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DECOMPOSITION PRODUCTS OF PLUTONYL NITRATE AND PLUTONIUM OXALATE

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ABSTRACT

A study has been made of the decomposition products of $PuO_2(NO_3)_2$ and $Pu_2(C_2O_4)_3$ in an effort to isolate either a higher oxide in the case of the plutonyl nitrate or a carbonate or sesquioxide in the case of the Pu(III) oxalate.

The results of these experiments show that both of these compounds decompose directly to the dioxide with no evidence of formation of any intermediate compounds.

DECOMPOSITION PRODUCTS OF PLUTONYL NITRATE AND PLUTONIUM OXALATE

DECOMPOSITION OF PLUTONYL NITRATE

A solution of plutonyl nitrate was dried in a micro platinum boat at ~40°C. The dried nitrate was then placed in a 10-mm. pyrex tube heated in a micro "regenerative block" and the temperature raised stepwise while aspirating air through the tube. The boat was cooled and weighed at intervals. Results are given in Table I and plotted in Fig. 1.

From these results it seems unlikely that any PuO_3 was formed since the weight dropped directly to PuO_2 at 230°C. Further heating resulted in only a very slight weight decrease. Horizontal lines on the graph show weights which corresponded to the formation of PuO_3 (not found) and PuO_2.

DECOMPOSITION OF Pu(III) OXALATE

A small sample of partially dehydrated $Pu_2(C_2O_4)_3$ was weighed into a micro platinum boat. The boat containing oxalate was placed in a quartz reaction tube in a Fisher micro combustion furnace. A slow stream of CO_2 was allowed to flow through the system. Temperature regulation was by means of a Capacitrol controller. The time at each temperature and the weight of sample after each treatment are shown in Table II, and the results are plotted in Fig. II. The black line shows results of the first series of experiments, with horizontal lines corresponding to theoretical values for $Pu_2(CO_3)_3$ and Pu_2O_3. These theoretical values were calculated from the weight of the oxide that had been ignited to 1100°C. The dotted line is a second experiment conducted in the same manner as the first, but with smaller temperature intervals near the decomposition temperature.

From these two series of experiments it can be seen that the weight drops directly to PuO_2 at 325° and does not stop at the carbonate or go on down to the sesquioxide in a CO_2 atmosphere.

The above experiments were repeated except that a stream of H_2 instead of CO_2 was used for the inert atmosphere. The data for this experiment are given in Table III, and the results are plotted in Fig. III. During this treatment with H_2, the material reached a constant weight at 300°. A sample of 94.8 mg was removed for carbon analysis at this time and the remainder was weighed. The treatment was continued until the dioxide was formed. The carbon analysis indicated 0.71 mg of carbon present. This weight of carbon does not correspond to any definite compound.

As in the experiment with CO_2 no evidence of the formation of a stable carbonate or sesquioxide was found.

Two attempts were made by S. Bakes to prepare the sesquioxide by reduction of the dioxide with H_2 at 1000°C for several hours. Neither showed any loss in weight as would be necessary for the formation of a sesquioxide.

TABLE I

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Time In	Temp. In °C	Time Out	Temp. Out °C	Weight, mg
9:35	Room Temp.	10:26	138	16.52
10:48	152	11:10	160	16.05
11:17	162	11:47	190	15.43
1:37*	167	2:07*	210	15.21
2:20*	205	2:44*	230	14.00
2:51*	222	3:29*	226	11.47
3:47*	208	4:43*	229	10.91
4:54*	212	5:26*	238	10.89
7:30*	238	8:27*	230	10.84
8:34*	262	8:58*	310	10.82
9:03*	312	9:23*	355	10.78
overnight			355	10.77

* P.M.

TABLE II

CO_2 Atmosphere

Time In	Time Out	Temp. °C	Total Time	Weight, mg
10:35	11:20	200	45 min.	136.4
11:35	1:17*	250	102 min.	135.0
1:40*	8:53*	300	7 hrs. 13 min.	131.9
9:10*	8:15	350	11 hrs. 5 min.	123.2
9:34	9:07*	400	11 hrs. 33 min.	99.2
9:25*	9:15	450	11 hrs. 50 min.	98.7
10:40	8:15	500	21 hrs. 35 min.	97.1
		1100 (air)		95.5
9:45	9:55	200	10 min.	92.2
10:05	11:15	225	70 min.	91.8
11:35	1:03*	250	88 min.	91.6
1:25*	2:15*	275	50 min.	91.2
2:35*	3:50*	300	75 min.	89.7
4:05*	5:05*	325	60 min.	87.4
5:30*	9:10*	325	3 hrs. 40 min.	74.9
9:30*	8:20	325	10 hrs. 50 min.	68.5
9:45	10:50	1100°(air)	1 hr.	66.7

* P.M.

Table III

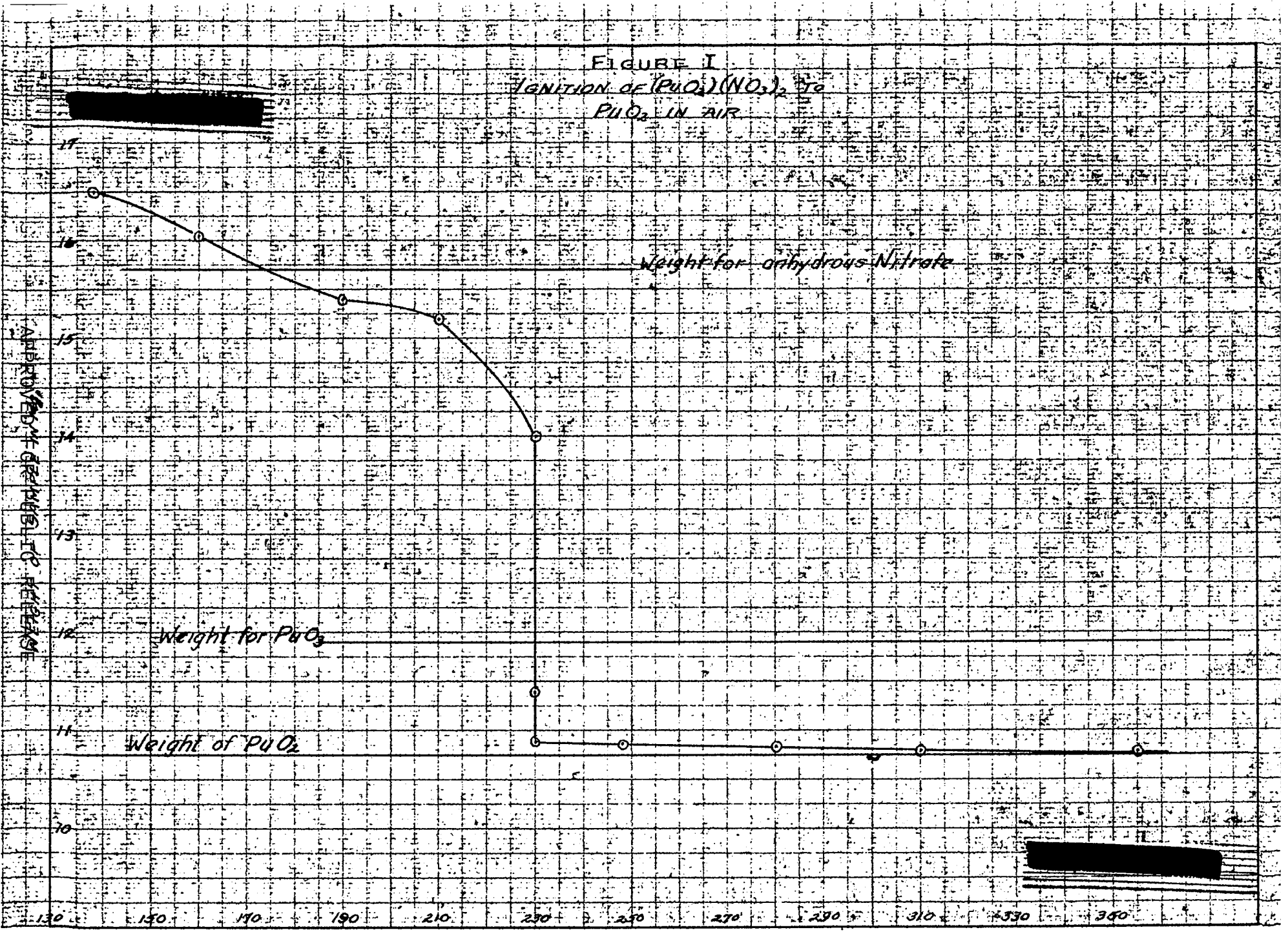
(90-92) H_2 Atmosphere

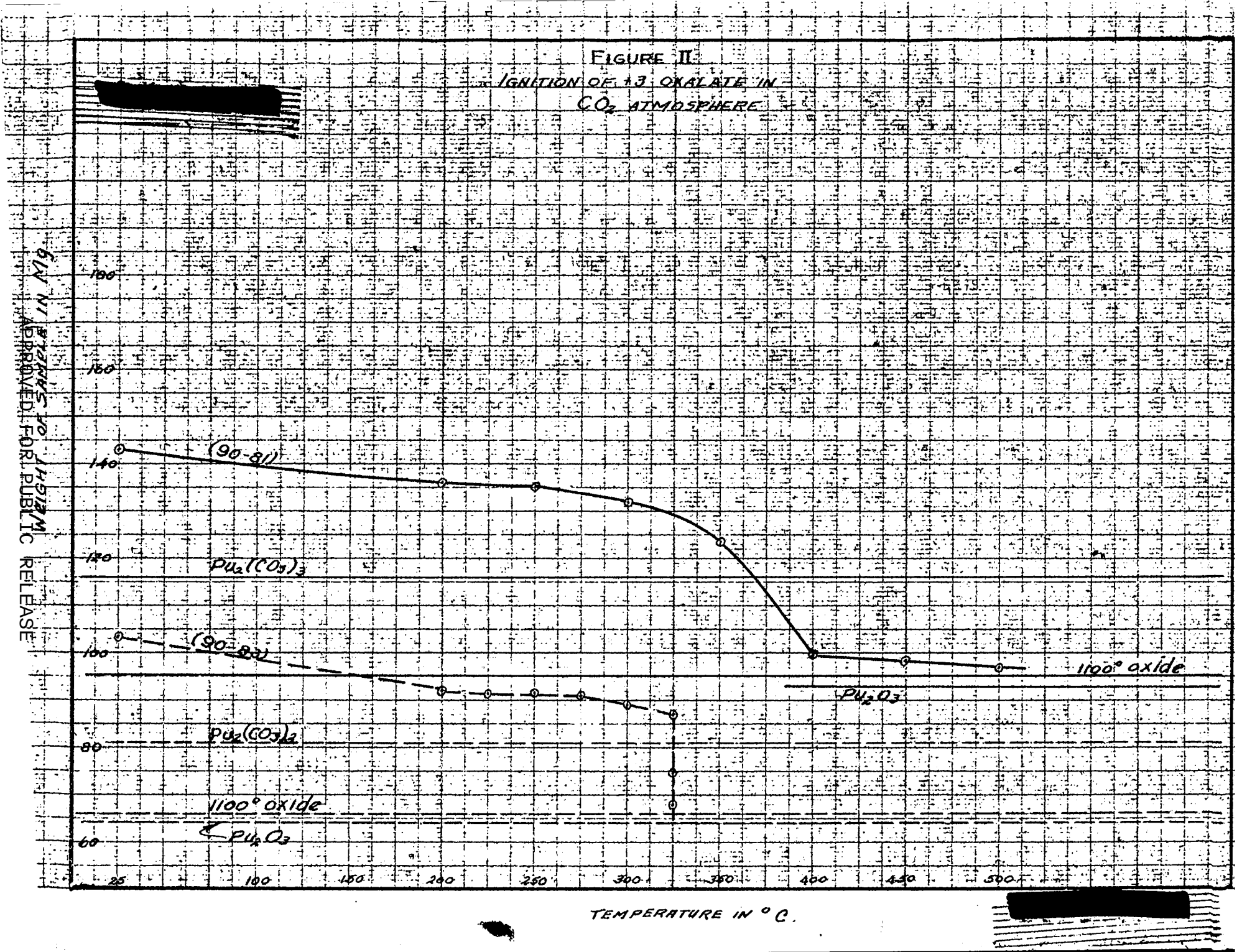
Time In	Time Out	Temp, °C	Total Time	Weight, mg
3:25*	4:25*	200	1 hr.	614.8
5:00*	7:25*	200	2 hrs. 25 min.	612.8
9:00	10:00	225	1 hr.	611.6
10:45	1:05*	250	2 hrs. 20 min.	609.4
2:05*	3:15*	275	1 hr. 10 min.	607.3
4:04*	5:04*	275	1 hr.	606.5
9:25	11:25	290	2 hrs.	591.3
2:08*	4:08*	290	2 hrs.	569.2
5:20*	8:40*	290	3 hrs. 20 min.	533.9
10:00*	8:05	290	10 hrs. 5 min.	476.0
9:30	12:55*	290	3 hrs. 25 min.	469.5
2:00*	3:45*	290	1 hr. 45 min.	471.8
9:15	1:05*	290	3 hrs.	467.9
2:15*	7:15*	290	5 hrs.	467.0
9:30*	8:15	300	11 hrs. 15 min.	466.3
9:15	9:45	310	30 min.	468.8
Sample removed for carbon analysis. Weight left				380.1
6:00*	8:20	360	14 hrs. 20 min.	373.0
9:50	1:05*	360	3 hrs. 15 min.	373.1
2:15*	8:10	400	17 hrs. 55 min.	373.1
9:30	1:05*	500	3 hrs. 35 min.	370.2
		1000° (air)	1 hr.	363.3

* P.M.

FIGURE I

IGNITION OF $(PuO_2)(NO_3)_2$ TO PuO_3 IN AIR

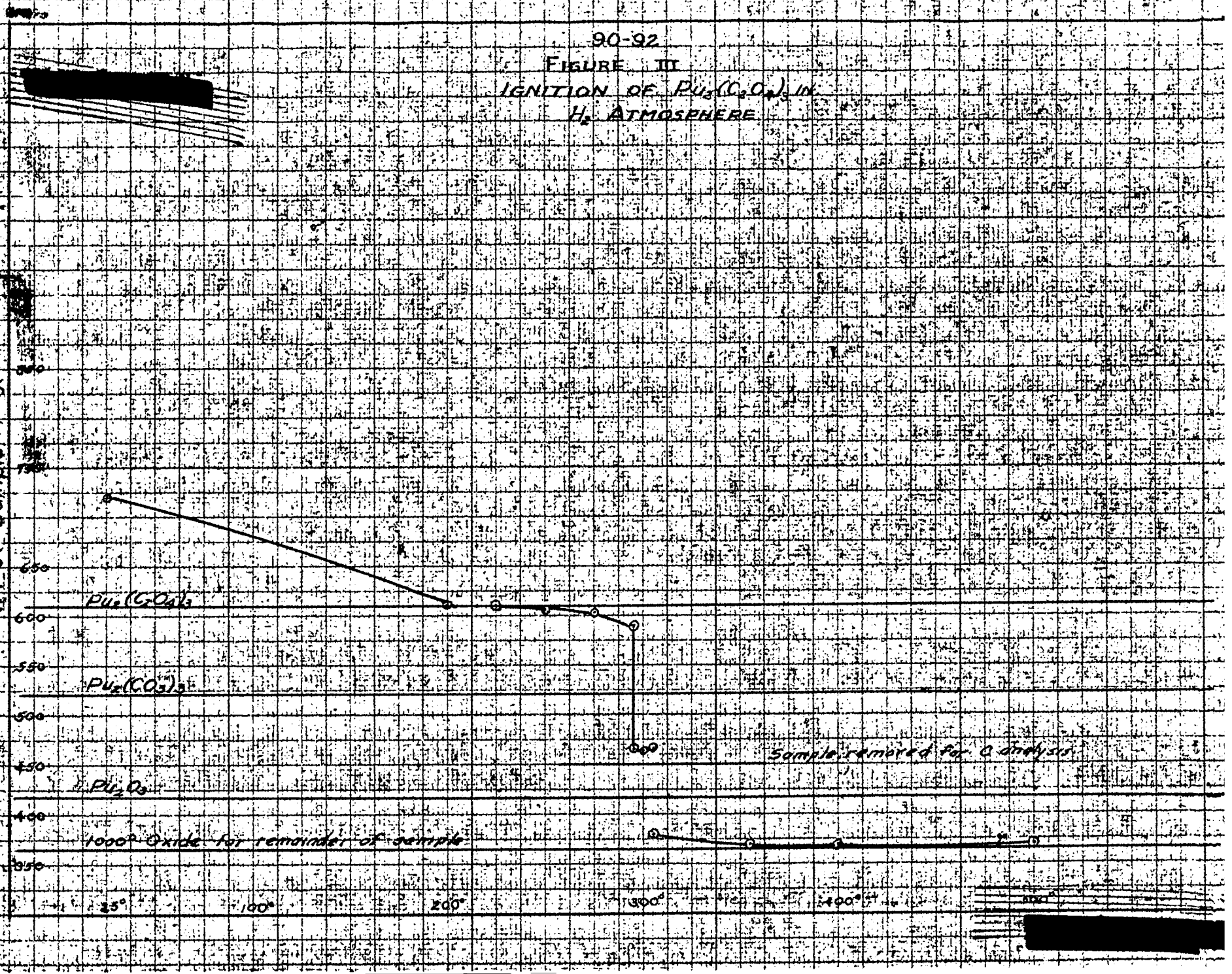




90-92

FIGURE III

IGNITION OF $Pu_2(C_2O_4)_3$ IN H_2 ATMOSPHERE



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