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PROJECT 9536

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Route List

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DATE 10-10-46

SUBJECT Pile Chemistry ~~and General~~
Week Ending October 9, 1946
WEEKLY ABSTRACTS Section C-II

TO File

FROM O. C. Simpson

COPY NO. [redacted]

A. B. Greninger

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BY JP Derouin DATE 2-16-82

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JE Savely 4/2/03
JE Shreve 4/23/03

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ANL-OCS-69
This document consists
of 11 pages, and
figures. [redacted]

WEEKLY ABSTRACTS October 10, 1946

Section C-II

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IV.
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Pile Chemistry and General Chemistry

Week Ending October 9, 1946

HIGH TEMPERATURE PILE PROGRAM

(C. A. Boyd)

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Reaction of Steam and BeO (M. G. Berkman)

Experiments were carried out to determine the effect of varying water vapor pressure on the rate of reaction between BeO and steam. Steam (generated from degassed water) was passed over a BeO pellet kept at 1400°C. An oil pump was used to obtain the desired pressure.

Run	Temp. of BeO Pellet (°C)	Rate of Steam Condensation (ml/min)	Water Vapor Pressure (mm Hg)	Weight of BeO Lost (%)
43	1400	0.4	130	0.40
44	1400	0.3	70	0.40

Additional experiments will be carried out to obtain more data on the effect of water vapor pressure on the reaction between BeO and steam.

Crushing Strength of Neutron Irradiated BeO and BeO-UO_2 Samples (S. R. Gaarder, J. L. Weeks, D. H. Rich)

Crushing strength determinations have now been completed for all the types of 1/4 inch pellets in the 63 day Hanford bombardment. The results are shown in Table I. Included for comparison are the values obtained from the twenty-four day irradiated material.

Inspection of the data for the mixed oxide pellets shows that in every case the crushing strength has shown an increase over the values for the twenty-four day bombardment. This may be due to uncertainties in the measurements. Certainly no marked effects were produced by the longer bombardment. It may indicate an approach to a saturation value. This same trend was noticed in connection with linear dimension changes of the mixed oxide samples (average expansion went from 0.56% in twenty-four days to 0.60% in sixty-three days - see abstract of September 25, 1946).

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The results of the tests on the pure BeO pellets are also erratic but suggest that the crushing strength of pure BeO is not affected appreciably by neutron irradiation. Expansion of this material was also slight - less than 0.3% (see abstract of September 25, 1946).

EFFECT OF NEUTRON IRRADIATION ON THE CRUSHING STRENGTH OF $BeO-UO_2$ AND BeO

	Norton Lot No. & Description	Crushing Strength (in psi)		
		Unirradiated Standards	24 Days Irradiation	63 Days Irradiation
Mixed Oxide	II-90 2% UO_2 Low Density (2.744)	57,300	53,900 (94%)	75,600 (133%)
	II-90 2% UO_2 High Density (2.789)	131,700	106,600 (81%)	115,500 (88%)
	II-89 10% UO_2 Low Density	155,900	105,200 (68%)	109,000 (70%)
	II-89 10% UO_2 High Density (3.048)	188,300	137,200 (73%)	144,100 (77%)
Pure BeO	H-78 Pure BeO High Density (2.885)	168,700	172,700 (102%)	164,700 (98%)
	H-79 Pure BeO High Density (2.913)	171,900	205,800 (120%) 183,300 (107%)	195,600
	II-65-1 Pure BeO Low Density (2.821)			84,400
	II-65-2 Pure BeO Low Density (2.891)		124,900	166,700

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Graphite Impregnation (P. F. Dinsmore)

All of the organic solvents that have been used for impregnating baths possess the disadvantage that the solutions of uranyl nitrate are unstable. The aqueous solutions used heretofore have the disadvantage that the uranyl nitrate hexahydrate tends to melt and come to the surface of the graphite before decomposing to the oxide. However, all of the aqueous solutions were saturated solutions. It was thought that possibly by using a dilute aqueous solution and thus depositing less uranyl nitrate in the graphite, the uranyl nitrate could lose its water of crystallization before melting and thus it would decompose before coming to the surface. This was tested this week.

The samples were impregnated according to the procedure in ANL-OCS-87 except that a water solution of uranyl nitrate was used (34.5 g of uranyl nitrate hexahydrate per 100 cc of solution) instead of an organic solvent. After impregnation the samples were dried in a vacuum instead of in air. One set of samples was refluxed for ten minutes and increased in weight by 1.7%. A second set was refluxed for thirty minutes and was not dipped in dibutyl cellosolve. These samples increased in weight by 1.7% also. These samples had very little uranyl nitrate coming to the surface before decomposing.

It should be pointed out that these samples increased in weight by only 1.7% while samples impregnated from an organic solvent bath of the same uranium concentration increased in weight by 2.5 to 3.0%. This lower weight increase may be due to one of two causes. It may be that aqueous solutions do not penetrate the graphite as far as do organic solutions. The other possibility is that not all of the water is removed from the sample before it is fired. This water would then react with the graphite and thus some graphite is lost. These possibilities will be investigated next week.

Work is continuing on a report of the program.

Fission Product Diffusion (J. E. Wilson)

Three hour diffusion measurements at 1200°C. on 3.1 density beryllia gave 0.32, 0.40, and 0.56% of xe lost. These results can be compared with earlier results at other temperatures.

Temperature, °C.	Percent xenon lost	D, $cm^2/min.$
1000	0.1	1×10^{-10}
1200	0.4	2×10^{-9}
1450	1.4	2×10^{-8}

These points are not really sufficient to determine the temperature dependence function, but they satisfy the following equation rather well:

$$D = (.060)\, e^{-\frac{5,000}{RT}}$$

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This may be interpreted to mean that the energy of activation for diffusion is 5,000 cal; this can be compared with 32,000 cal for argon diffusing through silica glass.

<u>Irradiation Effects on Elastic Modulus (L. Bupp)</u>

Measurements using the remote control apparatus have been completed on pure BeO prisms irradiated for a 63 day period. The results appear in the accompanying table together with those obtained on 24 day irradiated material.

It will be noted that high and low density react similarly showing a relatively insignificant change in modulus of the same order of magnitude as found with 24 day irradiation.

The modulus apparatus is being transferred to a mobile cabinet constructed for that purpose. Measurements will then be attempted on mixed oxide prisms from the 63 day irradiation.

(LDLO=low density, light oxide; HDLO=high density, light oxide)

Sample No.	Modulus E Initial 10^{10} dynes/cm^2	 Final 10^{10} dynes/cm^2	% Change
	24 Day Irradiation		
LDLO-24			
B-38	283.59	280.81	-0.98
B-39	285.60	281.32	-1.50
B-40	278.22	275.97	-0.81
B-41	288.72	281.94	-2.35
	63 Day Irradiation		
LDLO-28			
B-37	263.63	259.78	-1.12
B-42	275.46	270.94	-1.64
B-43	270.84	269.26	-0.58
B-44	282.85	278.80	-1.43
HDLO-28			
B-27	341.89	340.33	-0.46
B-29	342.77	342.99	+0.07 ?
B-30	325.92	320.05	-1.82
B-31	331.18	328.40	-0.84
HDLO-28			
B-6	353.44	348.53	-1.39
B-7	348.22	347.81	-0.12
B-8	349.50	344.57	-1.41
B-9	350.21	345.99	-1.21

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Mockup (M. P. Walling, G. R. Boyd)

Two runs have been made in which the mockup was operated at power levels simulating 2400 and 1200 KW respectively in a pile comprising 505 channels 8 1/2 feet long. These runs were discontinued after fairly short periods of steady-state operation (24 and 30 hours respectively) because of variac failures. However, equilibrium data were obtained on temperature distributions through the mock-up channels and pressure drops. In addition, certain data relating to the stability of the pile materials (BeO-UO_2 and BeO) will be obtained by post-mortem examination of the fuel tubes and bricks which were used in the mock-up tests. This will include dimensional changes, uranium loss, and mechanical changes (development of cracks, change in crushing strength, etc.). No deterioration of either the fuel tubes or the bricks was detected in a cursory autopsy.

Preparations are under way at present to ship the mockup and accessory equipment to Oak Ridge where it will be operated (probably after certain changes have been made) as a long-time test.

ANALYTICAL PROBLEMS

(F. S. Tomkins)

Spectrographic Laboratory (F. Tomkins, J. K. Brody, S. Patterson, J. [illegible], R. Walsh)

A. Service Analysis:

The four Be metal samples mentioned last week were repeated. A blank of pure BeO run with the metal samples showed appreciable contamination probably from pyrex glassware. The samples were repeated using the available quartz ware and vicor crucibles. These were transferred to platinum crucibles for ignition. This time the black showed contamination of about .1 PPM. It will be necessary to obtain more quartz beakers, crucibles, etc., if we are to get reliable results on Be metal samples. Steps have been taken to do this.

The following analyses were also completed this last week:

1. Four samples BeO quantitatively for impurities.
2. Na - K alloy semi-quantitatively for Pt and other impurities.
3. Propylene glycol for aluminum.

B. Research:

The restandardisation of the copper spark is proceeding. Three elements have been standardized covering a range from 2100 A° to 6500 A° in five plates. Three additional elements have been standardized in the two lower regions.

The development of microphotometer methods for the analysis of BeO has again been resumed. One of the main advantages of the visual comparison method is in the very wide concentration range which can be

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utilized in a single exposure. The maximum concentration for the microphotometric method is 10 p.p.m. when no sector nor other means of reducing the intensity are used. In order to increase the useful range of the microphotometer method, the exposures will be taken through a step sector whose step ratio is 1:2. If four steps are used, this will give a range from .1 p.p.m. to 80 p.p.m. The higher standards are being made and the Baird step sector is being adapted to the Bausch and Lomb optical bench.

Two rotameters were received this week and will be used to control the He, O_2 gas mixture for our special D.C. arc gas source. A mount for the rotameters was designed and the carpenter shop is making it.

Wet Chemical Analysis (R. Bane, R. Hospelhorn, K. Hansen, R. Telford)

A. Services:

1. One manganese-copper alloy was analyzed in duplicate for copper and aluminum.
2. Two uranium oxide samples were analyzed for moisture and loss on ignition.
3. The final two beryllium standard samples were analyzed for nitrogen.
4. The micro-fluorine apparatus has been set up and work started on the determination of fluorine in beryllium.
5. Six barium hydroxide solutions containing barium carbonate were titrated for the base content and the carbonate content.
6. Three zirconium solutions were analyzed for the zirconium content.
7. It is believed that in the analysis of BeO rods for boron erratic results are due almost entirely to variations in the titration of the dissolved sodium carbonate melt. Experiments indicate that from 15% to 20% of the boron may be lost on ignition of B_4C.

B. Research:

The train for the analysis of BeO in Be was set up. A preliminary run was made and it was evident that extensive modifications were needed. Work is continuing on this analysis.

SPECIAL PROBLEMS
(O. C. Simpson)

Pure BeO Thermal Conductivity Rod (G. Steel, S. Harder, H. C. Andrews)

The pure BeO thermal conductivity rod from the 63 day bombardment was monitored 9/12/46, giving the following results:

Distance	No. Shield	Metal Screen	Metal & Plastic	Instrument
3"		3000 mr.	900 mr.	Zeus
6"	10,000 mr.			Zeus
3"	~4 mr.	Through shipping pot		Binder

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It was again monitored October 4, 1946 giving the following results:

Distance	No. Shield	Metal Shield	Metal & Plastic	Instrument
3"	40,000 mr.	2400 mr.	600 mr.	Zeus
6"	8,000		80	Zeus
9"	600		44	Zeus
12"	56		22	Zeus
15"	30		12	Zeus
1"	~5½-9½	Through shipping pot		Binder

It has proven difficult to measure the soft activity from the BeO rod at the surface. It is planned to measure the radiation on contact from a pure BeO pellet. From this data it may be possible to calculate the total activity from the pure BeO rod.

Quantitative Determination of Hydrogen in Na-K Alloy (F. L. Belletire, H. C. Andrews)

A sample of Na-K alloy has been received from site B for quantitative analysis of hydrogen. It is planned to react a known weight of Na-K with an excess of heavy water whose D to H ratio is known. Hydrogen (H and D) will be liberated from the heavy water by the Na-K. The hydrogen (H) which is present in the NaK will also be liberated. All of the hydrogen thus liberated will be catalytically united with O_2 and condensed. The water will then be distilled away from the alkalis analyzed by means of a mass spectrometer and the ratio of D and H determined. Given this new ratio, the amount of H_2 in Na-K can be calculated.

[illegible] trains have been designed for the Na-K alloy, and the glass shop is finishing the more difficult pieces. The reaction chamber is being designed now and it is planned to have the glass shop complete building it next week.

Temperature of BeO-UO_2 Prisms During Bombardment in HEW Pile (M. E. Kolenak, F. L. Belletire, H. C. Andrews)

Another prism has been prepared for temperature determination as planned (ANL-OCS-57, p. 4). The thermocouple junction is placed at the center of the prism through a small hole in the end. Preliminary runs at 20 watts indicate a prism temperature of about 280°C. with the aluminum can held at 40°C. Previous values obtained with the thermocouple placed at the end of the prism were considerably lower, 142°C. at a can temperature of 40°C. and 20 watts input. It is planned to run the experiment over 20, 30, 40, 50 watt ranges and at can temperatures ranging from 40° to over 100°C.

Attempts have been made to calculate the temperature on the basis of the change in the electrical resistivity of platinum by the formula

$$t = \frac{R_t - R_t}{R_{to}\alpha} + [illegible]$$

where t = Hot temperature to be determined.

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R_t = Resistance at temperature t.

R_{t_o} = Resistance at room temperature.

t_o = Room temperature.

α = Temperature coefficient of electrical resistivity.

R_t is determined by $\frac{E_t}{I_t}$ (the voltage and current measured at t). It is necessary to determine E and I quite accurately. We are looking for better instruments now in order to do this.

Remote Control Development (F. L. Belletire, M. E. Rebenak, R. W. Holmes, R. Miller, H. [illegible] Andrews)

Drawings for the improved cave periscope have been completed and prints are available in Room B-5 or at Ryerson for inspection. Work is being continued on hydraulic tongs, a remote control crane, high activity hot laboratory design, etc.

This type of design work entails considerable engineering, drafting, and shop work and it is felt that more help will be needed in some spots if the work is to proceed at a rate in line with the demand for remote control equipment.

Heat of Sublimation and Vapor Pressure of Graphite (R. J. Thorn)

Two attempts to extend the range of the measurements to a temperature of 50° to 100°C. higher than that employed in the first series of runs has resulted in either the quartz condenser or the beryllia support for the graphite cracking, or both. In view of these failures, it is impossible to extend the range without changing almost entirely the present experimental set-up. Since this will require more work than can probably be justified, no attempt will be made to redesign the apparatus at present. Instead, we will be satisfied with an upper limit of about 2110°C. and a range of 200°C.

GRAPHITE PROGRAM

(T. J. Neubert)

Isothermal Release of Stored Energy (Dunking Experiments) (C. M. Nelson, E. Avery)

A series of dunking experiments has been made with unirradiated graphite samples and an evaluation of these results is in progress. The apparatus consists of an all-glass vacuum-tight system in which there is a nickel cup containing Rose's metal alloy insulated from the glass envelope by a quartz peg and mica spacers. A mineral oil bath, which is heated by a coil of Nichrome wire, surrounds the vacuum jacket. The oil bath has asbestos packing insulation around the

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A graphite sample is pressed on an alundum tube which contains thermocouple leads. This tube is connected to a long metal plunger which has an iron cylinder at the top and and which can be moved by means of a solenoid from the outside. This part of the system is connected by a stopcock to a vacuum system. Helium can be admitted to the calorimeter part of the system while the calorimeter is heated to the desired temperature by the oil bath. The helium is pumped out of the system after the proper temperature is reached and a vacuum of 10[illegible] to 10^{-6} mm. Hg attained before dunking. Thermocouples are placed in the metal alloy bath and in the oil bath. A L & N type K-2 potentiometer and a galvanometer are used to measure the thermocouple temperatures. A photoelectric cell and galvanometer are arranged to control the temperature of the oil bath within ± 0.03°C.

A plot of the log of the temperature drop ΔT of the metal bath against time t was made and extrapolated linearly to zero time. The heat capacity C_p of the graphite sample was calculated from an equation taken from Bureau of Mines Bulletin 371 (1934):

$$C_p = 2.673 + 2.617 \times 10^{-3}T - 1.169 \times 10^{5}T^{-2}$$

and the quantity of heat q(T) from $\int C_p dT$. As a first approximation, the rate of heat leak in the calorimeter is given by $\frac{dq}{dt} = k\Delta T$. A more exact mathematical analysis of the data is being made using the equation $\frac{dq}{dt} = K(T_0^4 - T^4)$ which describes the conditions more fully. The results will be given next time.

From above $\frac{dq}{dt} = k\Delta T$, then $-C\frac{d\Delta T}{dt} = k\Delta T$ and $-\frac{d\Delta T}{\Delta T} = \frac{k}{Cdt}$ so that $\ln\frac{\Delta T_0}{\Delta T} = \frac{k}{Ct}$ and $\frac{k}{C} = \frac{\ln\Delta T_0/\Delta T}{t}$ where ΔT_0 is ΔT at $t = 0$ and C is the heat capacity of the calorimeter system. Table I gives the results of the measurements.

TABLE I

Expt.	T, Orig. (°K)	$T_{extrapolated}$ (°K)	k/C (min^{-1})	k (cal min^{-1} deg^{-1})	C (cal deg^{-1})
10	413.3	409.6	0.02320	0.2357	10.16
11	383.4	380.8	0.01954	0.2034	10.41
12	442.6	437.9	0.0330	0.3402	10.31
13	502.7	495.5	0.04881	0.4968	10.18
14	473.4	467.5	0.03964	0.4186	10.56
15	533.2	524.7	0.05808	0.5994	10.32
16	399.4	396.3	0.02076	0.2221	10.70

Since the values of C vary so much, the C_p values as calculated from the Bureau of Mines equation is not sufficiently accurate and C will be determined with a metal (e.g., Cu) of known specific heat. Also several more dunkings of cold samples will be made before neutron irradiated samples are used and the amount of stored energy contained in them determined.

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Measurement of Stored Energy by Sykes Method (B. W. [illegible], R. [illegible])

In the Sykes experiment, an irradiated and an unirradiated sample of graphite are heated together in a furnace, and the temperature differences between the samples and their surroundings measured. These data are used to calculate the amount of stored energy from neutron bombardment released by annealing and to give information about the manner in which it is released.

The ratio of the temperature differences may be shown to be equal to the ratio of the apparent specific heats of the samples. This ratio is plotted against the temperature of the irradiated sample. In the first heat the curve tends upward from unity as the release of stored energy raises the temperature of the "hot" graphite, and should eventually return to some value near unity after the stored energy of disturbances has been healed (see figure).

The maximum temperature of the furnace used is about 750°C. At this temperature the C_H/C_C curve does not usually close to unity (and actually in some cases tends to drop further toward zero). The furnace is allowed to cool to room temperature over night and a second run made the following day. These data usually reveal a near linear change of C_H/C_C with C_H approaching unity from above. The hypothesis is that the difference of the ratio at the end of a first heat and at the beginning of a second heat represents stored energy released during cooling over night, or more likely during the first half-hour after power to the heating unit is turned off at about 700°C.

To investigate this phenomenon, a heat was made in the usual manner carrying the furnace to a temperature of 560°C, where it was certain from the ratio of the temperature differences that stored energy was being released, and the power was cut, readings being taken of the cooling of the system to room temperature.

Since the temperature differences are very small and the variations in them at the time of reversal from heating to cooling relatively large, their ratio becomes quite irregular until cooling equilibrium is established (see figure 1st heat).

A second heat was made the following day (see figure 2nd heat) and proved to be quite uniform. A comparison is made in the accompanying figure between this second run on the partially annealed sample and the first heat on a previous run, both runs employing an irradiated sample from [illegible] and approximately the same operating conditions.

The manner in which the partially annealed curve joins that of the unannealed sample seems to indicate that differences between first and subsequent heats may be ascribed (at least to a large extent) to the incomplete release of stored energy.

A second run of this type on a "T" bar sample has been completed and is now being calculated. It is also contemplated in the near future to freeze various portions of the incompletely healed stored energy in graphite samples by rapid immersion in a cool bath and studies will be conducted on the healing of the remaining energy.

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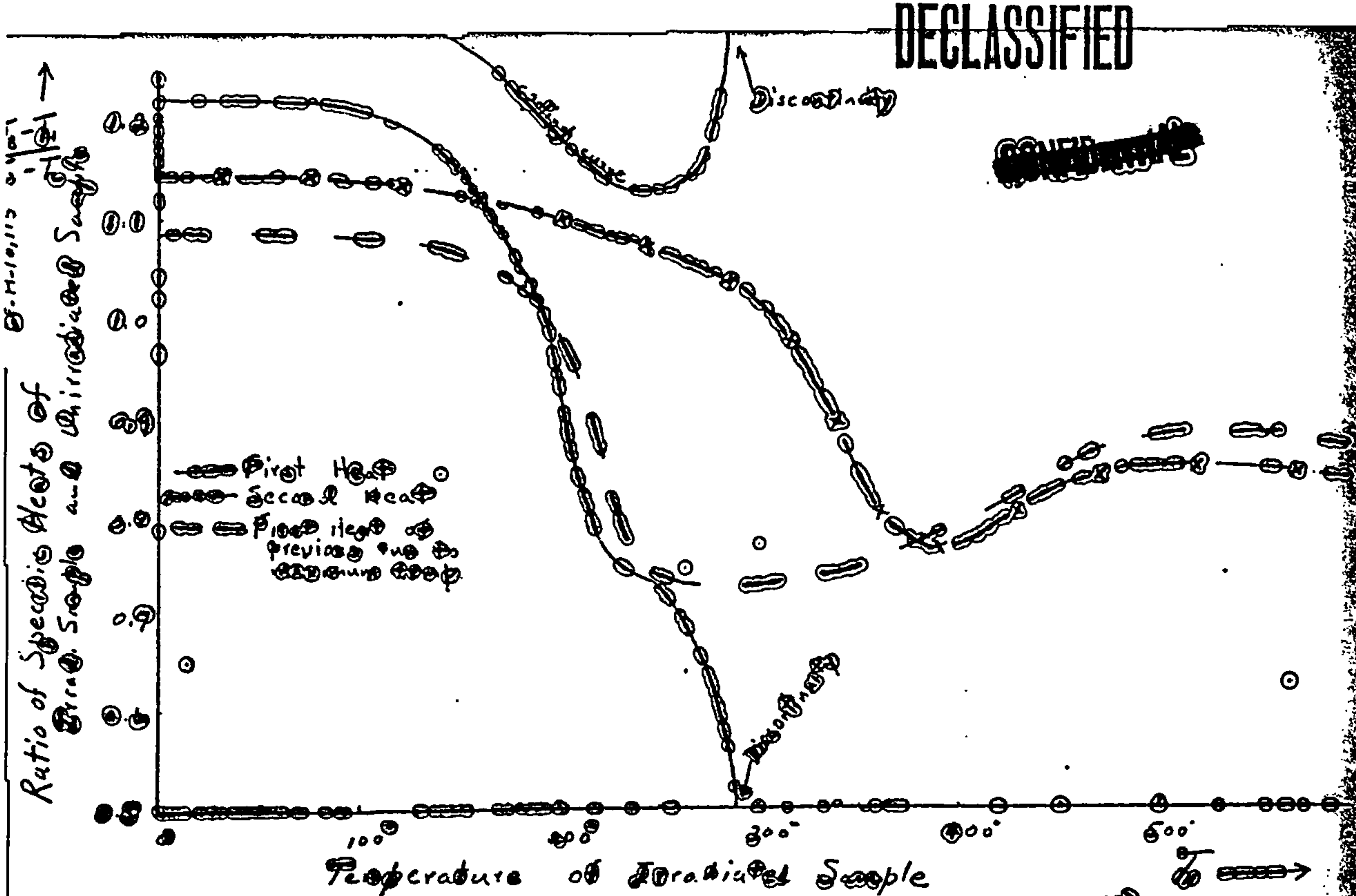

Syres Apparatus

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C^{14} Distribution in Graphite (W. [illegible])

The experiment previously described, in which powdered Hanford graphite was burned in small successive increments by admitting controlled amounts of air or oxygen to the graphite in a quartz tube at 770°C, has been repeated at ca. 500°C. The graphite was lightly ground with silica gel in a mortar. It was expected that silica gel would assist penetration of oxygen to all graphite particles and help prevent hot spots when pure oxygen was admitted. The temperature was so adjusted as to burn less than 1 mg. of graphite in ten minutes. Thirty-three fractions were collected from a 36 mg. sample of graphite, totally burned. Although performed under somewhat different conditions, results of these experiments are in complete qualitative agreement. The results, obtained by the method just described, are adequately represented by the following equations (where E = differential enrichment ratio, B = total % burn):

0 – 60% burn, [illegible] 37
60 – 80% burn, [illegible] 12
80 – 100% burn, [illegible] 3.5

The specific activities of the last two samples ([illegible] and 92 – 100%) were only 0.08 and 0.05 times the sample average. This fact indicates that approximately 5% of dislocated atoms heal to holes in the lattice if C^{14} atoms are representative of all dislocated atoms.

In an earlier experiment a sample of the same graphite was thoroughly evacuated, then fractionally burned at 565° with the following results:

E	6.[illegible]	5.[illegible]	[illegible]	[illegible]	[illegible]
B	0.098	[illegible]	[illegible]	[illegible]	[illegible]

If, as indicated above, dislocated atoms heal preponderantly to edges, then each crystallite may be supposed to have a sheath of healed dislocated atoms, and the initial value of E should correspond roughly to (total number atoms)/(number dislocated atoms) = 5.5.

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