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QUARTERLY TECHNICAL PROGRESS REPORT ON ARPA PROMPT FISSION YIELD PROJECT WORK FOR THE PERIOD 1 NOVEMBER, 1970 - 31 JANUARY, 1971

JANUARY 31, 1971

PREPARED FOR

THE UNITED STATES ATOMIC ENERGY COMMISSION

UNDER CONTRACT AT(45-1)-1830

BATTELLE NORTHWEST
PACIFIC NORTHWEST LABORATORIES DIVISION
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January 31, 1971

(a) This paper is based on work performed under United States Atomic Energy Commission Contract AT(45-1)-1830.

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Pneumatic Sample Transfer Facility

A mockup has been fabricated and tested in the laboratory of a switch which will sense the arrival and departure of a rabbit capsule at the irradiation location. A drawing is shown in Figure 1. The switch makes electrical contact as follows: The incoming rabbit strikes the movable piston, which causes the piston to contact a spring. The spring is attached to an insulated electrical lead. The electrical lead is thus grounded when the rabbit arrives. When the rabbit is sent out, gas pressure forces the piston away from the spring and thus breaks contact with the insulated wire.

An improved version of the mockup switch is being fabricated out of stainless steel for future installation.

Fast Radiochemical Separation of Bromine and Krypton

The apparatus for bromine collection has been considerably modified compared with the illustration in Figure 1 of the July-October Quarterly Report. Provision has been made for collection of other gases liberated from the sample during evolution of bromine. Bromine is removed from these gases by means of a quartz tube cooled with dry

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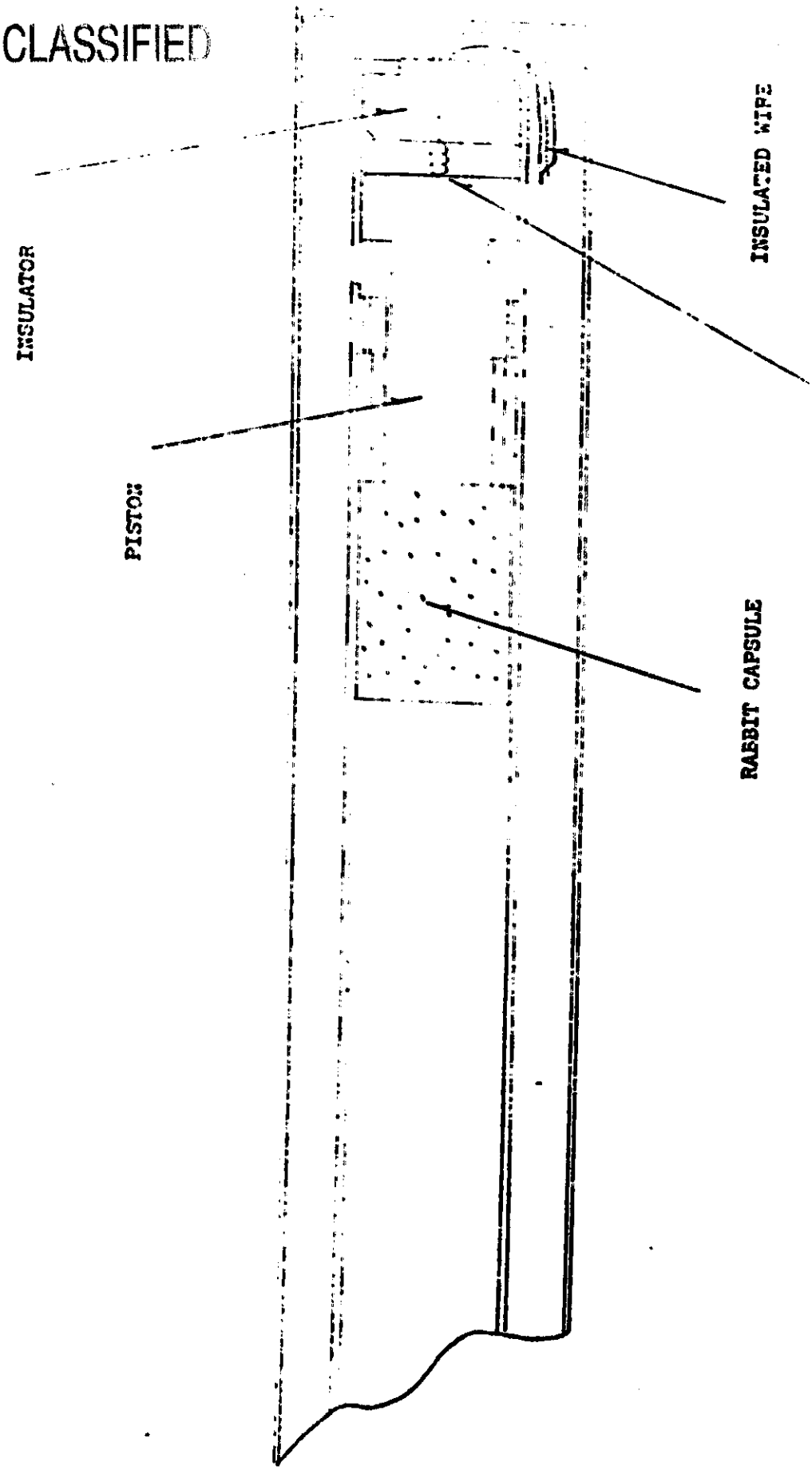

Figure 1. SCHEMATIC OF REACTOR IN-TIME SWITCH

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ice and a carbon tetrachloride trap. Since this gas sample is collected before appreciable decay of ^{85}Br and ^{87}Br, the ^{85m}Kr and ^{87}Kr activity is mainly due to independent production of these nuclides by uranium fission. With a short irradiation time and 2-second bromine collection interval the ^{88}Kr present in this sample would also be mainly due to independent production. Thus it is hoped to measure the independent fission yields of these nuclides as well as those of ^{85}Br, ^{87}Br and ^{88}Br with the same apparatus.

Another modification we have made allows us to sweep the Kr isotopes resulting from bromine decay into a separate trap, instead of having to count the bromine sample directly. This reduces interference due to large amounts of ^{82}Br from the (n,γ) reaction on uranyl bromate. Both rare gas samples are collected by sweeping the respective samples with helium through charcoal traps cooled with liquid nitrogen.

In order to perform the additional steps in the separation process, the control system which operates the solenoid valves was considerably modified and further automated. The entire sequence after the rabbit is sent into the reactor is now automated by means of solid state timing circuits and a 35mm tape programmer.

Considerable effort has been spent trying to produce a sealed uranyl bromate container so that no loss of rare gases will occur before chemical separation. In the previous report it was mentioned that a satisfactory sample container was prepared by wrapping the uranyl bromate powder with aluminum foil and shaping this into a ball.

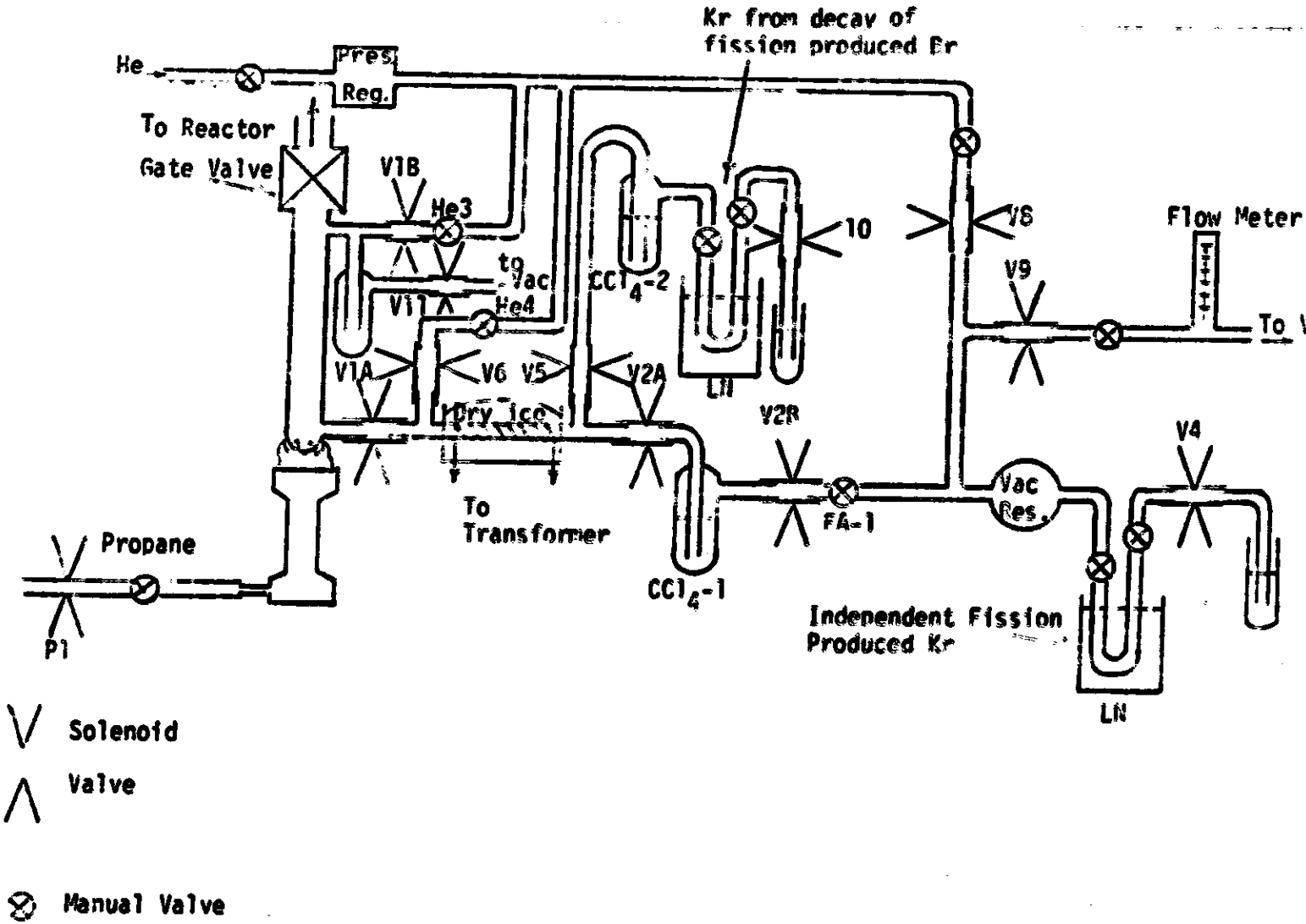

Figure 2. SCHEMATIC OF BROMINE/KRYPTON SEPARATION SYSTEM

This technique has worked well except that since the sample is not completely gas-tight, some rare gases could escape into the rabbit capsule during the irradiation. Such a loss would not be of consequence for measurement of bromine independent fission yields, but it could cause difficulties in krypton indenendent yield determinations since no krypton carrier is present in the sample during irradiation. Experiments are planned to measure how much krypton actually leaks out of aluminum foil-wrapped samples. A way of eliminating the problem is to place the sample in a gas-tight container. We have experimented with several possibilities of this sort. Any material containing organic compounds (such as plastics, epoxy, glue) decompose to carbon and other compounds which absorb the desired bromine when the sample is heated. Glass ampoules cannot be easily and quickly broken and heated to sufficient temperature. Small metal cans sealed with solder heat rapidly and the solder melts before the pressure inside the container ruptures it explosively. However, liquid solders apparently have an unusually high affinity for bromine, and chemical yields with this type of container are unacceptably low.

A screw-cap, thin-bottomed aluminum capsule appears to have possibilities. It is difficult to machine capsules with very thin (2 mil) bottoms, but if this is done the container bottom will melt in the heating chamber without violently rupturing, and bromine recoveries are satisfactory. The capsules are heavier than foil-wrapped samples which may present problems when very rapid heating is required because of the

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heat capacity of the capsule. Also, the capsule may break the heated quartz tube when it falls in. Further tests are planned.

Selenium Chemistry

In order to measure the independent fission yields of bromine isotopes it is necessary that the rapid radiochemical procedure separate bromine from the fission-produced precursors selenium and arsenic. An experiment was devised to directly measure fission-product selenium in both the separated bromine sample and in the U_3O_8 residue after the uranyl bromate had been decomposed. After an irradiation of uranyl bromate and chemical separation of bromine, the U_3O_8 residue was dissolved in concentrated HCl which contained selenium carrier. The solution was brought from the reactor to the radiochemistry laboratory and selenium was separated and counted on a Ge(Li) diode. The bromine fraction from the dry ice trap was treated in similar manner. Sufficient ^{83}Se activity (23 minute half-life) was present in the two fractions to determine that a significant amount of selenium activity is carried along with bromine and deposits in the dry ice trap. The selenium presumably comes over as a relatively volatile bromide compound. Thus we will have to find some means of rapidly separating this selenium compound from bromine if we are to be successful in measuring the independent fission yields of bromine isotopes. It is suspected that arsenic may behave similarly since it also forms relatively volatile bromine compounds, although we have not confirmed this by direct measurement.

Data Handling

A great deal of labor and time has been saved through extensive use of the PDP-9 computer for analysis of the gamma spectral data. A program called GASPAN (Gamma Spectral Analysis), which was originally written by B. Barnes in England for use on an IBM 360 computer, has been adapted for the PDP-9 by D. Donati of Lowell Technical Institute, Lowell, Massachusetts, and further modified by J. E. Fager of this laboratory. This program accepts paper tape readout from a pulse-height analyzer, stores the gamma spectra on magnetic tape (DEC tape), and searches each spectrum for photopeaks. The area of each peak is computed (after Compton correction), and the energy of the center of each peak is estimated. The photopeak counting rate may be corrected for decay back to the time of irradiation, and for detector counting efficiency. Once a photopeak has been identified as resulting from the decay of a known radionuclide, the program will label the peak with the corresponding radionuclide. After the detector has been calibrated the program will also output an absolute desintegration rate estimate for each nuclide if desired. The program also has the valuable feature of being able to resolve doublet photopeaks. Figure 3 illustrates typical computer output from GASPAN.

Preliminary Results

Radionuclides observed in the first test of the modified separation apparatus include $^{85m,87,88}Kr$, $^{135,138}Xe$, ^{138}Cs, ^{88}Rb and

[redacted]

[illegible]		END TIME	32[illegible].	10-4 DAYS
[illegible]	1	LIVE TIME	104.2	10-4 DAYS
[illegible]	24	LIVE TIME	15.0	MIN
[illegible]	1	CAL	[illegible]	DEL 3
[illegible]	[illegible]	TIME ZERO	3200	0. 10-4 DAYS

[illegible] 1.000 SAMPLE SIZE 1.00 EFFICIENCY CURVE 1

NUCLIDE	ENERGY KEV	STD LCC	MN	AREA CPIC-4DY	ERROR	ABND PCT	ESTIMATE DPIC-4 DY	ERROR	NUCLIDE
KR-88	196	C	88	5.80E+02	5.1E+01	38	7.9CE+04	6.9E+03	KR-88
	2392			4.94E+01	2.1E+00	34	1.00E+05	4.2E+03	
	[illegible]			4.63E+01	2.3E+00	15	2.20E+05	1.1E+04	
	1531			2.83E+01	2.7E+00	11	1.11E+05	1.0E+04	
	1240			1.01E+02	9.2E+00	1	6.97E+06	3.5E+05	
	836			5.90E+01	5.3E+00	13	1.00E+05	9.7E+03	
	302			6.04E+01	5.8E+00	3	1.80E+05	1.7E+04	
	100			8.59E+03	1.4E+02	7	1.70E+06	9.1E+04	
KR-87	402	C	87	3.77E+00	2.1E+01	49	6.34E+04	3.5E+03	KR-87
	1353			4.07E+01	4.3E+00	1	2.02E+06	1.9E+05	
XE133	257	C	13	9.77E+04	4.6E+03	100	5.95E+06	3.0E+05	XE133
	421			3.64E+03	9.7E+02	63	6.14E+05	1.6E+05	

*** OTHER NUCLIDES POSSIBLE AT FOLLOWING ENERGIES ***

ENERGY K V	AREA CPIC-4DY	ERROR	CANTAS PER 10-4DY,	ERROR	EFFICIENCY PER CENT
2709	2.10E+00	2.37E-01	1.86E+03	2.09E+02	0.1157
2042	2.28E+01	1.57E+00	2.44E+04	1.33E+03	0.1103
2871	2.72E+00	4.11E-01	7.85E+03	3.30E+02	0.1212
[illegible]	2.00E+00	2.05E-01	7.63E+03	7.35E+02	0.1219
2082	6.40E+01	3.69E+00	4.64E+04	2.65E+03	0.1393
2139	5.47E+00	6.00E-01	3.63E+03	4.59E+02	0.1451
2217	5.09E+00	9.71E-01	3.71E+03	6.33E+02	0.1533
2277	1.55E+01	1.92E+00	1.00E+04	1.25E+03	0.1541
1937	2.79E+01	1.83E+00	1.64E+04	1.08E+03	0.1698
1779	9.76E+00	1.99E+00	4.93E+03	6.22E+02	0.1752
1702	7.57E+00	9.99E-01	4.15E+03	5.47E+02	0.1827
1625	4.76E+00	7.37E-01	2.69E+03	3.98E+02	0.1854
1611	1.51E+01	1.03E+00	8.35E+03	6.60E+02	0.1935
1530	4.11E+01	1.36E+00	2.03E+04	6.63E+02	0.2040
1477	5.12E+00	6.89E-01	2.61E+03	1.38E+04	0.2012
1371	1.05E+01	1.91E+00	5.40E+03	6.46E+02	0.2338
1347	6.13E+00	1.31E+00	3.16E+03	5.40E+02	0.2357
1156	3.01E+01	3.[illegible]	1.69E+04	1.89E+03	0.2727
1134	1.31E+01	1.09E+00	4.30E+03	6.96E+02	0.2978
1130	2.01E+00	4.45E-01	3.77E+04	1.37E+04	0.3066

Figure 3. Sample of GASPAN Output

[redacted]

tentatively ^{139}Ba, ^{89}Rb and ^{92}Sr. Nuclides with half-lives less than about 30 minutes cannot easily be identified due to the delay before the samples can be brought from KE reactor to the detectors in the radiochemistry building. The first test of the apparatus gave the activities listed in Table 1, in units of counts/10^{-4} day(peak height).

TABLE 1

Relative Activities (c/10^{-4} day) of Kr Isotopes

NUCLIDE	Kr (INDEP.)	Kr (Br DECAY)	PHOTOPEAK ENERGY (KeV)
^{85m}Kr	20	37	150
^{87}Kr	.56	.88	2560
^{88}Kr	190	38	195

The ratios of amounts of krypton isotopes in the independent samples to the amounts in the Br decay samples were: ^{85m}Kr 0.5, ^{87}Kr 1.1, ^{88}Kr 5.1. Since a 10 second irradiation was made with 6-second separation time, about half of the ^{88}Br would have decayed into ^{88}Kr. Future runs will be made for shorter irradiation and separation intervals.

Iodine Separation

No work on the iodine system has been performed since the last report. The control system for the iodine apparatus has been considerably modified for use with the bromine system. Problems with electrical contact in the reaction chamber of the iodine apparatus have not been solved yet, but no work has been done yet to solve this problem.

Computer Monitor/Control System

Parts have been received and advanced design is nearly completed on the computer monitor/controls system which will allow an experiment at the reactor to be monitored and controlled by means of the PDP-9 computer in the radiochemistry building. An order for acoustical couplers for the transfer of information to/from the computer has been sent out for bids. It is hoped that this system will record all pertinent data during an irradiation and separation and automatically modify the course of the experiment as necessary should this be desired.