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**GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION**

**STUDIES ON O/M RATIO DETERMINATION IN URANIUM OXIDE,
PLUTONIUM OXIDE AND URANIUM-PLUTONIUM MIXED OXIDE**

by

**S. Sampath, K. L. Chawla and
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**BHABHA ATOMIC RESEARCH CENTRE
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ABSTRACT

Thermogravimetric studies were carried out in unsintered and sintered samples of uranium oxide, plutonium oxide and uranium-plutonium mixed oxide under different atmospheric conditions (air, argon and moist argon/hydrogen).

Moisture loss was found to occur below 200° C for uranium dioxide samples, upto 700° C for unsintered plutonium dioxide and negligible for sintered samples.

The O/M ratios for non-stoichiometric uranium dioxide (sintered and unsintered), plutonium dioxide and mixed uranium and plutonium oxides (sintered) could be obtained with a precision of ± 0.002 . Two reference states $UO_{2.000}$ and $UO_{2.656}$ were obtained for uranium dioxide and the reference state $MO_{2.000}$ was used for other cases. For unsintered plutonium dioxide samples, accurate O/M ratios could not be obtained because of overlap of moisture loss with oxygen loss/gain.

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1. INTRODUCTION

a) Effect of O/M ratio on properties of fuels

The ratio of oxygen to uranium and/or plutonium in the oxide, which will be referred to hereafter as O/M ratio, affects some of the properties of the fuels which are important in fuel performance and life. Lyon and Bailey⁽¹⁾ found that the melting point of mixed oxide fuel, which is important in determining maximum specific power for a given fast reactor design, increases as O/M ratio decreases from 2.06 to 1.97, the most rapid change occurring between O/M of 1.985 and 1.97. They reported that uncertainty in M.P. by $\pm 150^\circ$ C corresponds to a range of 20% in specific power. The conductivity of a mixed oxide fuel has also been found to be influenced by changes in O/M ratio. Thus thermal conductivity of sintered uranium-plutonium mixed oxide with an O/M of 1.95 or 2.05 is reported⁽²⁾ to be less than thermal conductivity of an oxide with a stoichiometric ratio of 2.00.

The oxygen potential around fuel derived from O/M and fuel temperature provides the chemical driving force for attack of stainless steel cladding⁽³⁾, controls chemical state of fission products⁽⁴⁾ and vapour pressure of fuel components and affects the potential for interaction of liquid sodium coolant with the fuel matrix⁽⁵⁾.

b) Determination of O/M - the method developed

A survey of methods potentially useful for determination of O/M ratio in mixed oxides of uranium and plutonium was made by Lyon⁽⁶⁾, Florence⁽⁷⁾ and Metz et al⁽⁸⁾.

Among gravimetric methods, the age-old method of igniting uranium oxide in air to U_3O_8 and determining O/U ratio from weight change has been found to lead to gross errors because of moisture in the sample and uncertainty in the composition of U_3O_8 . But recently Macgowan et al⁽⁹⁾ reported that stoichiometric U_3O_8 could be consistently obtained by controlled heating in air and M_3O_8 is satisfactory reference state for mixed oxides up to 10% PuO_2 . Drummond et al⁽¹⁰⁾ reported a thermogravimetric method making use of weight vs time plot obtained upon reduction in pure hydrogen at 1150°C from a sample which has been first heated in argon and then in hydrogen at 1000° C. A similar method was used by Lyon⁽⁶⁾ using 94% He - 6% H_2 and a temperature of 700° C for reduction.

Another thermogravimetric method described by Chikalla and Mansilly⁽¹¹⁾ is based on following thermodynamic considerations. The variation of $\Delta\bar{G}(O_2)$ vs O/M for UO_2 , PuO_2 and UO_2 - PuO_2 is given by Markin^(12, 13). The value of $\Delta\bar{G}(O_2)$ at an O/M of 2.00 varies from about -75 to -115 k.cals/mole O_2 at 800° C and is virtually independent of the composition of mixed oxide up to 30% PuO_2 . This is also proved from our data⁽¹⁴⁾ and Figures 1 and 2 show variation of $\Delta\bar{G}(O_2)$ vs O/M for different temperatures (800°, 1000°, 1200° C) and compositions

(30%, 20%, 10% PuO_2). These figures are derived from our calculations for mixed oxides based on the model proposed by Blackburn for UO_{2+x} (15). From these considerations, it would appear that a one-step heat treatment in an atmosphere having an oxygen potential in the above said range should produce a stoichiometric oxide that was initially hypo- or hyperstoichiometric. The method of Chikalla and Moneilly involves measuring the weight change when a mixed oxide sample is heated at 800°C for 6 hours in 92% argon/8% hydrogen gas mixture saturated with water vapour at a partial pressure of 4 torr obtained by passing the gas mixture through a water-trap kept at 0°C . These atmospheric conditions produce an oxygen potential of -100 k. cal/mole (16). Chikalla and Moneilly first suggested cooling of reduction product in the same flowing gas and later modified and suggested cooling in dry gas mixture.

Mogowan et al (9) made a comparison between reference states produced by (a) equilibration at 800°C with 4% hydrogen/argon saturated with water vapour at 0°C and 20°C , (b) oxidation followed by reduction at 1000°C with dry 4% hydrogen/argon and, (c) oxidation followed by reduction with dry 4% hydrogen/argon at 1150°C and found that O/M ratios of the product of equilibration deviated from stoichiometric $\text{UO}_{2.000}$ by only ± 0.001 .

In the method of Chikalla and Moneilly, the samples after equilibration in moist argon/hydrogen is cooled in dry gas mixture and O/M calculated from the weight change measured at room temperature.

This involves possibly two sources of errors in O/M. Care has to be taken so that moisture is removed without change in stoichiometry prior to equilibration. In Chickalla's method, after the sample is heated to 800° C in 92% Ar/8% H₂ mixture containing water vapour at 4 torr, the stoichiometric reference state is obtained. But this is cooled in dry 92% Ar/8% H₂ mixture. This implies sudden increase in $\frac{[H_2]}{[H_2O]}$ ratio, leading to reduction in $\Delta \bar{G}(O_2)$ not compensated by decrease of temperature. This is likely to cause change in reference state especially in PuO₂ and UO₂-PuO₂ samples. The sudden reduction in oxygen potential during cooling may lead to further loss of oxygen from MO_{2.00} in PuO₂ and UO₂-PuO₂ samples.

The aim of the present investigations based on above principle is (1) to arrive at the reference state, MO_{2.00} about which there is no doubt as to the composition, (2) to make sure that total conversion to the reference state is achieved, (3) to ensure that the change in weight corresponds solely to the change in oxygen content (all these in turn would give an O/M ratio with accuracy and precision), (4) to determine the exact composition of the reference state, U₃O_{8-x} which in turn can be used to determine O/M ratio in uranium oxide samples and, (5) to extend the investigations to plutonium oxides and uranium-plutonium mixed oxides.

2. EXPERIMENTAL

In this laboratory, ways of forming stoichiometric UO₂, PuO₂ and UO₂-PuO₂ by oxidation-reduction cycles and by equilibration in an atmosphere of appropriate oxygen potential are compared. The uranium

oxide samples were obtained from Uranium Metal Plant, the unsintered PuO_2 powder from Plutonium Plant and sintered PuO_2 and sintered $\text{PuO}_2\text{-UO}_2$ (crushed and whole pellets) from Radiometallurgy Section. The experiments were carried out in a Mettler Thermoanalyser having a readability of 5 ug in extended TG region, temperature range ambient to 1600°C and vacuum down to 10^{-5} torr. The reference state $\text{MO}_{2.00}$ was obtained for all samples as follows: about 200 mg of sample (~ 1 gm for whole pellets) were taken in platinum crucibles in the analyser, evacuated to 5×10^{-5} torr. and filled with the gas mixture of 92% argon/8% hydrogen. The composition of gas mixture was ascertained by controlling with flowmeters. The gases were purified from traces of oxygen by passing through bivalent vanadium solution. The purifying columns were prepared by adding zinc amalgam to vanadyl sulfate solution containing sulfuric acid. This has been used as an effective oxygen remover by Meites (17).

The purified gases were mixed in a water trap kept at 0°C and then passed through sample chamber. This ensured a partial pressure of water of 4 torr in the gas mixture. The sample chamber after filling up with the gas mixture was flushed for an hour and then heated to 800°C and kept constant at 800°C until constant weight is obtained.

For studying plutonium samples in the Mettler Thermoanalyser, a glove box was designed and fabricated. A photograph (Fig. 3) of the Mettler Analyser enclosed in the glove box is reproduced. The salient

features of the glove box are:

1. For easy access to various parts of the analyser, gloves are provided as much as possible.
2. A port hole is provided on the front for removal of the balance chamber and servicing if and when necessary.
3. The thermostat is fixed outside the glove box for easy servicing.
4. A double suction arrangement is made, with sufficient air inlet so that the air activity inside the box is kept minimum. This also effectively reduces heating of the glove box to the minimum when the furnace is heated to beyond 1000° C.

For taking in and taking out radioactive samples, Perspex Crucible holders were made, suitable for DTA and micro DTA (18).

3. RESULTS AND DISCUSSION

3.a) Studies in uranium oxides

The urania samples were subjected to the above equilibration experiments. A typical thermogram obtained during reduction of urania sample in moist 92% argon/8% hydrogen is shown in Fig. 4. The thermogram shows loss of moisture in the temperature range 25-200° C. Oxygen loss starts around 300° C and is completed at 650° C. The wt. remains constant upto 800° C, which corresponds to weight of stoichiometric dioxide.

The O/M of oxide was obtained from the expression

$$O/M = 2 + \frac{M(W_1 - W_2)}{16W_2}$$

M = mol. wt. of stoichiometric dioxide.

W₁ = wt. of sample after deducting moisture loss.

W₂ = weight after conversion to reference state.

The weight of the reference state was taken at 800° C after accounting for apparent weight gain due to buoyancy (corresponding to 0.645 mg up to 800° C for the atmospheric conditions employed). This was preferred over cooling the reduced sample in either dry 92% argon/8% H₂ or wet gas mixture which may introduce uncertainty in the composition of the reference state.

In another series of experiments, the uranium samples were heated in air to 800° C at a rate of heating of 4°/minute and kept constant at 800° C for 4 hours after which the sample was cooled and subjected to the reduction procedure.

The thermograms obtained during oxidation-reduction cycle are given in Fig. 5 and 6 respectively in a typical case.

Table 1 gives the data obtained from thermograms on moisture loss, oxygen loss and O/M ratios calculated for various samples together with the calculated precision.

Table 2 gives the data on O/M obtained from oxidation-reduction cycles.

Table 3 gives a comparison of data obtained from the two processes. It is found that O/M ratios could be obtained with a precision of ± 0.002 .

It was found that in all our experiments in controlled heating in air, that the composition of the product at 800° C remains constant

over two hours. This composition was obtained assuming the reduction product in oxidation-reduction cycle was $\text{UO}_{2.000}$ and the data is given in Table 4. The average composition of the oxidation product was found to be $\text{UO}_{2.656}$.

Our studies on uranium oxide show that O/M ratio of uranium oxide can be obtained with high precision. The moisture loss is distinctly separated from oxygen loss and a reproducible reference state is obtained.

3.b) Studies on unsintered PuO_2 powder

Five PuO_2 powder samples obtained from Plutonium Plant were heated in air, argon and moist argon/hydrogen upto 1200°C . A typical weight loss curve of a plutonium oxide sample in argon is shown in Fig. 7. The figure shows steep weight loss upto about 200°C (the loss upto 200°C is 70% of the total loss observed). The loss of water is not complete upto 700°C . The total loss is about 2% of weight of sample.

Weight change due to oxygen could not be separated from moisture loss and Table 5 gives the apparent O/M ratios of PuO_2 samples obtained in air, argon and moist argon/hydrogen. In this data the stoichiometry of PuO_2 in moist argon/hydrogen at 800°C is assumed as $\text{PuO}_{2.000}$.

Our studies on unsintered PuO_2 powder showed:

1. The reference state is reproducible in moist argon/hydrogen at 800°C .

2. Since moisture loss is not separated from weight change due to oxygen, accurate O/M ratios cannot be obtained for unsintered PuO_2 powders.
3. Fig. 8 shows behaviour of sample in moist argon atmosphere. The sample was heated to 400°C (Fig. 8a) cooled to room temperature in moist argon, reheated to 800°C (Fig. 8b). Cooled again and reheated (Fig. 8c). This shows that samples heated to 400°C and cooled pick up significantly more amount of moisture compared to samples heated to 800°C and cooled.
4. Our studies on heating and cooling in the temperature range $400\text{--}800^\circ\text{C}$ in steps of 50°C showed that upto 500°C the samples pick up on cooling the same amount of water lost in heating in any temperature range.

3.e) Studies on sintered PuO_2 and sintered $\text{UO}_2\text{-PuO}_2$ samples

A number of PuO_2 sintered samples, 2% $\text{PuO}_2\text{-UO}_2$ samples and 30% $\text{PuO}_2\text{-UO}_2$ crushed pellets and whole pellets were analysed for O/M ratio. In these cases the samples were first heated in dry argon upto 400°C and then in moist argon/hydrogen upto 800°C .

Figures 9, 10 and 11 show the behaviour of sintered PuO_2 , 2% $\text{PuO}_2\text{-UO}_2$ and 30% $\text{PuO}_2\text{-UO}_2$ samples. The reference state was very steady. The amount of water lost was very small (except in 2% $\text{PuO}_2\text{-UO}_2$ samples) and hence did not create any problem in calculating O/M ratio.

4. SUMMARY

Our gas equilibration studies in moist argon/hydrogen on UO_2 , PuO_2 and $\text{UO}_2\text{-PuO}_2$ have shown that it is necessary to take the

reference state as $\text{UO}_{2.00}$ at 800°C (and not the cooled product in dry or wet gas mixture as recommended by Chickalla). This ascertains the composition of the reference state which in turn determines the accuracy of O/M ratio obtained. Total conversion to the reference state is obtained by heating at 800°C until weight remains constant (in-situ measurements).

For uranium oxides, moisture loss occurs below 200°C , whereas oxygen loss is observed above 300°C . For unsintered plutonium oxides, moisture loss is noticed upto at least 700°C . For sintered uranium-plutonium mixed oxides, moisture loss is small as expected. Hence weight change due to oxygen can be obtained accurately for uranium oxides and for sintered uranium-plutonium mixed oxide.

For uranium oxide the composition of the reference state U_3O_{8-x} was determined from our studies in oxidizing atmospheres to be $\text{UO}_{2.656}$. This, we have found, also can be used to determine O/M ratio.

Recently, Blackburn has calculated oxygen potential from O/M and temperature for uranium assuming equilibrium between U^{2+} , U^{4+} and U^{6+} in the fuel. We are attempting to calculate oxygen potential for mixed oxide fuels from O/M and temperature.

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Table 1

DATA ON O/M RATIO OBTAINED BY REDUCTION IN 92% ARGON/8% HYDROGEN

Sample No.	Run No.	Amount of sample	Moisture loss	Oxygen loss	O/M ratio	Average O/M ratio	Standard deviation
Sample I	1	189.30	0.48	1.08	2.097	2.097	
	2	198.52	0.51	1.12	2.097		
Sample II	1	196.25	0.49	0.975	2.083	2.084	0.0016
	2	200.10	0.51	0.99	2.084		
	3	198.45	0.505	0.985	2.082		
	4	195.32	0.47	0.90	2.086		
	5	106.45	0.50	0.99	2.084		
Sample III	1	198.10	0.49	0.98	2.084	2.084	0.0007
	2	203.40	0.52	1.005	2.084		
	3	187.52	0.47	0.92	2.083		
Sample IV	1	202.5	0.56	2.25	2.105	2.105	0.0017
	2	200.15	0.55	1.23	2.104		
	3	195.85	0.53	1.20	2.103		
	4	205.40	0.56	2.285	2.107		
Sample V	1	200.80	0.81	2.35	2.115	2.115	0.0016
	2	206.50	0.84	1.33	2.114		
	3	200.75	0.82	1.37	2.117		
Sample VI	1	197.55	0.45	1.345	2.116	2.1165	0.001
	2	199.00	1.370	2.118	2.116		
	3	201.45	0.40	1.365	2.116		
	4	184.05	0.43	2.280	2.116		
Sample VII	1	199.40	0.79	1.22	2.104	2.1035	
	2	205.05	0.81	1.24	2.103		

(contd.)

(Table 1 Contd.)

Sample No.	Run No.	Amount of sample	Moisture loss	Oxygen loss	O/M ratio	Average O/M ratio	Standard deviation
Sample VIII	1	204.25	0.008	0.80	2.065		
UO ₂ sintered	2	201.45	0.008	0.77	2.065	2.065	0.0007
	3	195.80	0.07	0.73	2.064		
UO ₂ sintered	1	2.0015 g	0.24	0.65	2.009		
Sample IX	2	200.45 mg	0.05	0.11	2.009	2.009	0.0007
	3	195.50 mg	0.04	0.095	2.010		
UO ₂ powder sample X		194.6	0.27	-0.68	2.059		
UO ₃ powder Sample XI		341.45	0.40	-12.70	2.653		

Table 2

DATA ON O/M RATIO FROM OXIDATION-REDUCTION CYCLE

Sample No.	Run No.	Amount of sample (mg)	Moisture loss (mg)	Oxygen gain in air (mg)	Oxygen loss (Ar/H ₂) (mg)	Net loss (mg)	O/M ratio	Average O/M	Standard deviation
Sample I	1	202.90	0.42	6.68	7.96	1.18	2.098	2.097	0.0002
	2	105.85	0.40	6.45	7.59	1.13	2.098		
	3	200.50	0.43	5.59	7.74	1.16	2.098		
Sample II	1	205.70	0.84	6.95	7.96	1.01	2.082	2.081	0.0007
	2	201.45	0.75	6.80	7.76	0.98	2.081		
	3	203.85	0.70	6.87	7.86	0.99	2.081		
Sample III	1	25.40	0.40	6.89	7.94	1.05	2.086		
Sample IV	1	199.55	0.39	5.50	7.72	1.22	2.104	2.104	0.0007
	2	199.50	0.41	6.495	7.715	1.22	2.104		
	3	202.45	0.41	6.56	7.80	1.24	2.105		
Sample V	1	199.90	0.34	6.38	7.74	1.35	2.116	2.116	
	2	201.20	0.40	6.30	7.67	1.37	2.116		
Sample VI	1	202.70	0.35	6.35	7.77	1.42	2.118	2.118	0.0007
	2	192.40	0.31	6.11	7.42	1.31	2.117		
	3	195.55	0.32	6.21	6.54	1.33	2.110		
Sample VII	1	200.05	0.45	6.51	7.75	1.24	2.106	2.106	0.0007
	2	211.15	0.19	6.84	8.14	1.30	2.106		
	3	198.40	0.05	6.44	7.66	1.22	2.105		
Sample VIII	1	201.30	0.05	7.02	7.88	0.76	2.064	2.064	0.001
	2	199.85	0.05	6.97	7.74	0.77	2.065		
Sample IX	1	220.45	0.025	8.35	8.51	0.16	2.011		

Table 3

COMPARISON OF O/M RATIO FROM REDUCTION METHOD
AND OXIDATION-REDUCTION METHOD

Sample number	O/M ratio got from $UO_{2+x} \xrightarrow{\quad} UO_{2.000}$	O/M ratio got from $UO_{2+x} \xrightarrow{\text{Air}} U_3O_8-y \xrightarrow{\quad} UO_{2.00}$ (y = 0.032)
I	2.097	2.097
II	2.084	2.081
III	2.084	2.085
IV	2.105	2.104
V	2.115	2.116
VI	2.165	2.118
VII	2.1055	2.106
VIII	2.065	2.064
IX	2.009	2.007

Table 4

COMPOSITION OF OXIDATION PRODUCT AT 800°C IN AIR

Sample	Run No.	Amount of $UO_{2.00}$ (mg)	Weight loss during $UO_{2.66-y} \rightarrow UO_{2.00}$ (mg)	Composition of oxidation product (O/M)
I	1	201.30	7.06	2.659
	2	194.32	7.58	2.6585
	3	198.91	7.74	2.6565
II	1	203.85	7.96	2.659
	2	199.72	7.78	2.658
	3	201.66	7.86	2.658
				Average O/M: 2.656
III	1	203.95	7.94	2.657
IV	1	197.80	7.72	2.659
	2	200.67	7.80	2.658
	3	197.80	7.72	2.659
V	1	198.20	7.74	2.659
	2	199.43	7.67	2.649
VI	1	200.93	7.77	2.653
	2	190.78	7.42	2.656
	3	198.90	7.54	2.656
VII	1	198.26	7.75	2.659
	2	199.36	8.14	
	3	196.73	7.66	2.657
VIII	1	204.61	7.94	2.653
	2	200.49	7.78	2.655
	3	199.03	7.74	2.656
IX	1	200.26	8.51	2.651

Table 5

UNSINTERED PLUTONIUM DIOXIDE SAMPLES

O/M ratios (without deduction of moisture and volatile impurities) of unsintered PuO_2 samples obtained under three different conditions assuming the stoichiometry of plutonium dioxide in Ar/H_2 at 800°C as $\text{PuO}_{2.000}$

Temperature	O/M ratio		
	Moist argon/hydrogen	Argon	Air
<u>Sample I</u>			
1200° C			2.000
1000° C		2.0005	2.014
800° C	2.000 (assumed)	2.025	2.035
Room temperature	2.163	2.162	2.180
<u>Sample II</u>			
1200° C			2.000
1000° C		2.000	2.013
800° C	2.000 (assumed)	2.015	2.026
Room temperature	2.269	2.270	2.268
<u>Sample III</u>			
1200° C			2.000
1000° C		2.000	2.009
800° C	2.000 (assumed)	2.012	2.019
Room temperature	2.290	2.297	2.289
<u>Sample IV</u>			
1200° C			2.000
1000° C		2.000	2.018
800° C	2.000 (assumed)	2.009	2.018
Room temperature	2.391	2.393	2.389

Table 6
**O/M RATIOS IN SINTERED PLUTONIUM OXIDES AND
 MIXED PLUTONIUM URANIUM OXIDE SAMPLES**

S. No.	Sample	Sample wt. (mg)	Moisture loss (mg)	Oxygen gain/loss (mg)	O/M ratio
1	PuO ₂ sintered sample I	189.42	0.15	+ 0.85	1.942 In
	II	200.01	0.16	+ 0.98	1.940 duplicate
2	PuO ₂ sintered sample	200.13	0.13	+ 0.46	1.961
3	30% PuO ₂ -UO ₂ sample	207.95	0.03	+ 0.24	1.981
4	2% PuO ₂ -UO ₂ sample I	209.43	0.28	- 0.29	2.022 In
	II	201.57	0.32	- 0.23	2.020 duplicate
5	30% PuO ₂ -UO ₂ sample I	200.6	0.04	- 0.09	2.008 In
	II	211.6	0.04	- 0.10	2.010 duplicate
6	30% PuO ₂ -UO ₂ sample	212.05	0.02	+ 0.08	1.996
7	30% PuO ₂ -UO ₂ sample	220.39	0.02	+ 0.15	1.993
8	30% PuO ₂ -UO ₂ sample 2	177.55	0.02	+ 0.52	1.956
9	30% PuO ₂ -UO ₂ sample	260.76	0.02	+ 0.62	1.960
10	30% PuO ₂ -UO ₂ sample	201.16	0.04	+ 0.37	1.969
11	30% PuO ₂ -UO ₂ sample	170.86	0.06	+ 0.25	1.975
12	30% PuO ₂ -UO ₂ sample	183.50	0.07	+ 0.23	1.989
13	30% PuO ₂ -UO ₂ sample	192.55	0.08	+ 0.20	1.983
14	30% PuO ₂ -UO ₂ whole pellet I	1.12296 g	0.02	+ 1.07	1.984 In
	II	1.0545 g	0.04	+ 0.96	1.983 duplicate
15	30% PuO ₂ -UO ₂ whole pellet I	1.42182 g	0.02	+ 2.24	1.973 In
	II	0.96917 g	0.02	+ 1.59	1.972 duplicate
16	30% PuO ₂ -UO ₂ whole pellet I	1.14989 g	-	+ 2.29	1.966 In
	II	0.8451 g	0.02	+ 1.70	1.966 duplicate

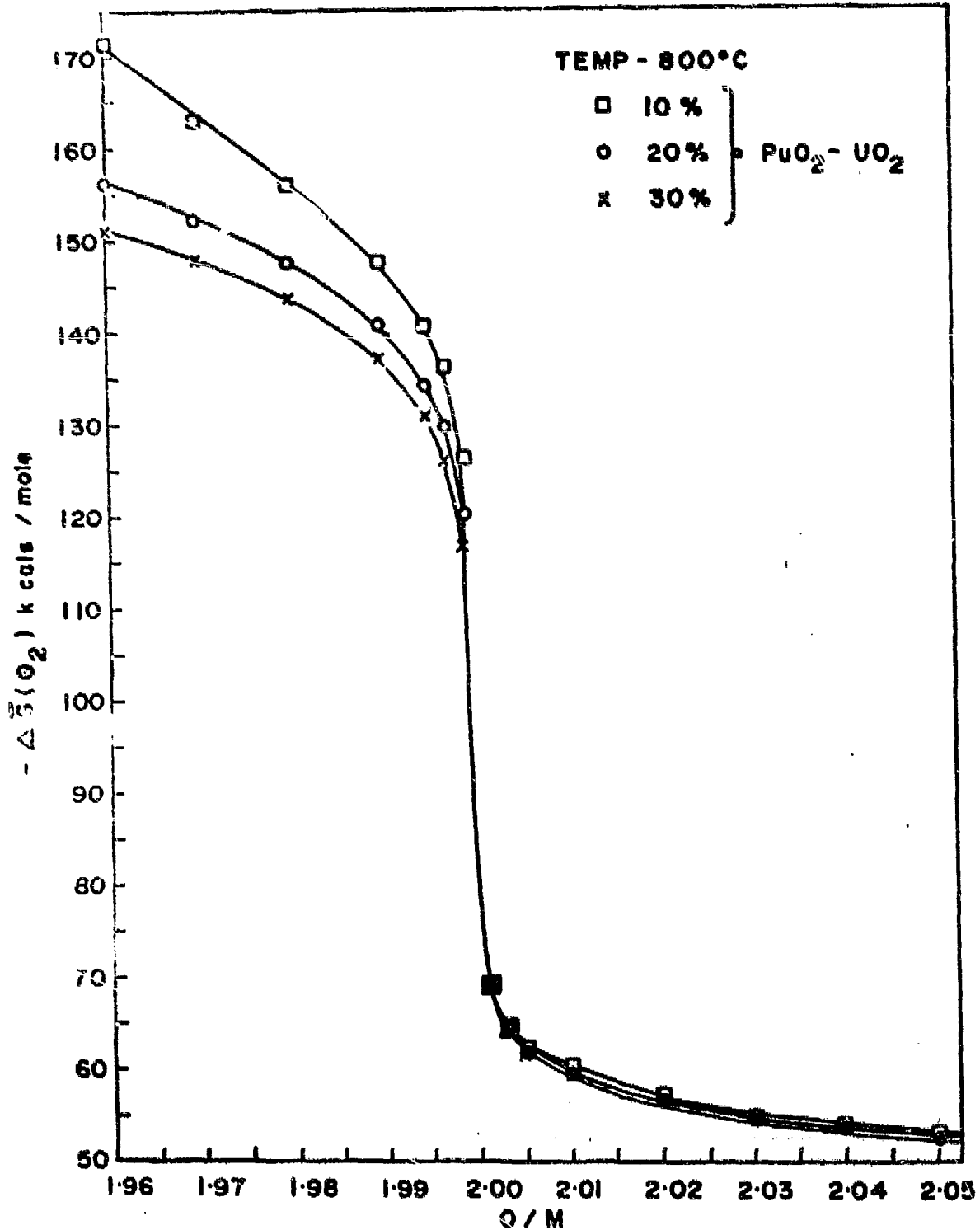


FIG.- 1. PLOTS OF $\Delta \bar{G}(\text{O}_2)$ Vs O/M

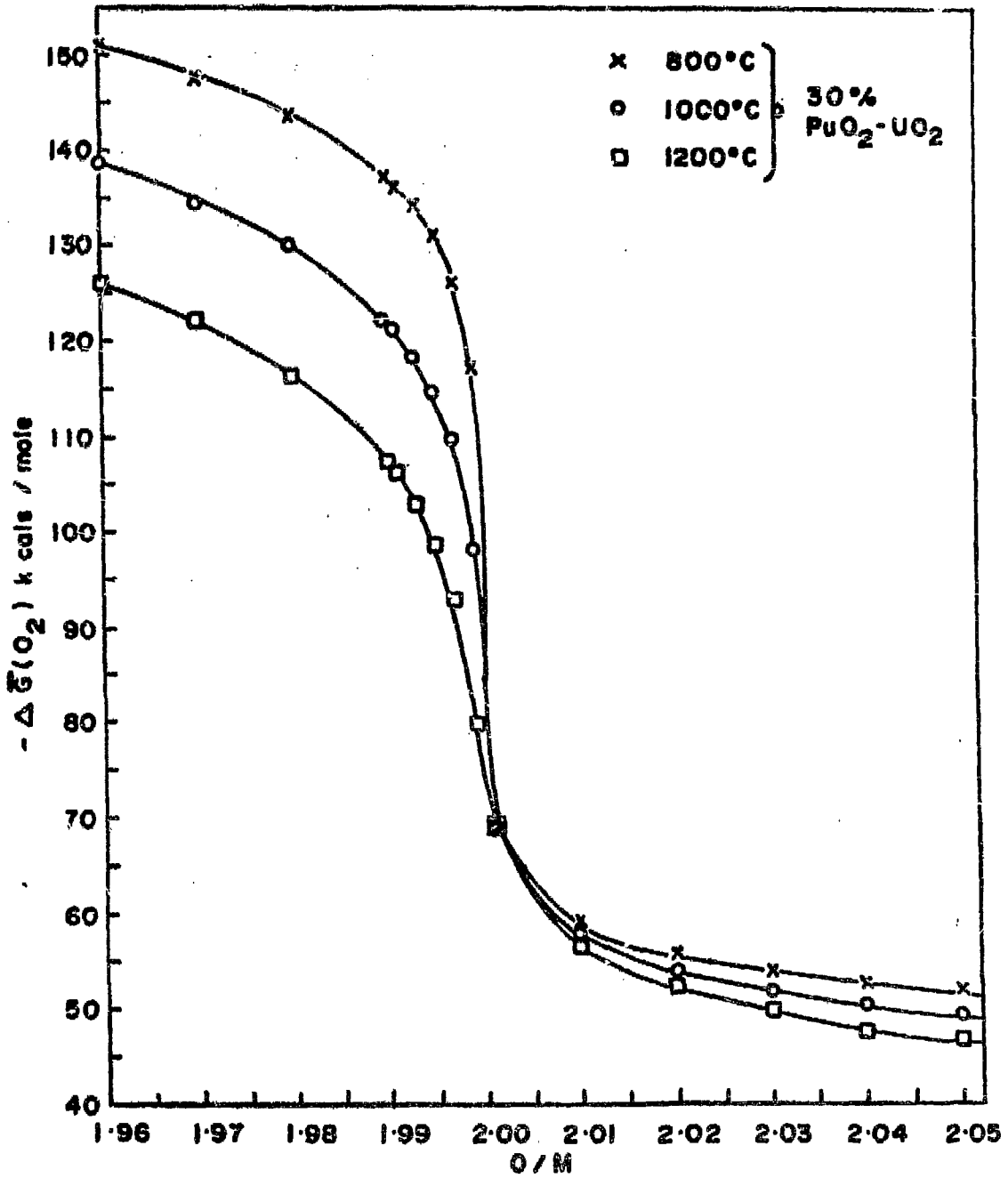


FIG.-2. PLOTS OF $\Delta\bar{G}(O_2)$ Vs O/M

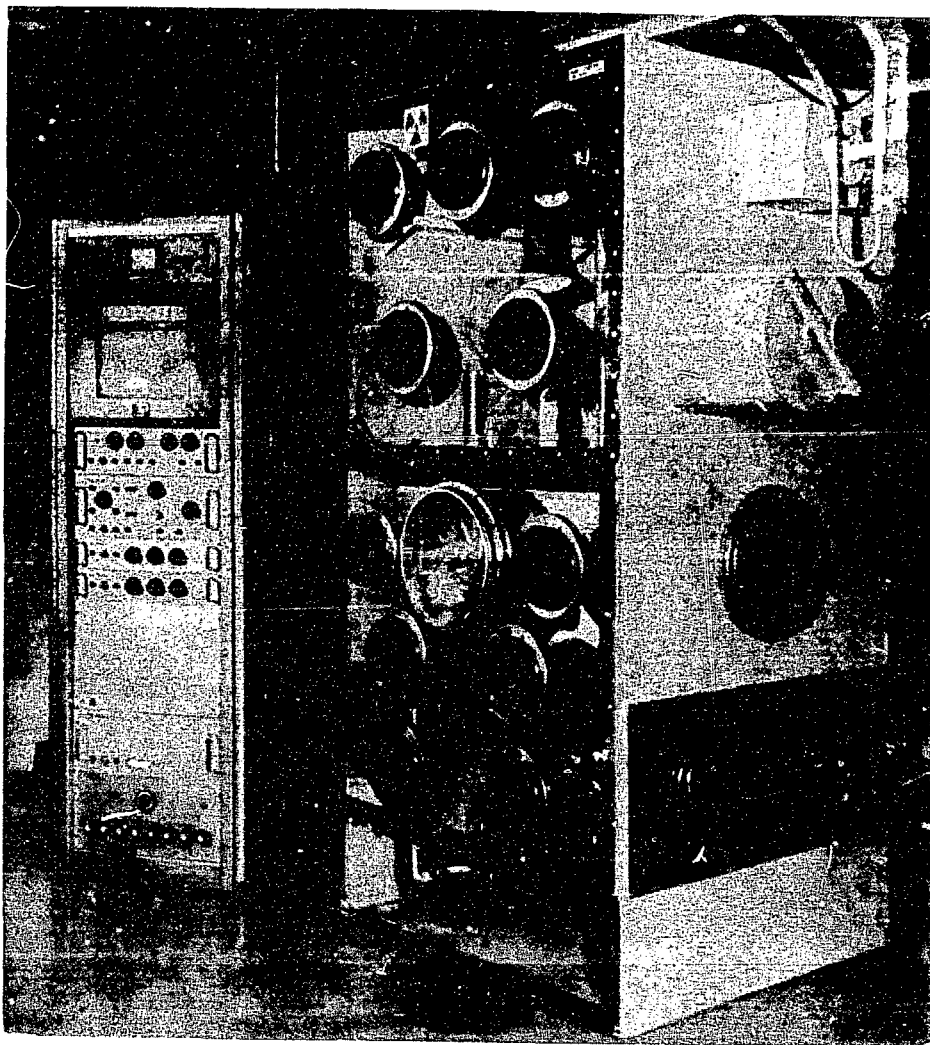


Fig. 3 Glove box assembly for the thermoanalyzer

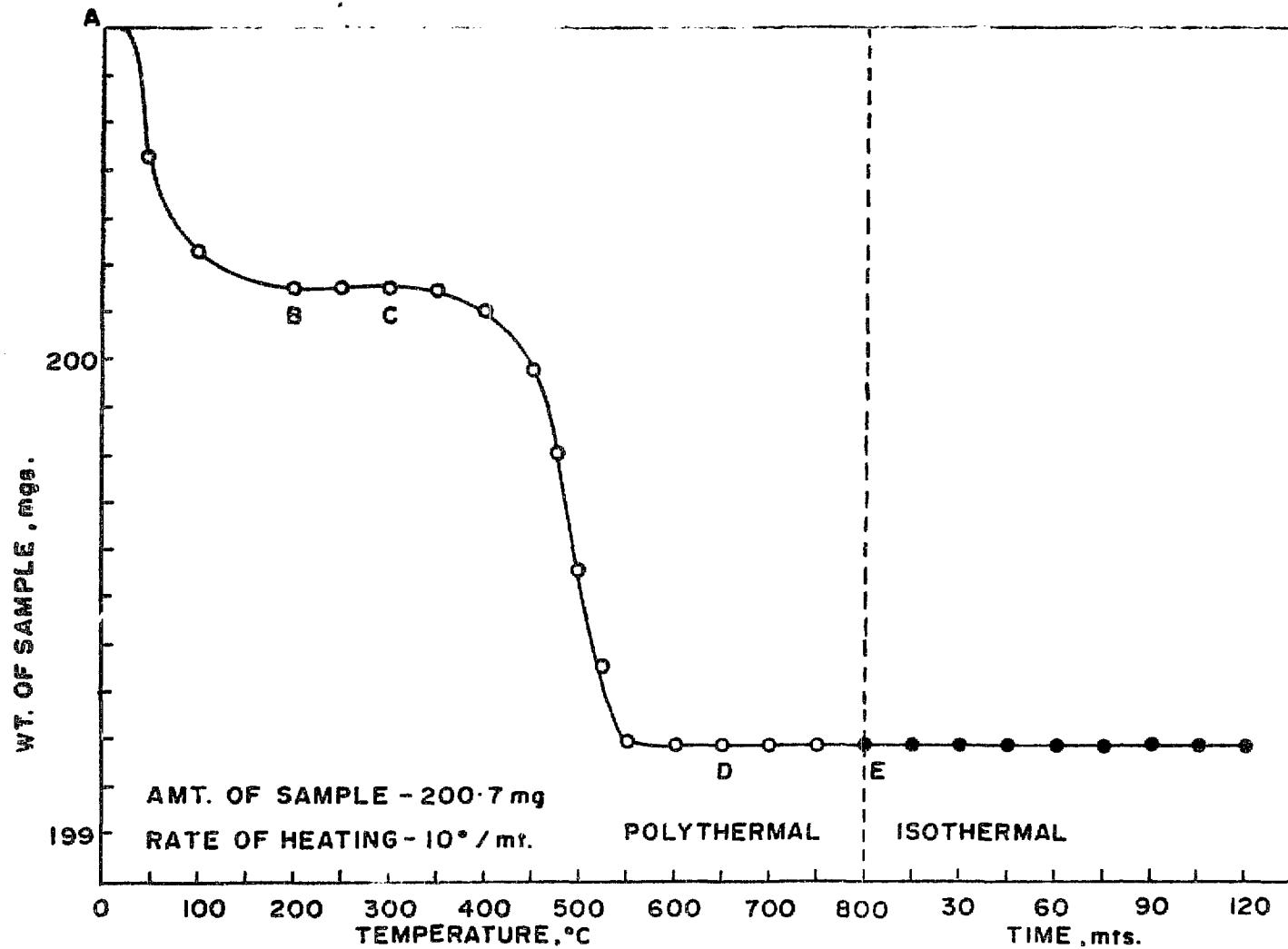


FIG.-4. TG CURVE OF UO_{2+x} IN 92% Ar / 8% H_2

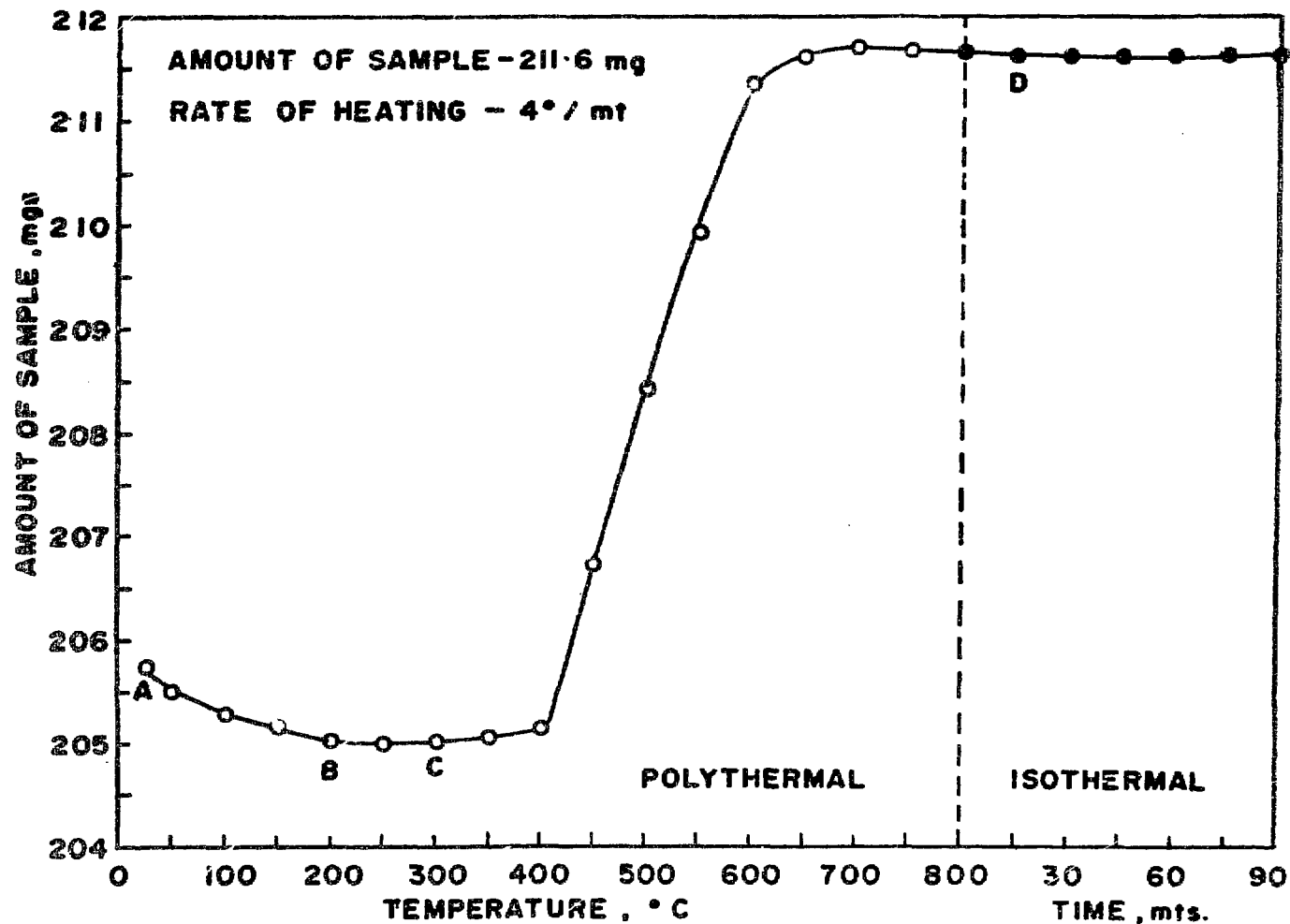


FIG - 3. TG CURVE OF UO_{2+x} IN AIR

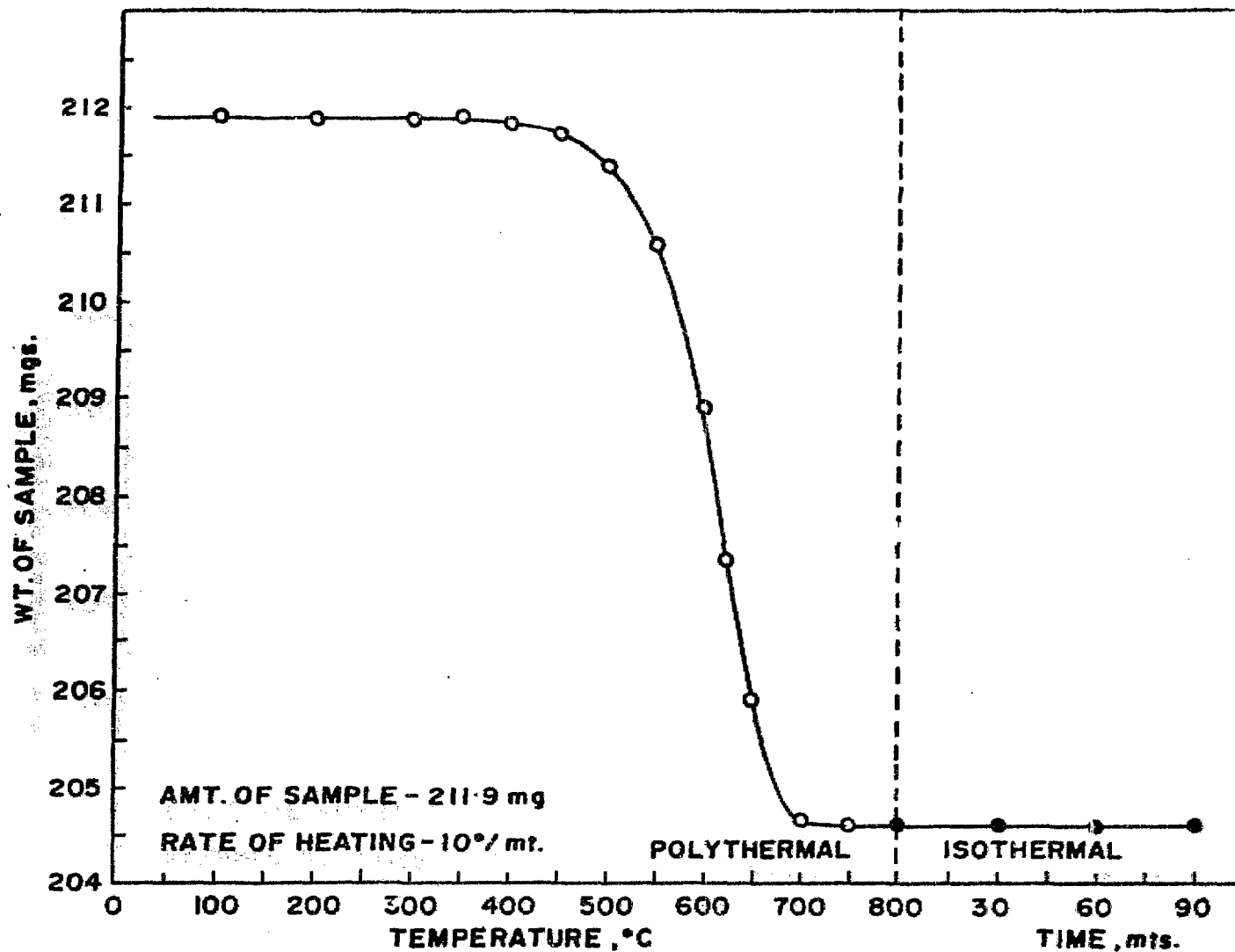


FIG.-6. TG CURVE OF U_3O_{8-x} IN 92% Ar / 8% H_2

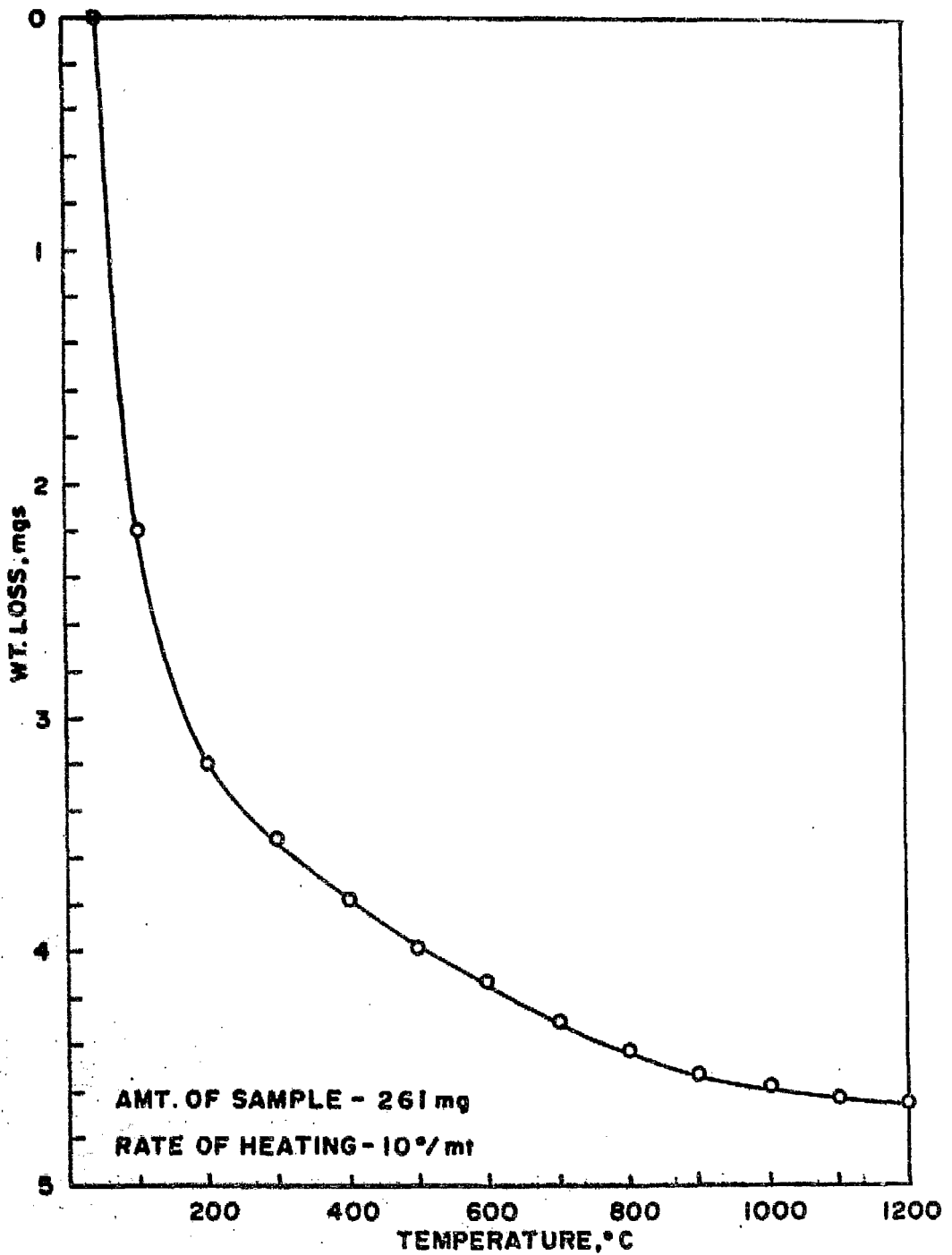


FIG.-7. WEIGHT LOSS BEHAVIOUR OF PLUTONIUM OXIDE POWDER IN DRY ARGON.

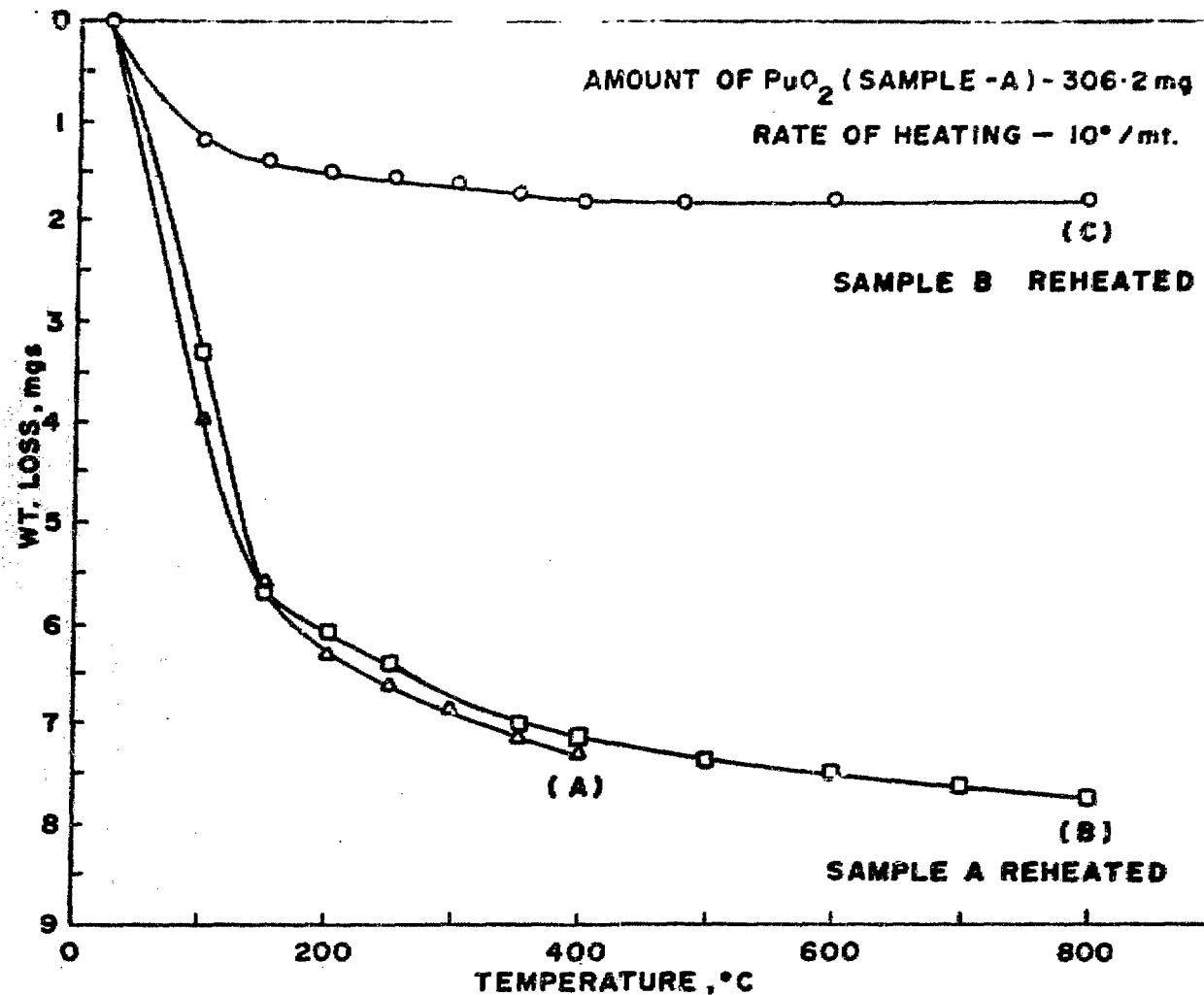


FIG.-8. WEIGHT LOSS BEHAVIOUR OF PLUTONIUM OXIDE IN MOIST ARGON.

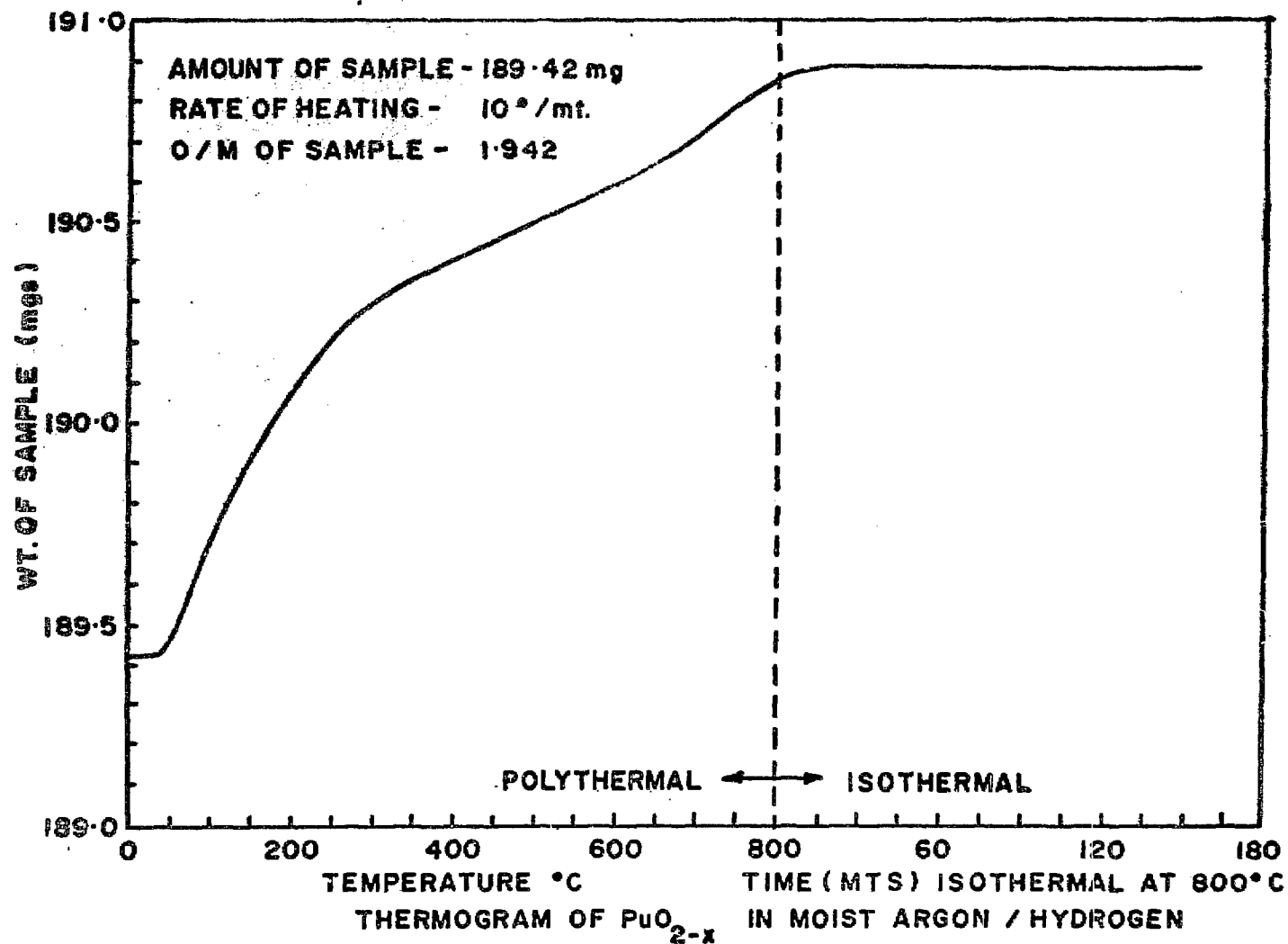


FIG.- 9.

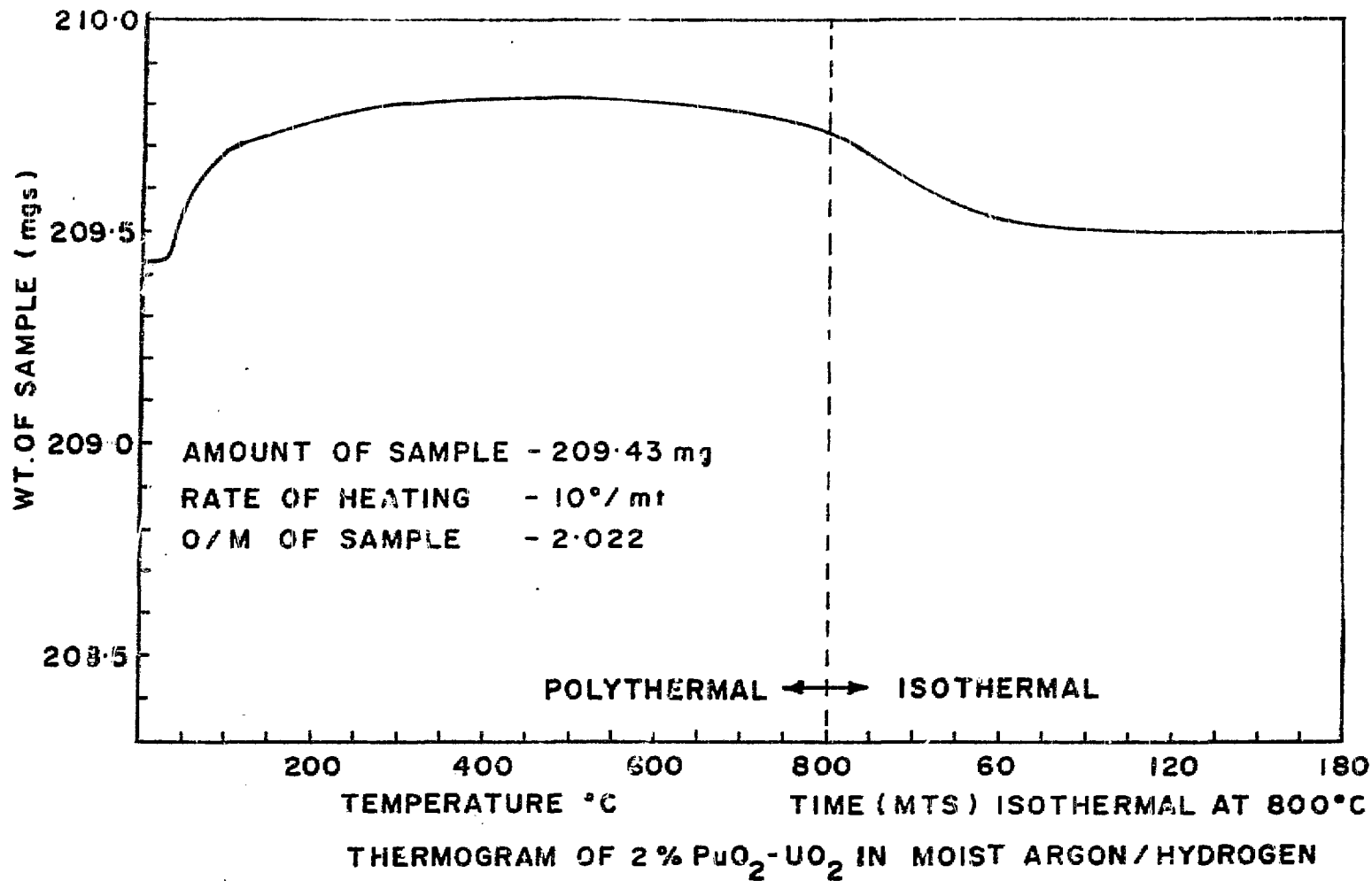


FIG.-10.

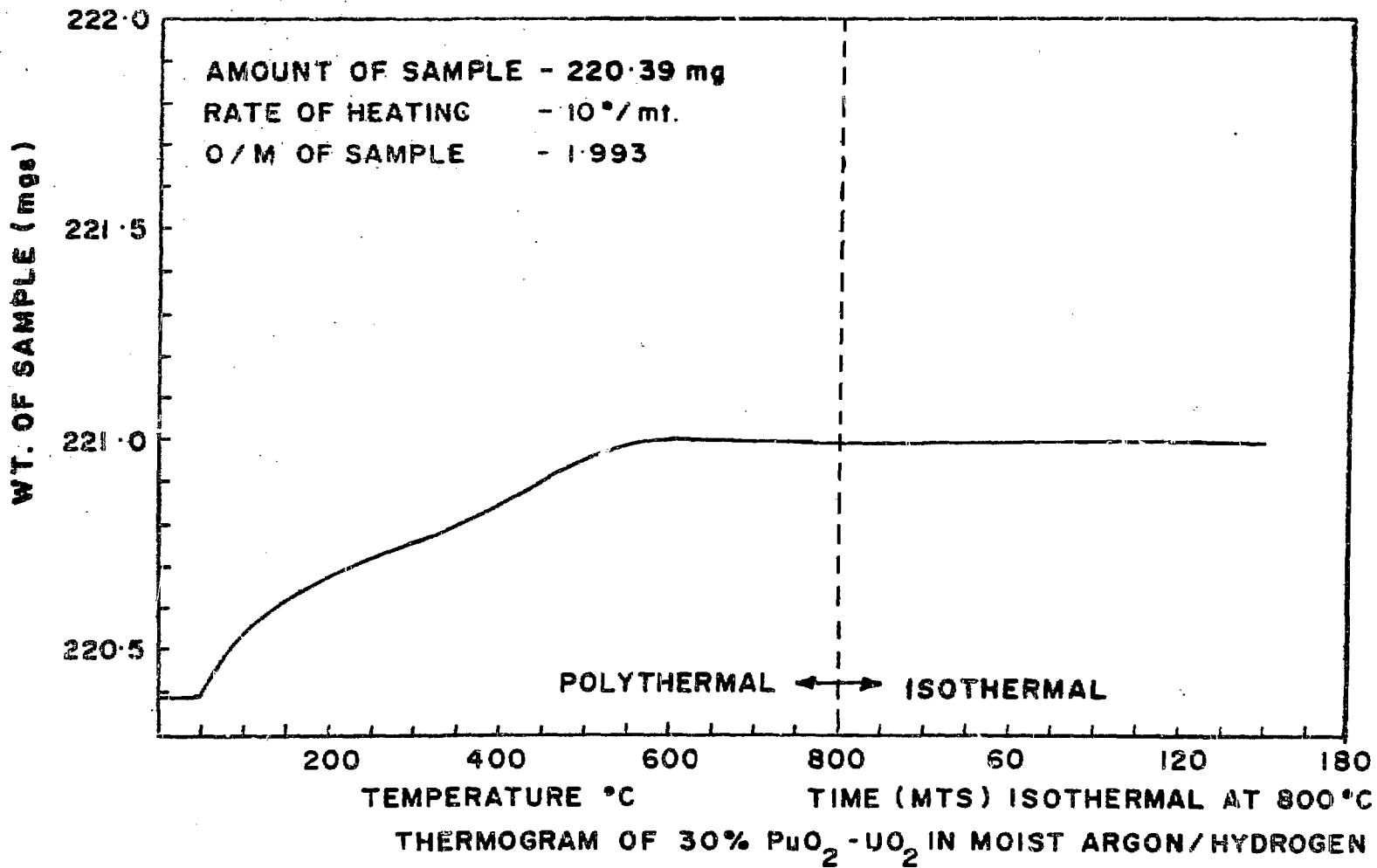


FIG.-II.