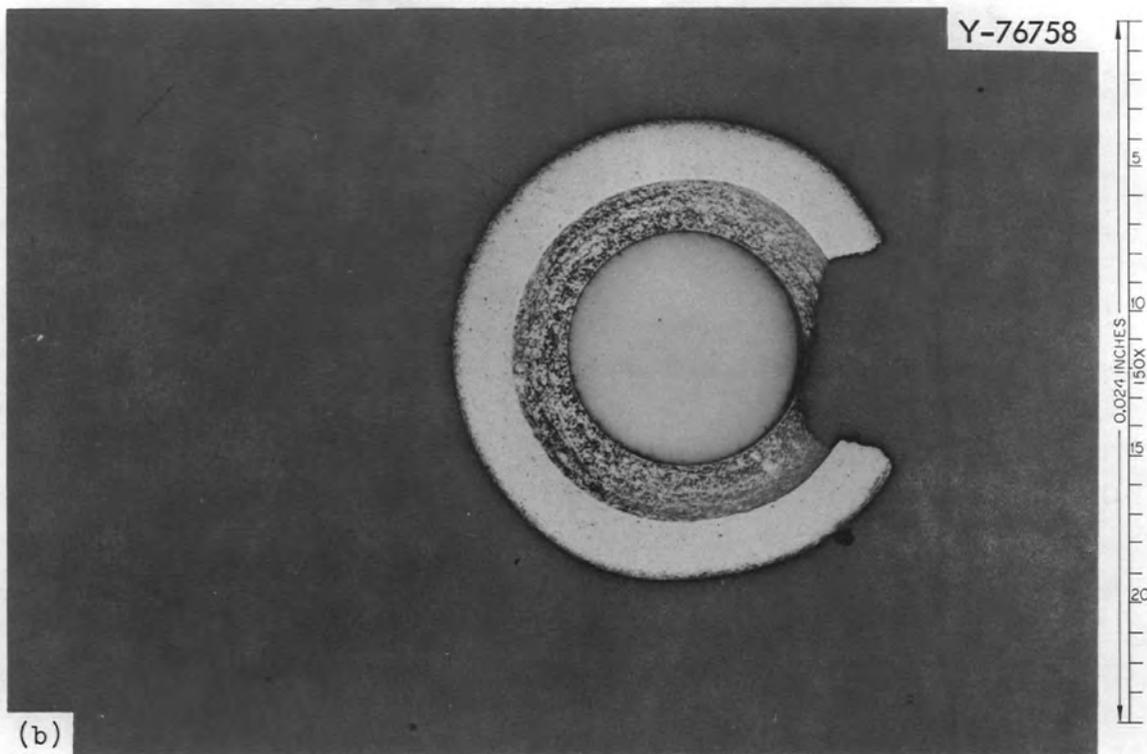
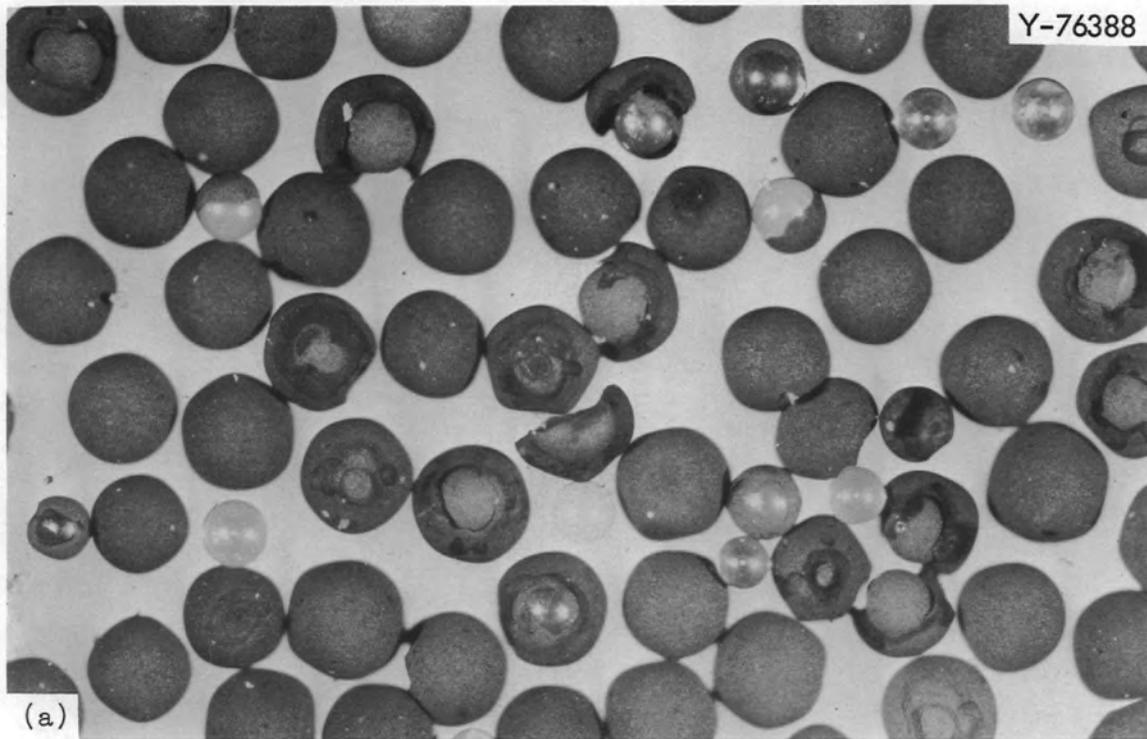


Fig. 9. Unbleached YZ-134 Coated Particles After 24 hr Exposure at  $1200^{\circ}\text{C}$  to Helium with a Water Vapor Concentration of 1000 ppm. (a) 33X. (b) Sectioned particle. 150X.

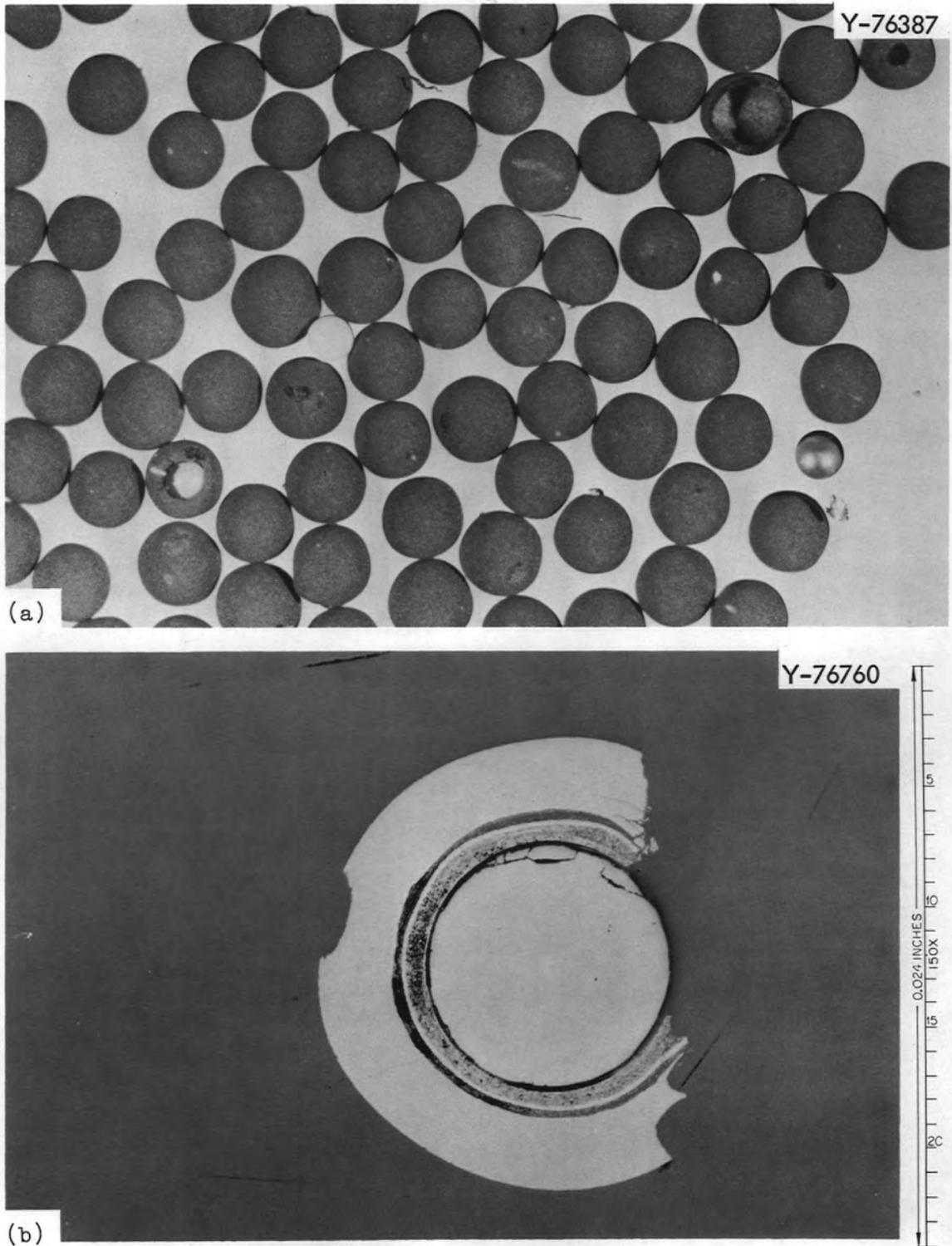


Fig. 10. Unleached OR-689 Coated Particles After 24 hr Exposure at  $1200^{\circ}\text{C}$  to Helium with a Water Vapor Concentration of 1000 ppm. (a) 33X. (b) Sectioned particle. 150X.

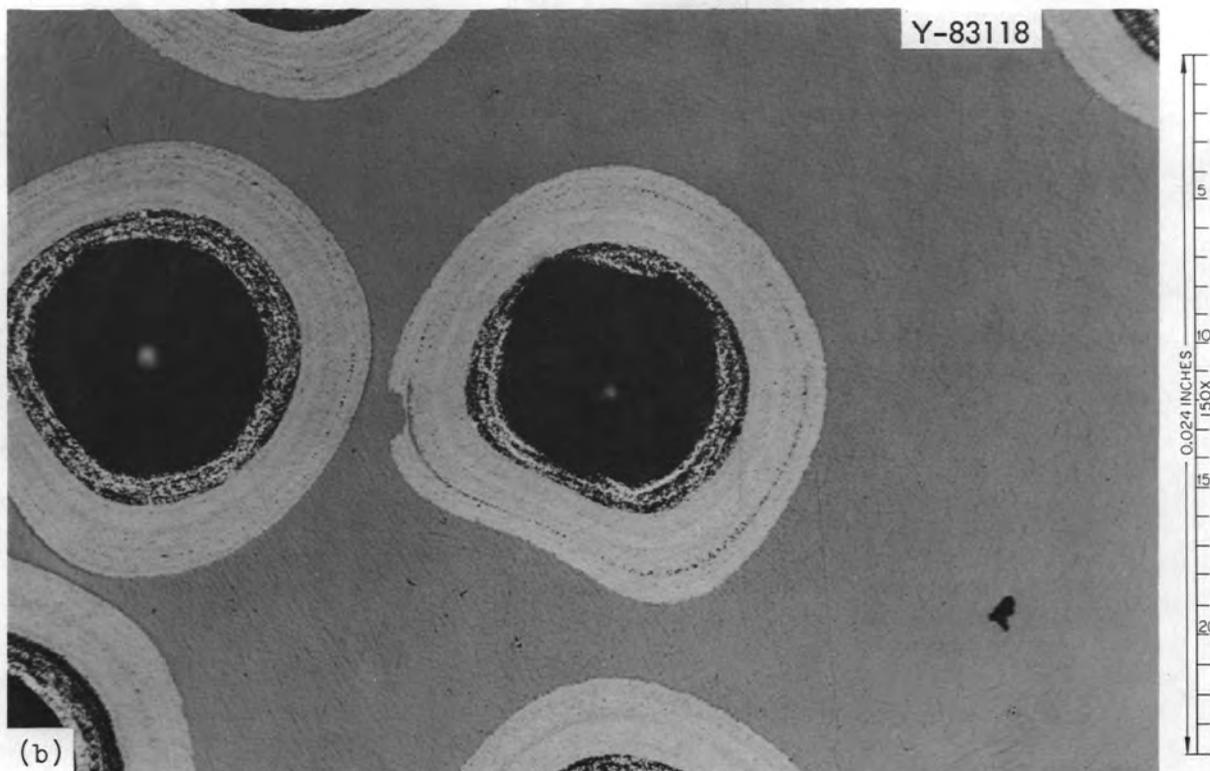
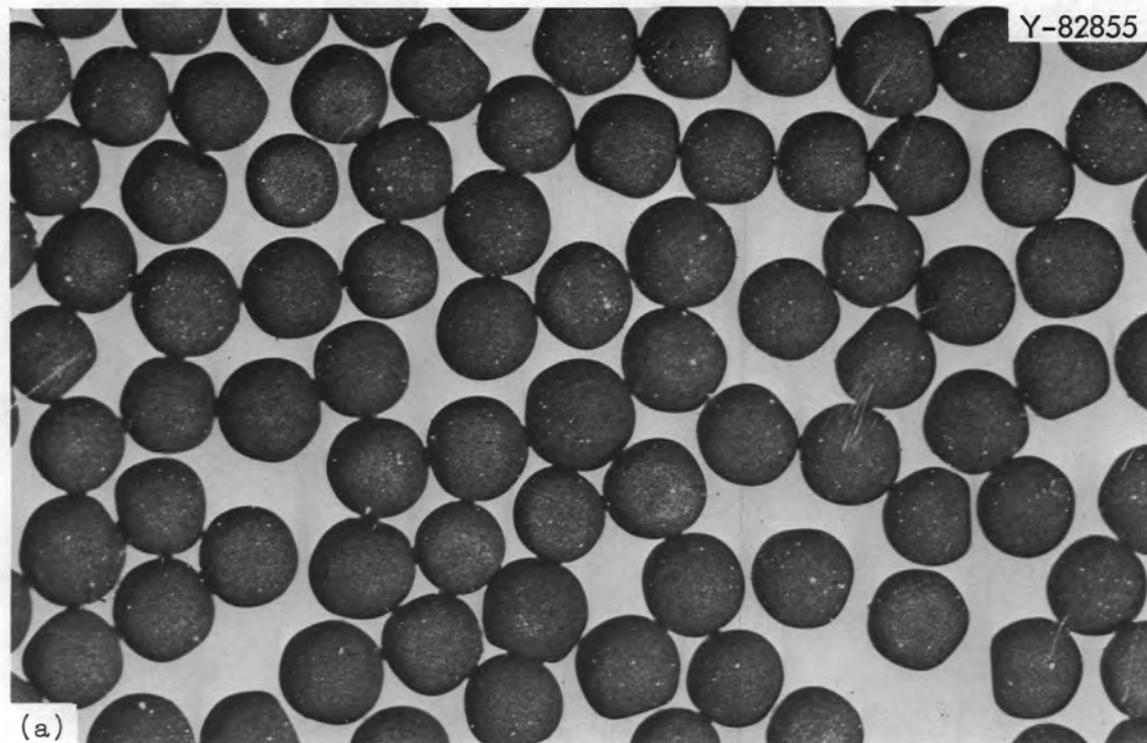


Fig. 11. Leached OR-790-C Coated Particles After 24 hr Exposure at  $1100^{\circ}\text{C}$  to Helium with Water Vapor Concentration of 1000 ppm. (a) 33X. (b) Sectioned particles. 150X.

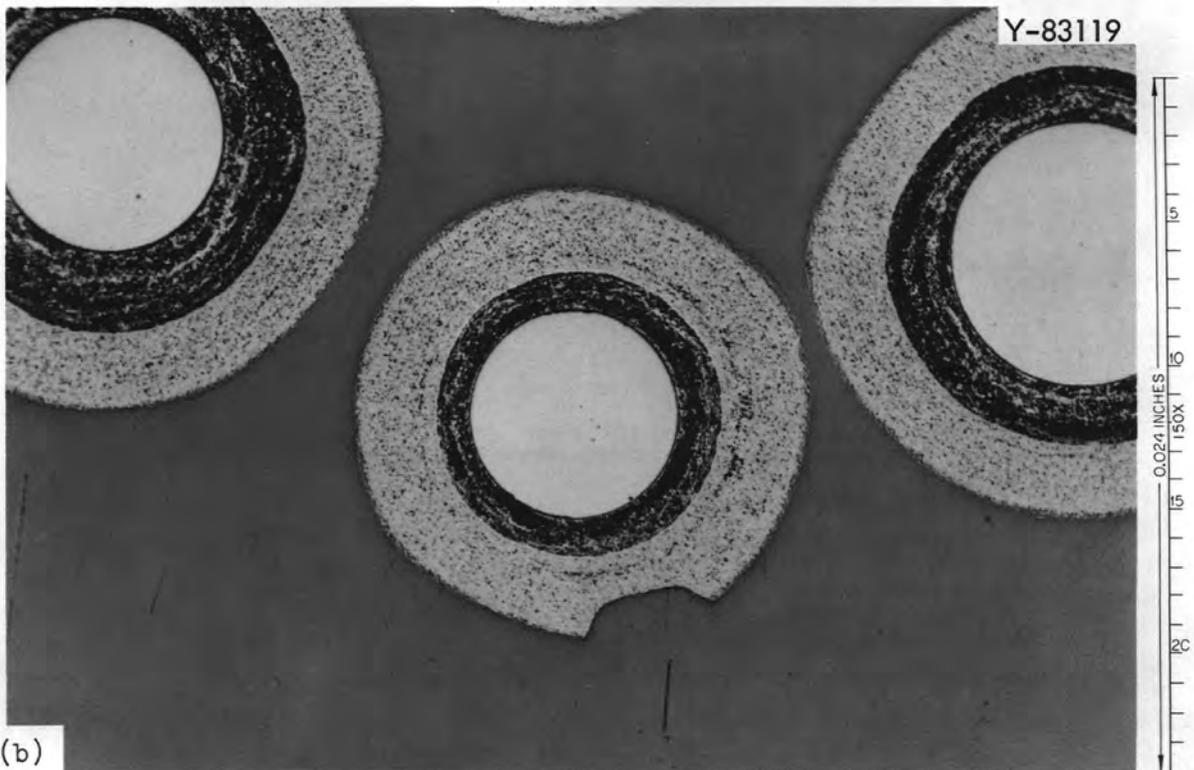
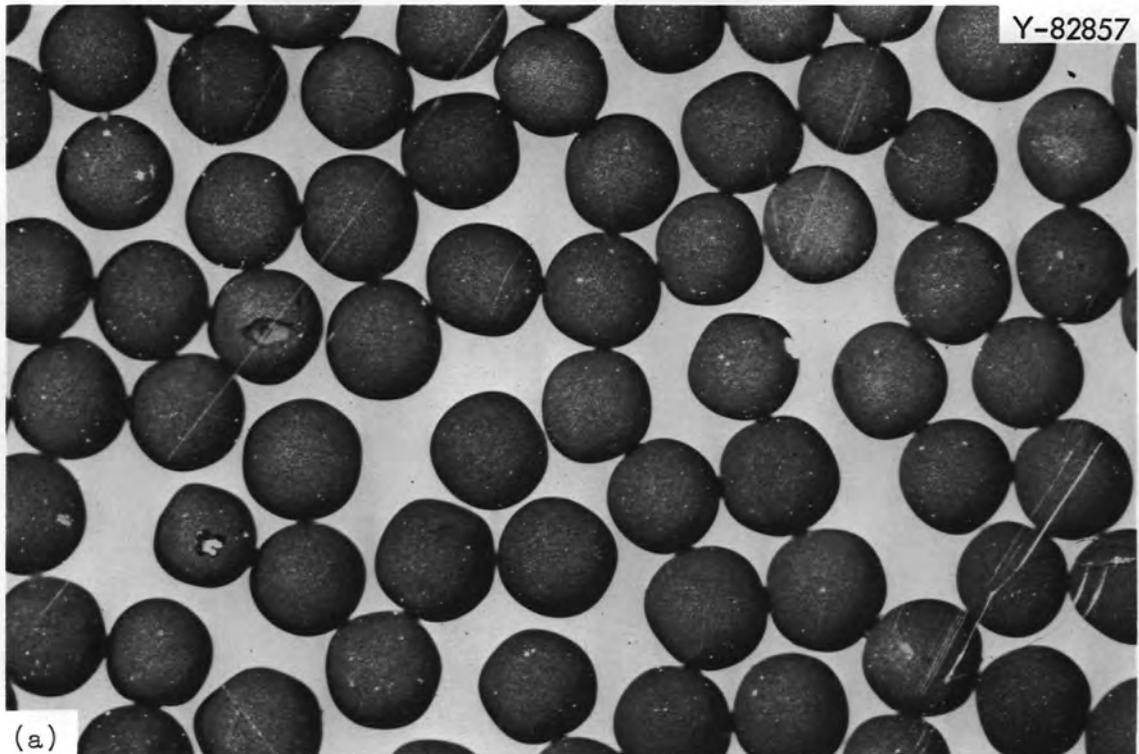


Fig. 12. Leached OR-813-R Coated Particles After 24 hr Exposure at  $1100^{\circ}\text{C}$  to Helium with a Water Vapor Concentration of 1000 ppm. (a) 33X. (b) Sectioned particles. 150X.

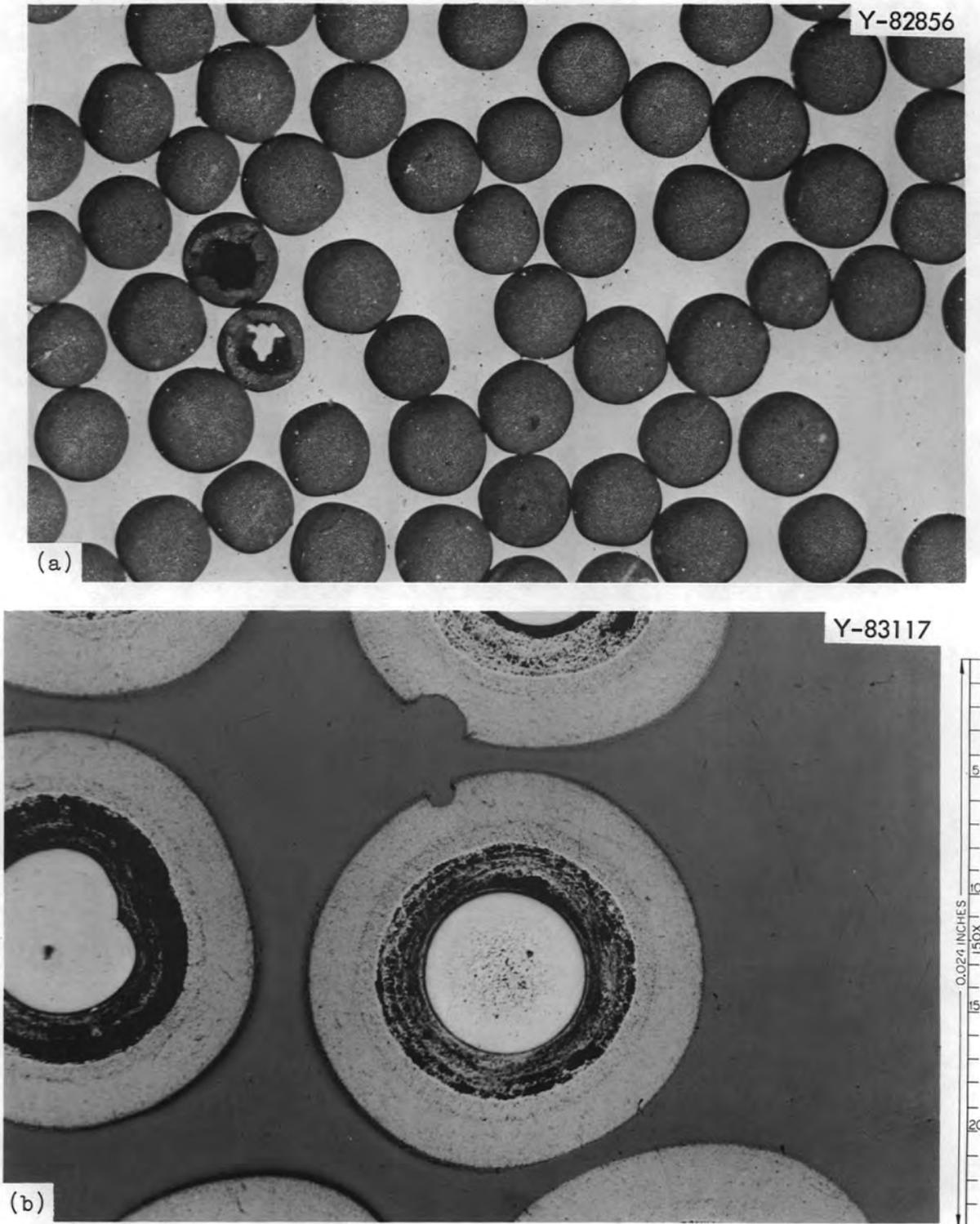


Fig. 13. Leached OR-814-R Coated Particles After 24 hr Exposure at  $1100^{\circ}\text{C}$  to Helium with a Water Vapor Concentration of 1000 ppm. (a) 33X. (b) Sectioned particles. 150X.

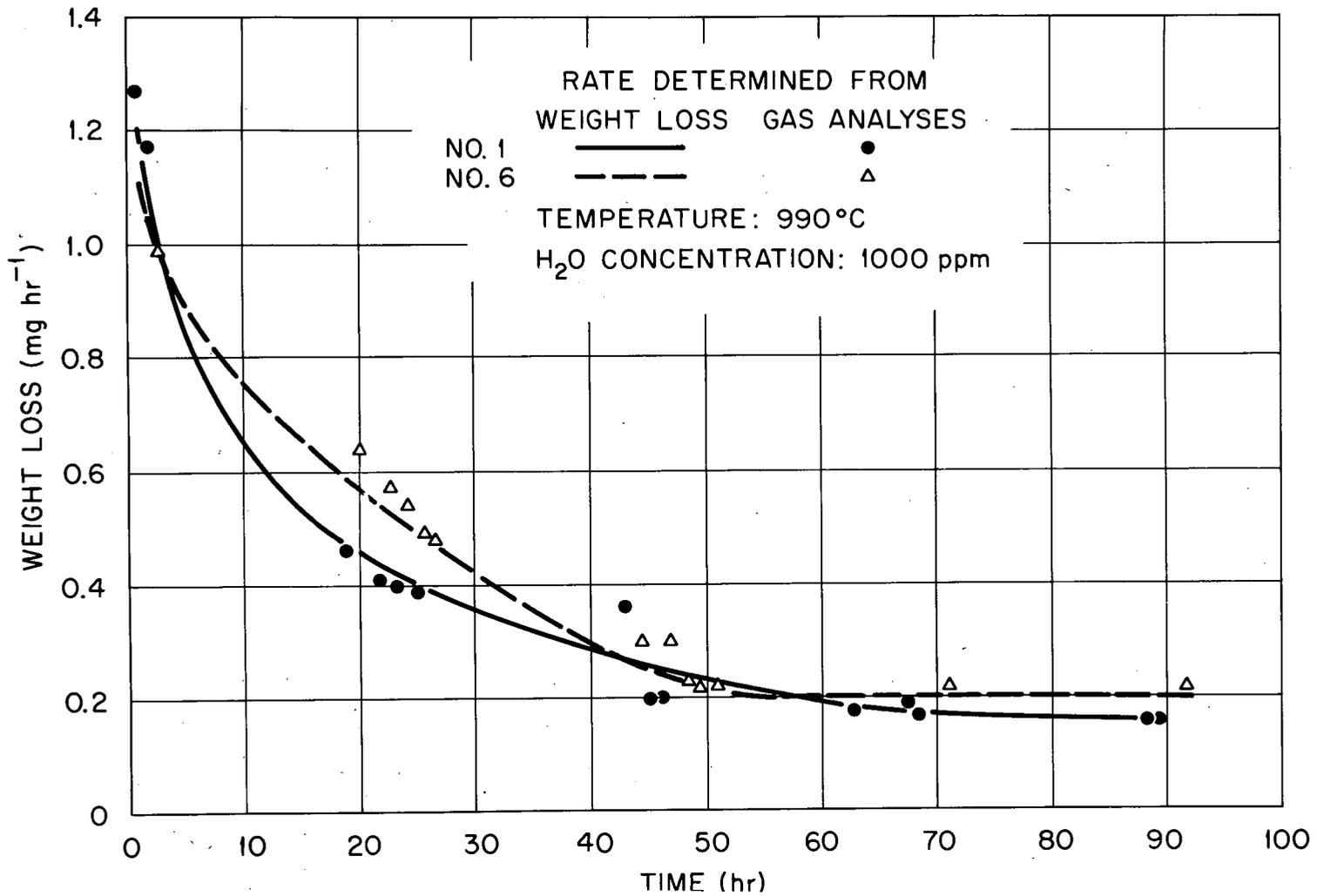


Fig. 14. Rates of Oxidation of Carbon in Fuel Sticks by Water Vapor.

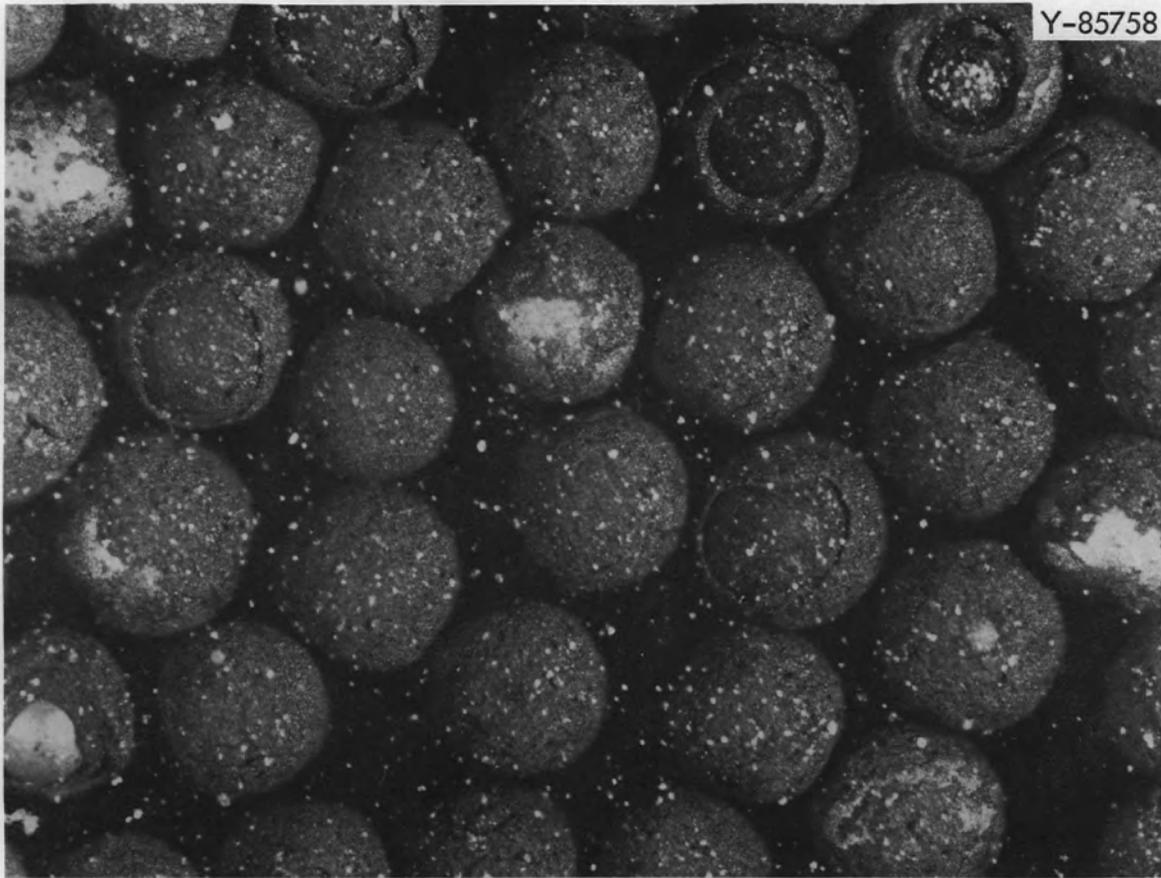


Fig. 15. Damaged Coated Particles from Oxidized Fuel Stick No. 6.

**APPENDIX A****Additional Oxidation Data**

Additional oxidation data are included in Appendix A for the various batches of coated fuel particles. It includes additional reaction rates determined from weight changes but consists mainly of data obtained from analyses of effluent gases.

Table A-1. Data From Effluent Gas Analyses

Run No.	Batch Designation	Time (hr)	Temp. (°C)	Flow Rate (cm <sup>3</sup> STP/min)	H <sub>2</sub> O Concentration (ppm)	Effluent Gas Analyses (normalized to flow of 200 cm <sup>3</sup> STP/min)			CO/CO <sub>2</sub>	Average Correction Factor	Burnoff (wt %)	Rate of Pyrocarbon Removal		
						H <sub>2</sub> (ppm)	CO (ppm)	CO <sub>2</sub> (ppm)				From Effluent Gas Analyses (mg/hr)	From Weight Change (mg/hr)	
19	Granular 4	1-2	1300	410	1000	230	74	93	0.79	0.97	2.4	1.00	0.95	
		20-21	1300	410	980	420	154	115	1.3			47	1.62	1.63
22	Granular 4	1-2	1100	200	1000	18	3	14	0.21	0.34	0.29	0.11	0.12	
		22-23	1100	195	990	-	2	9	0.21			3.3	0.07	0.04
8	Isotropic 5	2-3	1100	200	1000	139	100	33	3.0	0.78	3.3	0.80	0.80	
		4-5	1100	200		88	49	30	1.6			5.3	0.48	0.32
		23-24	1100	200	1000	39	11	27	0.38			14	0.23	0.20
9	Isotropic 5	1-2	1200	200	1000	181	100	54	1.8	0.91	1.9	0.93	0.93	
		18-19	1200	200	1000	171	62	59	1.1			29	0.73	0.72
10	Isotropic 5	1-2	1300	200	1000	186	77	77	1.0	0.92	2.2	0.91	0.76	
		19-20	1300	200	1000	297	162	89	1.8			49	1.51	1.44
11	Isotropic 5	1-2	1400	200	1000	200	99	72	1.4	0.96	3.2	1.02	1.34	
		5-6	1400	200		335	200	91	2.2			16	1.14	1.42
		7-8	1400	200		425	283	88	3.2			23	2.23	2.04
		11-12	1400	200	1000	560	395	78	5.1			36	2.84	2.21
13	Isotropic 6	1-2	1200	200	980	25	5	18	0.29	0.53	0.31	0.14	0.14	
		19-20	1200	200	1000	16	4	17	0.24			3.0	0.12	0.10
14	Isotropic 6	1-2	1300	200	1000	102	27	54	0.50	0.86	1.0	0.49	0.51	
15	Isotropic 6	1-2	1400	200	980	200	84	81	1.0	0.95	2.1	0.98	1.12	
		22-23	1400	200	1000	314	165	88	1.9			42	1.52	1.34
16	Isotropic 6	1-2	1300	410	1000	115	16	58	0.27	0.89	1.0	0.44	0.47	
		19-20	1300	425	1000	130	18	67	0.27			14	0.51	0.46
17	Isotropic 6	1-2	1300	400	500	76	16	40	0.40	0.82	0.70	0.34	0.31	
		20-21	1300	400	500	60	14	36	0.39			8.3	0.30	0.26
29	Isotropic 6	1-2	1200	200	995	14	2	13	0.85	0.46	0.22	0.09	0.10	
		22-23	1200	200	1000	22	5	24	0.75			3.4	0.17	0.10
45	Isotropic 6	1-2	1100	200	1020	10	2	8	0.24	0.87	0.01	0.06	0.03	
		4-5	1100	200		5	1	3	0.29			0.19	0.03	0.02
		23-24	1100	200	1020	5	1	3	0.29			0.76	0.03	0.02
53	Isotropic 6	1-2	1300	400	250	45	15	20	0.75	0.97	0.67	0.21	0.14	
		5-6	1300	400		43	12	20	0.60			1.3	0.20	0.13
20	Isotropic 7	1-2	1300	410	1000	175	44	82	0.54	0.94	1.3	0.76	0.63	
		20-21	1300	400	990	121	29	65	0.46			17	0.56	0.52

Table A-1. Data From Effluent Gas Analyses (continued)

Run No.	Batch Designation	Time (hr)	Temp. (°C)	Flow Rate (cm <sup>3</sup> STP/min)	H <sub>2</sub> O Concentration (ppm)	Effluent Gas Analyses (normalized to flow of 200 cm <sup>3</sup> STP/min)			CO/CO <sub>2</sub>	Average Correction Factor	Burnoff (wt %)	Rate of Pyrocarbon Removal	
						H <sub>2</sub> (ppm)	CO (ppm)	CO <sub>2</sub> (ppm)				From Effluent Gas Analyses (mg/hr)	From Weight Change (mg/hr)
25	YZ-134	1-2	1200	200	1000	177	85	56	1.5	0.91	1.8	0.85	0.75
		22-23	1200	195	1050	163	73	56	1.3		35	0.78	0.71
28	YZ-134	1-2	1200	200	1000	96	34	40	0.86	0.93	0.93	0.45	0.54
		20-21	1200	200	1000	27	4.5	20	0.23		10.8	0.15	0.14
50	YZ-134	1-2	1100	200	1010	10	1	6.5	0.15	0.93	0.11	0.05	0.02
		3-4	1100	200		10	1	6.5	0.15		0.23	0.05	0.03
		5-6	1100	200		10	1.5	7.5	0.20		0.36	0.05	0.03
		23-24	1100	200	1020	7	1.5	6	0.25		1.5	0.05	0.04
23	YZ-135	1-2	1200	200	1000	148	71	51	1.2	0.91	1.4	0.77	0.76
		2-3	1200	200		218	128	67	1.9		4.2	1.17	0.84
		22-23	1200	190	1000	188	97	59	1.6		44	0.93	0.89
24	YZ-136	1-2	1200	200	1000	63	15	36	0.41	0.85	0.83	0.31	0.29
		22-23	1200	195	930	129	58	46	1.3		29	0.63	0.46
26	OR-688	1-2	1200	205	1000	77	24	34	0.69	0.86	0.89	0.35	0.34
		20-21	1200	210	1010	103	38	40	0.94		17	0.47	0.47
27	OR-689	1-2	1200	205	980	79	22	33	0.66	0.83	1.1	0.33	0.41
		22-23	1200	210	1000	91	35	37	0.93		18	0.43	0.44
30	OR-689	1-1.5	1190	200	510	29	14	26	0.53	0.44	0.64	0.23	0.32
		22-23	1180	200	510	24	16	27	0.58		7.8	0.25	0.21
31	OR-689	1-2	1190	200	260	32	26	16	1.6	0.53	0.62	0.26	0.27
		19-20	1180	200		14	8	17	0.46		5.4	0.15	0.11
		22-23	1180	200	260	15	9.5	16	0.58		6.1	0.16	0.12
33	OR-689	1-2	1290	205	520	131	56	51	1.1	0.94	1.5	0.64	0.52
		4-5	1290	205		101	35	49	0.73		5.0	0.50	0.45
		23-24	1290	200	540	115	46	50	0.91		23	0.57	0.47
34	OR-689	1-2	1290	215	260	56	24	20	1.1	0.96	0.8	0.26	0.22
		5-6	1290	215		57	24	21	1.1		2.6	0.27	0.21
		23-24	1290	205	280	57	26	20	1.2		12	0.27	0.33
35	OR-689	1-2	1290	205	1000	232	79	83	0.95	0.99	2.7	0.97	0.69
		4-5	1290	205		304	125	106	1.0		8.3	1.38	1.05
		23-24	1290	205	1020	315	135	106	1.4		55	1.44	1.19
49	OR-689	1-2	1110	200	1000	35	9	15	0.60	0.95	0.13	0.14	0.06
		3-4	1110	200		31	7.5	15	0.48		0.44	0.14	0.11
		5-6	1110	200		32	7.5	14	0.54		0.96	0.13	0.15
		23-24	1110	200	950	40	12	15	0.81		6.7	0.17	0.18

Table A-1. Data From Effluent Gas Analyses (continued)

Run No.	Batch Designation	Time (hr)	Temp. (°C)	Flow Rate (cm <sup>3</sup> STP/min)	H <sub>2</sub> O Concentration (ppm)	Effluent Gas Analyses (normalized to flow of 200 cm <sup>3</sup> STP/min)			CO/CO <sub>2</sub>	Average Correction Factor	Burnoff (wt %)	Rate of Pyrocarbon Removal	
						H <sub>2</sub> (ppm)	CO (ppm)	CO <sub>2</sub> (ppm)				From Effluent Gas Analyses (mg/hr)	From Weight Change (mg/hr)
52	OR-689	1-2	1200	200	1020	95	27	38	0.70	0.99	1.0	0.39	0.35
		3-4	1200	220		109	34	44	0.52		2.7	0.47	0.45
		5-6	1200	205		112	36	45	0.81		4.5	0.49	0.51
		23-24	1250	215		1040	163	57	65		0.88	25	0.74
40	OR-788	1-2	1100	200	980	62	20	24	0.83	0.96	0.5	0.26	0.22
		5-6	1100	200	28	8	11	0.77	1.7		0.12	0.12	
		22-23	1100	205	980	14	3	12	0.26		4.5	0.09	0.10
36	OR-789-C	1-2	1195	200	1000	184	73	79	0.91	0.98	1.8	0.91	0.90
		5-6	1195	200	209	96	67	1.4	11		0.98	1.20	
		22-23	1195	200	900	257	149	82	1.8		59	1.40	3.50
37	OR-789-C	1-2	1100	210	1000	77	18	35	0.53	0.97	0.6	0.32	0.35
		5-6	1100	210	96	27	39	0.70	3.3		0.40	0.60	
		23-24	1100	210	1040	270	128	80	1.6		39	1.25	1.30
42	OR-790-C	1-2	1100	200	980	168	87	53	1.6	0.98	2.5	0.85	0.66
		5-6	1100	200	55	23	22	1.0	5.5		0.26	0.25	
		23-24	1115	200	1020	65	26	22	1.2		15.7	0.29	0.31
44	OR-813-R	1-2	1200	205	990	53	12	29	0.40	0.98	0.57	0.25	0.27
		5-6	1200	200	85	23	36	0.64	2.5		0.36	0.32	
		22-23	1200	200	1040	66	14	33	0.43		14	0.28	0.27
46	OR-813-R	1-2	1190	200	1010	41	6	22	0.27	0.98	0.35	0.17	0.13
		3-4	1200	200	45	8	21	0.38	0.93		0.17	0.15	
		5-6	1200	200	48	10	21	0.50	1.5		0.19	0.19	
		22-23	1210	200	800	72	19	31	0.63		9.5	0.30	0.26
48	OR-813-R	1-2	1110	205	1010	16	4	9	0.39	0.95	0.20	0.08	0.08
		3-4	1110	205	17	4	11	0.33	0.48		0.09	0.05	
		5-6	1110	205	16	4	9	0.44	0.70		0.08	0.05	
		22-23	1110	205	1040	20	4	11	0.36		2.9	0.09	0.09
41	OR-814-R	1-2	1100	205	980	32	9	12	0.75	0.95	0.42	0.13	0.16
		5-6	1100	205	24	6	12	0.50	1.1		0.11	0.08	
		23-24	1100	205	980	29	10	13	0.80		5.3	0.14	0.15
43	OR-814-R	1-2	1200	200	990	98	32	37	0.88	0.97	1.0	0.42	0.35
		3-4	1200	200	76	26	29	0.90	2.0		0.33	0.31	
		5-6	1200	200	65	23	25	0.90	3.1		0.29	0.28	
		22-23	1200	180	600	68	27	26	1.1		12	0.32	0.29

Table A-1. Data From Effluent Gas Analyses (continued)

Run No.	Batch Designation	Time (hr)	Temp. (°C)	Flow Rate (cm <sup>3</sup> STP/min)	H <sub>2</sub> O Concentration (ppm)	Effluent Gas Analyses (normalized to flow of 200 cm <sup>3</sup> STP/min)			CO/CO <sub>2</sub>	Average Correction Factor	Burnoff (wt %)	Rate of Pyrocarbon Removal	
						H <sub>2</sub> (ppm)	CO (ppm)	CO <sub>2</sub> (ppm)				From Effluent Gas Analyses (mg/hr)	From Weight Change (mg/hr)
39	OR-815	1-2	1100	205	1000	187	86	62	1.4	0.97	2.0	0.89	0.62
		3-4	1100	210		96	46	31	1.4		4.1	0.46	0.35
		5-6	1100	210		60	28	23	1.2		5.4	0.30	0.26
		22-23	1080	200	1020	36	16	12	1.4		13	0.16	0.16
51	OR-815	1-2	1200	200	1010	243	123	76	1.6	0.99	3.2	1.20	0.94
		3-4	1200	200		215	100	73	1.4		7.0	1.04	0.83
		5-6	1200	200		215	103	73	1.4		10.4	1.05	0.70
		23-24	1200	205	1010	228	112	76	1.5		41	1.13	0.89
54	OR-818	1-2	1110	210	1020	45	9	21	0.43		0.53	0.18	0.17
		4-5	1110	210	1000	42	7	22	0.33		1.7	0.18	0.16
		46-47	1200	200	1000	107	58	54	1.1		39	0.67	0.51
		52-53	1210	200		112	49	43	1.1		46	0.56	0.55
		73-74	1205	200		73	24	27	0.91		71	0.31	0.45
		93-94	1210	200		28	5	17	0.29		91	0.13	0.34
		118-119	1210	200	1000	<1	<1	<1	-		100	<0.01	0.16
		142-143	1300	200	1000	171	88	46	1.9		-	0.81	0.36
		145-146	1295	200		35	3	14	0.25		-	0.11	0.10
		163-164	1350	200	1000	<1	<1	<1	-		-	<0.01	>0.01
		165-166	1400	200	1000	<1	<1	<1	-		-	<0.01	0.04
		212-213	1400	200	1000	<1	<1	<1	-		-	<0.01	0.04

## APPENDIX B

## Plots of Reaction Rates as Function of Burnoff

Some representative reaction rate vs burnoff curves are given for a number of batches of coated particles. Reaction rates are expressed in terms of the weight of pyrolytic carbon remaining at any particular time.

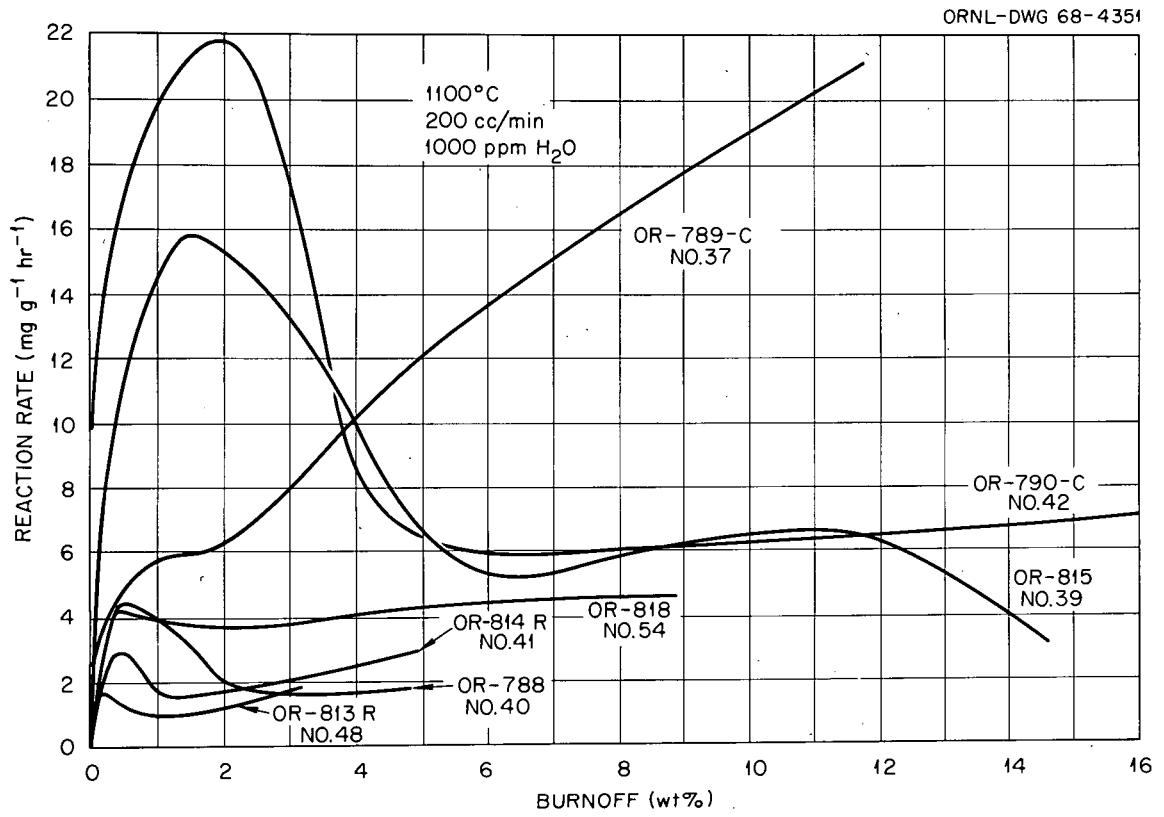


Fig. B-1

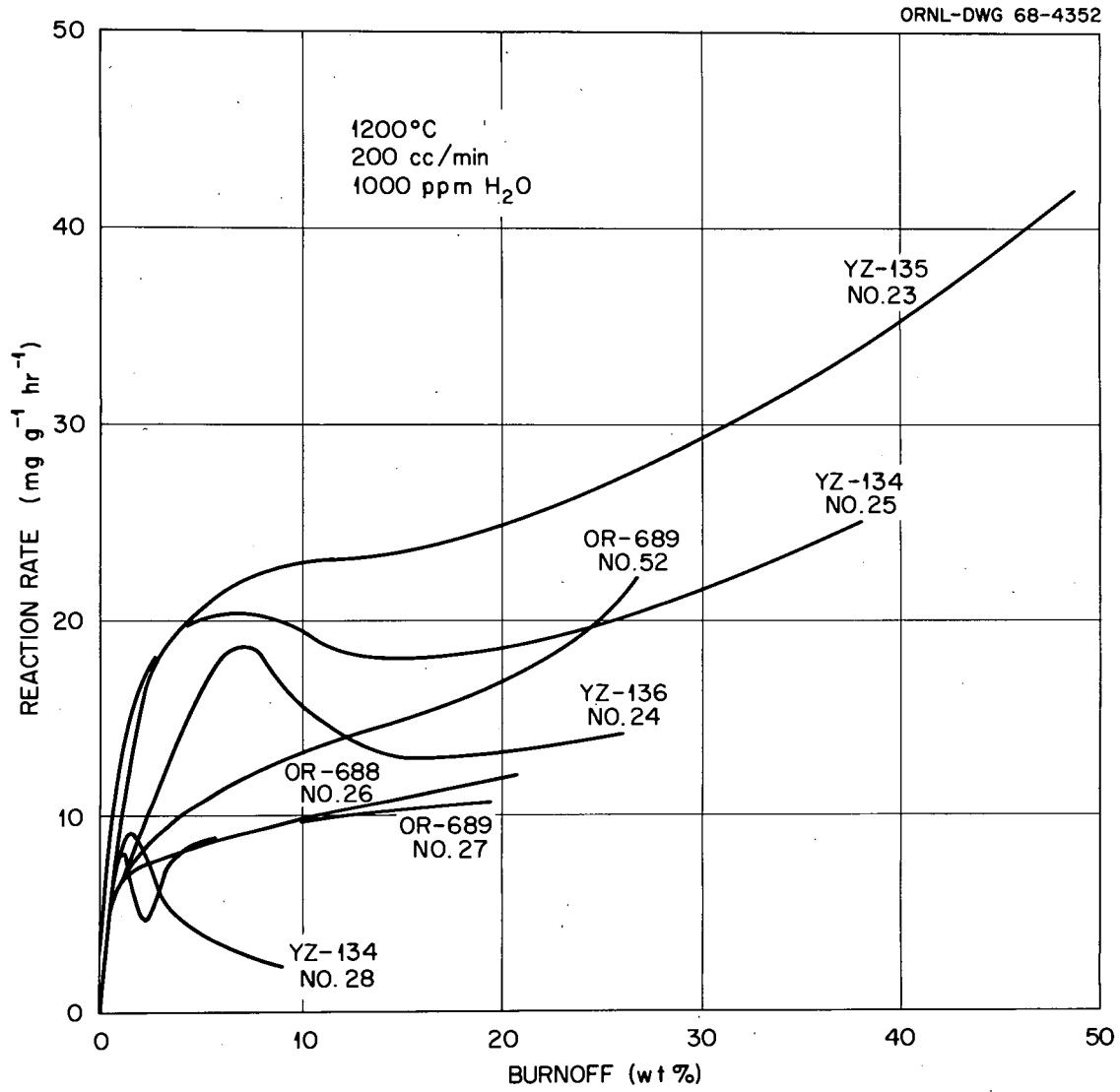


Fig. B-2

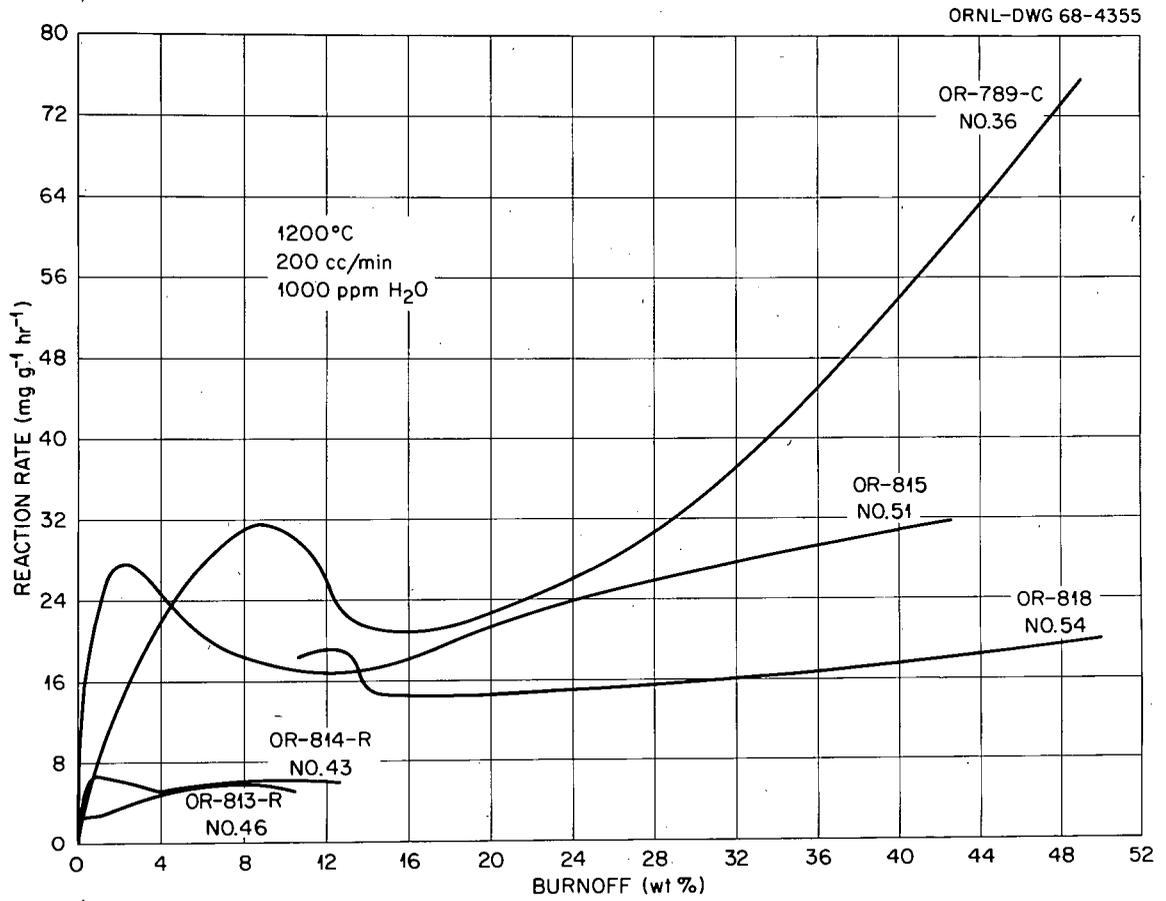


Fig. B-3

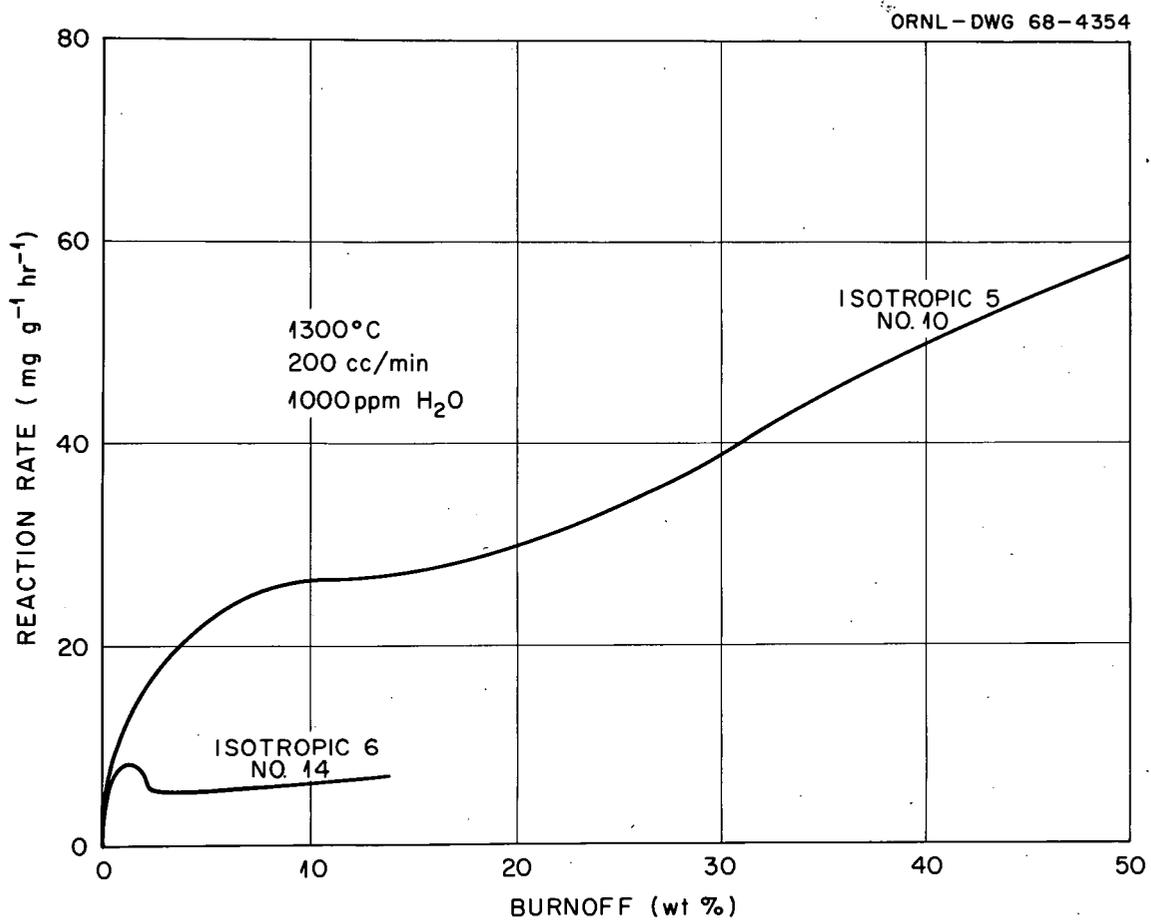


Fig. B-4

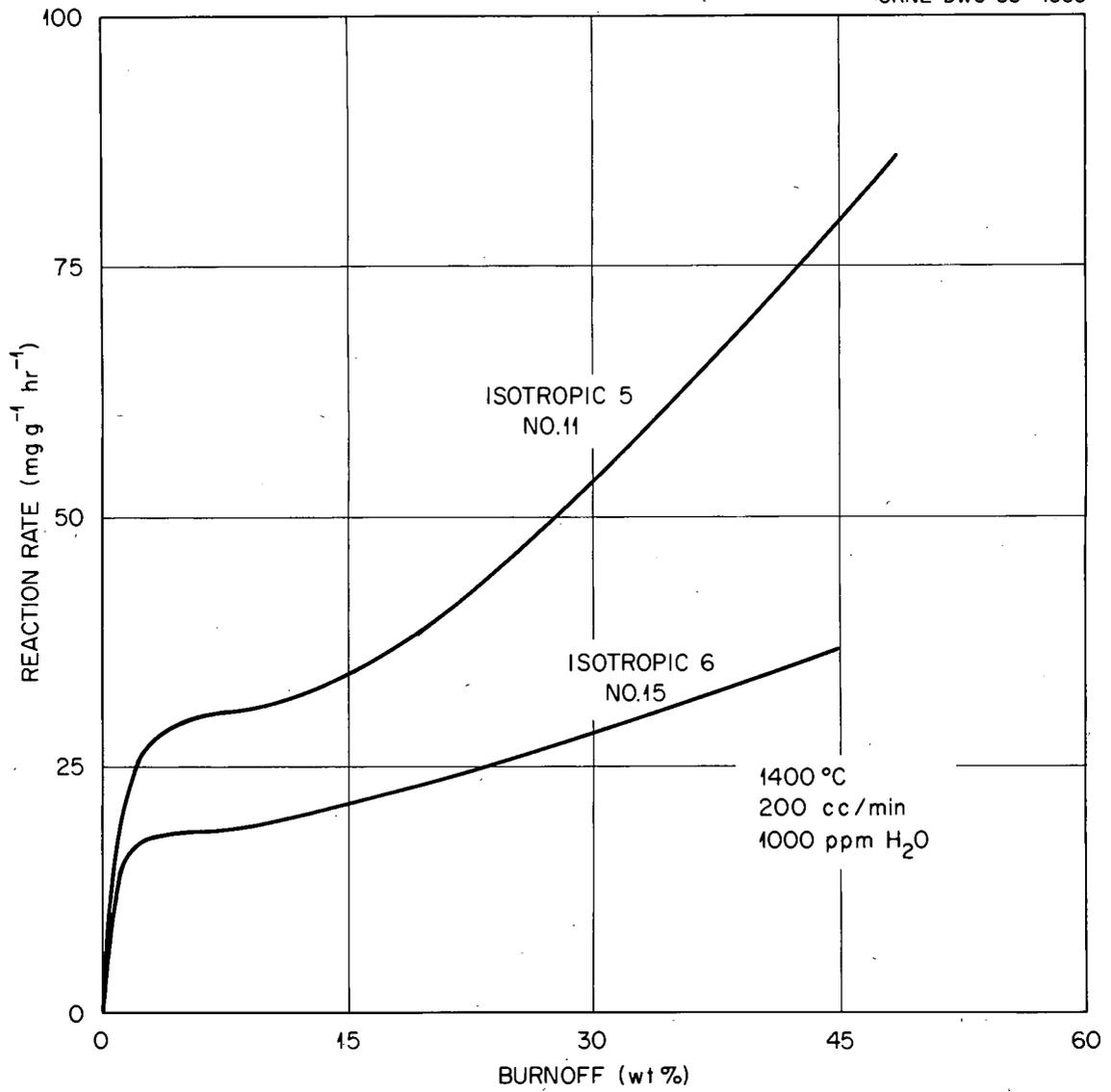
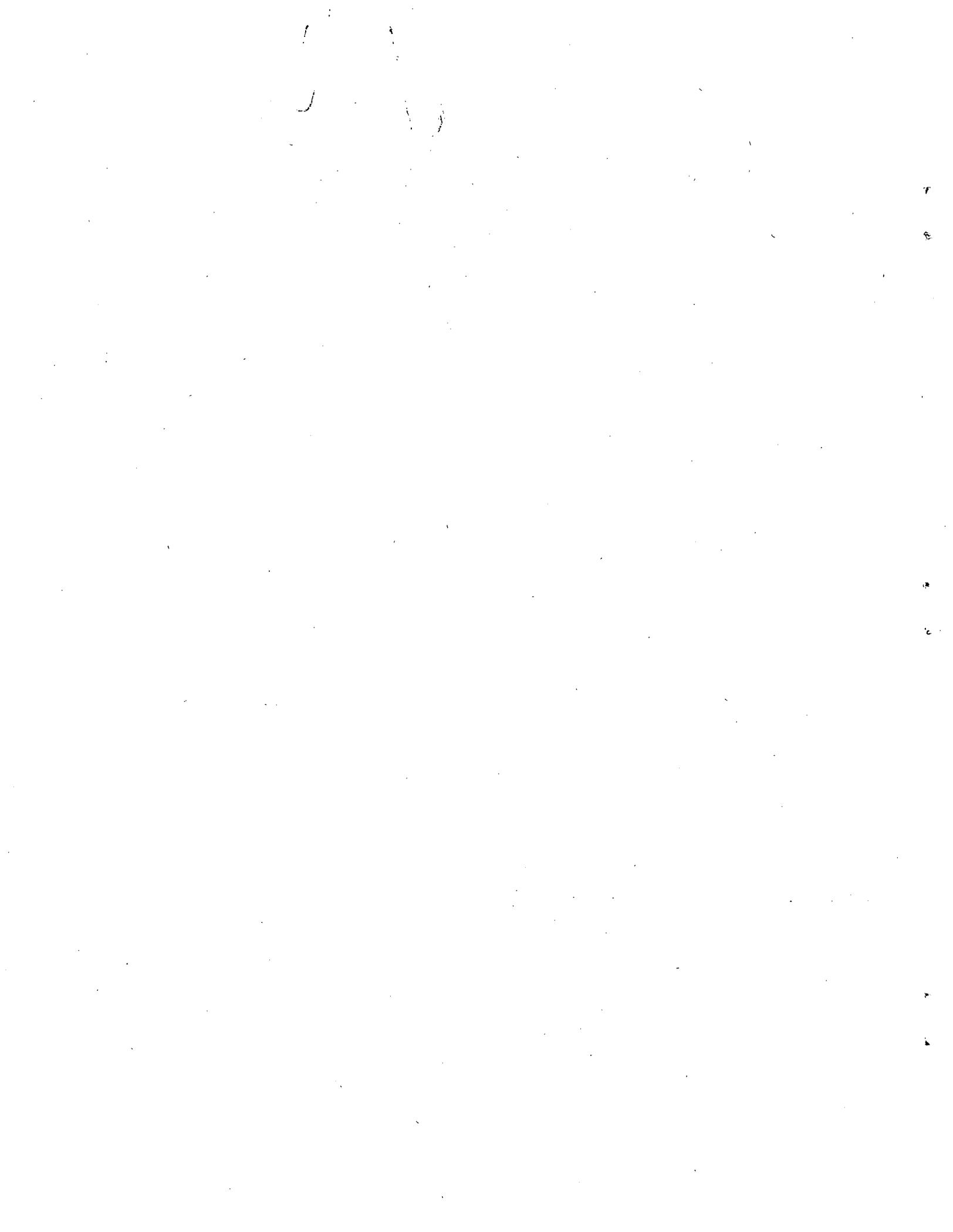


Fig. B-5



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