DFC

MLM-MU-77-62-0004

Decontamination of HEPA Filters

Quarterly Report

October, November, December, 1976

MOUND LABORATORY
Miamisburg, Ohio
operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company
for the
UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION
U. S. Government Contract No. EY-76-C-04-0053

Monsanto

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DECONTAMINATION OF HEPA FILTERS
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INTRODUCTION

Mound Laboratory, in its many years of plutonium processing experience, has recovered over 150 kilograms of plutonium-238. Much of this material has been recovered from HEPA filters and other secondary wastes such as sludge, slag, etc. The basic recovery nitric acid leach process at Mound for the decontamination of HEPA filters will be improved and modified in this project for processing filters from the Nuclear Fuel Cycle. Efforts this past quarter were directed toward determining the dissolution parameters of plutonium in HNO_3 which included a study of the optimum acid concentration and optimum dissolution temperature.

EXPERIMENTAL WORK

Contaminated HEPA filter media was prepared by mixing PuO_2[1] powder with shredded filter media. The resulting mixture contained approximately 0.2 curies (11mg) of Pu-238 per cubic centimeter of prepared media. Small samples of this prepared mixture ($\sim$ 3.0 grams) were put into beakers containing 250 ml of HNO_3 of the desired concentration. The beakers and contents were than heated to the desired temperatures. Samples were taken every 4 to 8 hours and the Pu-238 concentration of the acid solution determined. Stirring of beaker contents was done every hour, with one exception in which constant magnetic stirring was used. The solution volume and concentration were adjusted when necessary by adding nitric acid to the beaker to replace evaporated acid.

Table I shows results from dissolution tests using nitric acid of various concentrations (6N, 8N, 10N, 12N, and 15.7N). It

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(1) This powder has an average particle size of approximately 20 microns. It is a mixture of plutonium oxides (80% Pu-238, 16% Pu-239, 2.5% Pu-240, 0.8% Pu-241, 0.2% Pu-242) plus very small amounts of other actinides.

TABLE I

Acid Test Number	Acid Conc.	Acid Temperature	Time (Hr.)	PuO_2 * Dissolved (Milligrams)	PuO_2 * Dissolved Wt. %
1-1	6N	80°C	4	1.40	0.88
1-2	6N	80°C	8	1.89	1.18
1-3	6N	80°C	13½	2.73	1.71
2-1	6N	90°C	4	1.62	1.07
2-2	6N	90°C	8	2.22	1.47
2-3	6N	90°C	13½	3.28	2.17
3-1	6N	Boiling	2½	2.00	1.31
3-2	6N	Boiling	5½	2.82	1.84
3-4	6N	Boiling	10½	3.80	2.48
4-1	8N	85°C	4	2.40	1.62
4-2	8N	85°C	8	2.75	1.86
4-3	8N	85°C	13	5.50	3.72
5-1	8N	95°C	4	2.75	1.86
5-3	8N	95°C	8	3.50	2.36
5-4	8N	95°C	13	6.12	4.14
6-1	8N	Boiling	6	5.40	3.75
6-2	8N	Boiling	12½	5.80	4.03
6-3	8N	Boiling	18½	13.00	9.03
7-1	10N	85°C	6½	2.75	1.88
7-2	10N	85°C	11½	4.00	2.73
7-3	10N	85°C	16¼	4.14	2.82
8-1	10N	95°C	6½	4.50	3.20
8-2	10N	95°C	11½	5.85	4.15
8-3	10N	95°C	16¼	6.90	4.90
9-1	10N	Boiling	6½	3.75	2.39
9-2	10N	Boiling	11½	4.73	3.01
9-3	10N	Boiling	16¼	6.35	4.04
10-1	12N	85°C	6½	2.80	1.68
10-2	12N	85°C	11½	4.00	2.40
10-3	12N	85°C	16¼	5.13	3.08
11-1	12N	95°C	6½	4.00	2.39
11-2	12N	95°C	11½	5.40	3.22
11-3	12N	95°C	16¼	5.63	3.36
13-1	12N	Boiling	4½	5.64	3.78
13-2	12N	Boiling	11¼	8.80	5.90
13-3	12N	Boiling	18	8.48	5.68
14-1#	10N	95°C	4½	3.70	2.68
14-2#	10N	95°C	9¼	4.90	3.55
14-3#	10N	95°C	16¼	5.95	4.31
15-1	15.7N	85°C	5	4.41	3.35
15-2	15.7N	85°C	13	4.94	3.76
15-3	15.7N	85°C	16½	4.83	3.68
16-1	15.7N	95°C	5½	4.41	3.14
16-2	15.7N	95°C	13	3.83	2.72
16-3	15.7N	95°C	16½	5.52	3.93
17-1	15.7N	Boiling	6¼	6.67	4.70
17-2	15.7N	Boiling	13	6.60	4.65
17-3	15.7N	Boiling	16 3/4	6.67	4.70

*Expressed as Pu metal
#Beaker contents stirred continuously by magnetic stirring bar.

should be noted that the optimum acid concentration was 8N and optimum temperature at this concentration was $\sim 120^{o}C$ (boiling). The maximum percent PuO_2 dissolved was 9.03% after 18.5 hours of heating (see acid test No. 6-3).

The data in Figure 1 illustrates how the dissolution rates vary with temperature. This plot is for 6N HNO_3 and is, in general, similar to the plot for 8N, 10N, 12N, and 15.7N, with one exception. The exception is that for 10N HNO_3 the maximum dissolution rate was not at the boiling temperature, but at $95^{o}C$. Therefore, the tests with 10N HNO_3 will be re-run to investigate this phenomenon.

The data in Figure 2 illustrates the effect of stirring on the dissolution rates. As can be seen, the dissolution rates are essentially identical, the small difference could be due to experimental error. One explanation for the identical rates is that both solutions are actually undergoing constant mixing, the one due to convection currents caused by constant external heating.

The effect on dissolution of asbestos and fiberglass in the filter media is not presently known. Isotherms will be run to determine the effect, if any, the media has on the solubility and dissolution rates of the PuO_2.

CONCLUSIONS

Dissolution tests were conducted with 6N, 8N, 10N, 12N and 15.7N HNO_3 on filter media contaminated with PuO_2. The optimum acid concentration and temperature were 8N and boiling ($\sim 120^{o}C$) respectively. The weight percent PuO_2 dissolved after 18.5 hours was, however, only 9.0%. This work substantiates data collected in Recovery Operations at Mound Laboratory, that nitric acid alone is not an effective leach reagent. However, this data provides basic information on the effects of the filter media on the equilibrium reaction. It was found that stirring of the leach solutions had no effect on the dissolution rate. During the next quarter, other leach reagents will be investigated. These will include HNO_3 with dilute concentration of HF; HNO_3 in combination with small percentages of H_2SO_4; HNO_3 with ceric salts[1]; and combinations of HNO_3, HF and H_2SO_4[2]. Also, electrolytic oxidation[3] of dissolved Pu in HNO_3-HF solutions might be tried, as well as ultrasonic agitation of leach reagents.

Figure I. A Comparison of the Dissolution Rates of PuO_2 in 6N HNO_3 at Selected Temperatures

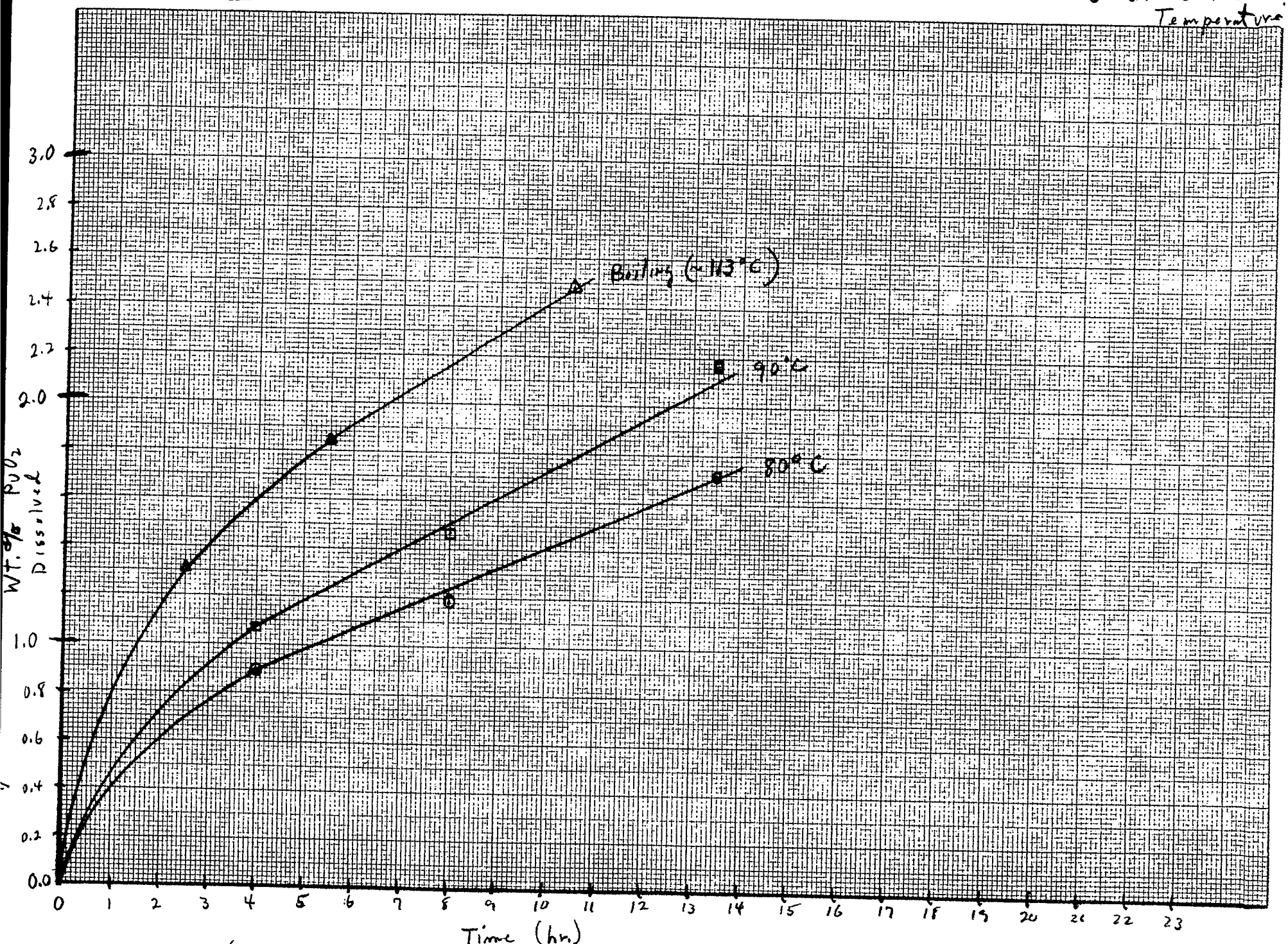
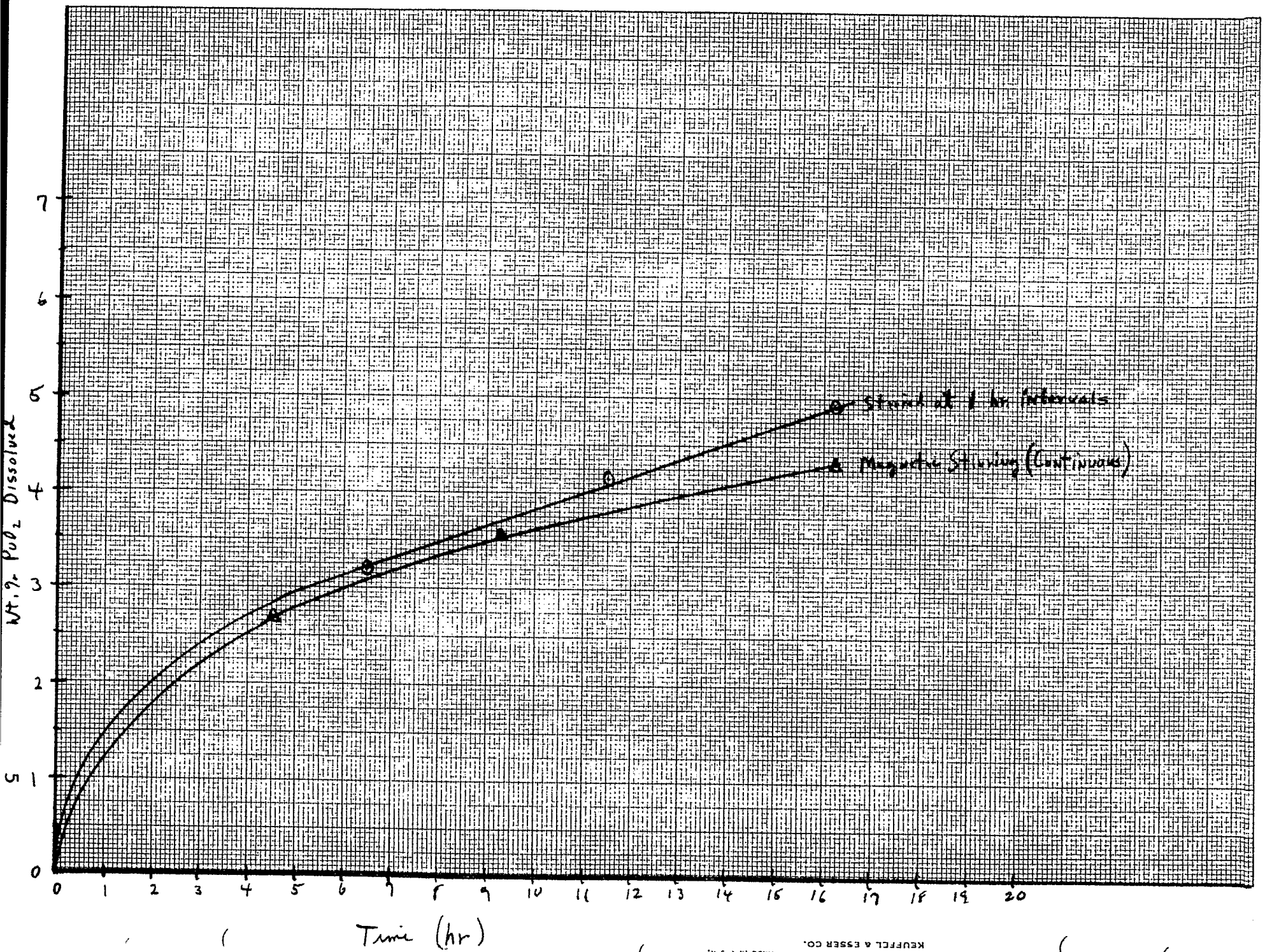


Figure II. Comparison of Stirring Methods on Dissolution Rate of PuO_2 in 10N HNO_3 at 95°C.



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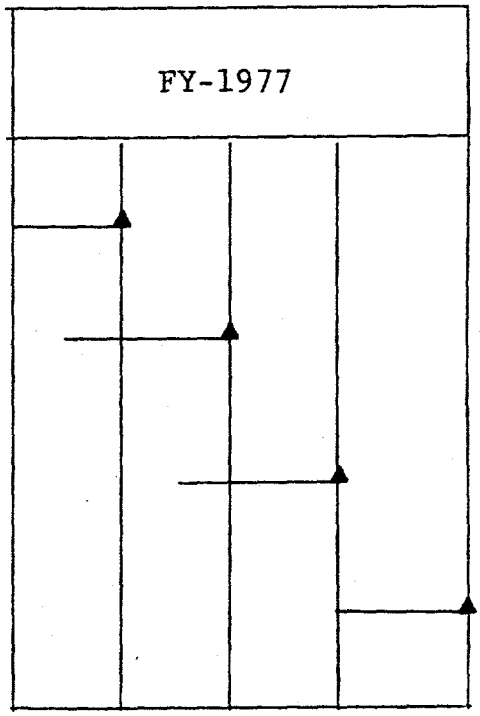

Phase I

A. Determine parameters of HNO_3 leach test with Pu.

B. Determine parameters of alternate reagent leach tests with Pu.

C. Determine leach parameters for other actinides.

D. Determine and compare waste streams from selected processes.

Milestone A has been completed except for repeating tests with 10N HNO_3 and running isotherms to determine the effect the presence of asbestos and fiberglass has on dissolution rate. In Part B, alternate leach reagents (listed previously) are to be investigated. Part C will involve determining the leach parameters for other actinides as U-234, Am-241 and Cm-244. Material balance will be performed and decontaminated factors determined for these decontamination procedures. Flowsheets will also be prepared. At this time, indications are that the decontamination process will be a two or three stage operation, the initial stage (or step) removing 99% + of the actinides and the final step(s) the remainder. In Part D, waste streams from the most feasible processes will be studied and comparisons made among them. Additional material balance will be done for the waste streams.

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