United States
Naval Postgraduate School

THESIS

SEARCH FOR SHORT LIVED ISOTOPES IN

VOLATILE REACTOR FUEL PRODUCTS

by

Ward Duane Fagan

JUNE 1969

41

Search for Short Lived Isotopes in Volatile Reactor Fuel Products

by

Ward Duane Fagan
Major, Chemical Corps, United States Army
B. A., University of Colorado, 1959

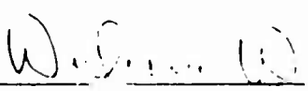
Submitted in partial fulfillment of the
requirements for the degree of

MASTER OF SCIENCE IN CHEMISTRY

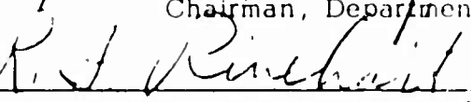
from the

NAVAL POSTGRADUATE SCHOOL
June 1969

Author 

Approved by: 
Thesis Advisor


Chairman, Department of Chemistry


Academic Dean

ABSTRACT

The volatile radioactive nuclides produced by thermal neutron fissioning of uranium oxide-polyethylene fuel of the AGN-201 Reactor were collected in an evacuated aluminum cylinder. The following constituent isotopes, identified by gamma ray spectrometry, present in 20 minute old gas are; Te^{131} Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{137}, Xe^{138}, Xe^{139}, Se^{81}, Se^{83}, Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, and Kr^{89}. Radioactive daughter products identified are Cs^{137}, Cs^{138}, Cs^{139}, Rb^{88}, Rb^{89}, and I^{131}. Other isotopes of iodine, tellurium, and bromine may be present. Spectra were taken with a NaI (Th) scintillation crystal.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF ILLUSTRATIONS

LIST OF DEFINITIONS OF ABBREVIATIONS AND SYMBOLS

1. AGN-201 - The model 201 nuclear reactor produced by Aerojet General Nucleonics Corporation, San Ramon, California

2. MEV - million electron volts

3. NPS - The Naval Postgraduate School, Monterey, California

4. Elemental symbols:

   Al - aluminum
   Au - gold
   Br - bromine
   Co - cobolt
   Cs - cesium
   I - iodine
   Kr - krypton
   Mn - manganese
   Rb - rubidium
   Se - selenium
   Te - tellurium
   U - uranium
   Xe - xenon

ACKNOWLEDGEMENTS

I wish to express my appreciation to Dr. William W. Hawes for his guidance and timely advice. His counsel and enthusiasum have contributed materially toward this endeavor.

I also wish to extend my sincere gratitude to Mr. Harold McFarland, reactor operator, for his outstanding assistance. Without his technical competence and devotion to duty this project could never have been successfully completed.

I would be remiss if I did not give credit to my wife Rogene for her encouragement and support. Her assistance in extracting data, preparing graphs, and typing of drafts has been a significant contribution to this work.

I. INTRODUCTION

In the past many attempts have been made to analyze both qualitatively and quantitatively the gaseous effluents given off from the core of the AGN-201 Nuclear Reactor at the Naval Postgraduate School. Bredderman [1] conducted a detailed analysis of the gamma ray spectrum of the gas given off by the core of the reactor and sampled two hours after peak of power operation. The principle radioactive isotopes present based on gamma ray photopeak energies and half lives were found to be Kr^{85m} 10.2%, Kr^{87} 9.0%, Kr^{88} 47.4%, Xe^{133} 18.6%, Xe^{135} 4.4%, and an unidentified activity 10.4%. He observed the total activity to be about 363 microcuries per milliliter. Laskaris [2] analyzed the reactor core gas by means of a gas chromatograph and found the gas to be comprised almost 50% by volume of hydrogen, with the remaining constituents being methane, ethane, ethylene, and other light saturated, unsaturated, and cyclic hydrocarbons. He stated the oxygen, carbon dioxide, and carbon monoxide detected in these gases was directly attributed to the impurities of the nitrogen gas used to pressurize the reactor. At the same time he made an activity analysis that demonstrated the existence of xenon and krypton radio-nuclides in the effluent gases. He further determined the percentage by volume of krypton and of xenon to be less than 0.5% each. In a later work by Cole [3] the gases produced by radiation of the polyethylene moderated uranium oxide fuel were analyzed for both radioactive and nonradioactive

species. He showed the hydrogen content to be about 60% by volume, the ethane-ethylene to be 17%, methane 19% with the remaining composition being that of the higher alkanes and cyclic hydrocarbons. His gas chromatograms showed xenon to be faintly detectable. Trace quantities of krypton and xenon were successfully separated without isotopic dilution using gas-solid chromatography. The radioactive species identified by Cole were Kr^{85m}, Kr^{87}, Kr^{88}, Xe^{133m}, Xe^{133}, Xe^{135}, Xe^{138}, I^{131}, Cs^{138}. Cole believed that the iodine present would be in the form of hydrogen iodide or compounds such as methyl iodide and ethyl iodide. However, it should be pointed out that no hydrogen iodide or methyl iodide was observable in his chromatograms. Cole further indicated the possible existence of Rb^{88}, Rb^{89}, Xe^{131}, Xe^{135m}, I^{133} and I^{135} based solely on photopeak identification with no supporting half life data. At the same time Lindsay [4] conducted a very thorough half life study of various photopeaks found in the gamma ray spectrum of the volatile reactor fuel products. He observed some 34 to 40 photopeaks of which several were not strong enough to make any detailed half life study. He did identify 11 constituent nuclides including two radioactive daughters by this method. They were I^{131}, Xe^{131}, Xe^{133m}, Xe^{133}, Xe^{135}, Xe^{138}, Kr^{85m}, Kr^{87}, Kr^{88}, Cs^{138}, and Rb^{88}. He further reported the possible presence of I^{133}, I^{134}, I^{135}, I^{136}, Xe^{137}, Kr^{89}, and Rb^{89} from a photopeak energy evaluation. Lindsay proposed the presence of Br^{84m}, and Br^{84} isotopes. These

bromine isotopes have half lives of 6 and 32 minutes respectively. This suggestion plus that of the possible presence of I^{136}, which has a half life of 86 seconds, required an extremely rapid method of extraction of the gas be developed before a positive identification could be made.

The thermal neutron fissioning of uranium 235, has been thoroughly investigated by numerous people and a fine compilation of fission product yields has been prepared by Katcoff [5]. The most probable masses of the fission fragments from a thermal neutron fission reaction are 93 and 134 atomic mass units with 7 and 8% fission yields respectively. Masses which have fission yields greater than 3% are 88 through 103 and 131 through 146. Most of the fission fragments are metallic atoms although nuclides of xenon, iodine, krypton, and bromine are chemically stable in gaseous form. Therefore, it is safe to assume that any gaseous products from a fission reaction should likely contain isotopes of these elements. Furthermore, the identification of one isotope of an element as a constituent in the reactor gas implies the other fission product isotopes of that element are also present. This leads to a mixture of isotopes of one or more elements whose spectrum may be difficult to resolve in areas where different gamma rays have similar energies.

Photopeaks are assumed to have from one to six decays of different half lives. This assumption is based on the number of nuclides, with prominent gamma ray energies equal to energies included in the

photopeak, believed to be present in the reactor gas. A positive identification of an isotope is made when the half life of that isotope is observed for one of the strong photopeaks of that isotope. Daughter products can be identified by observing the growth of a strong photopeak and then following the decay of that photopeak. A computerized program for the processing of radioactive decay data and analyzing the decay curve for up to 10 components in the curve has been prepared by Rogers [6].

II. EXPERIMENTAL

A. PRODUCTION OF GASES

The gaseous products from the fissioning of the uranium oxide-polyethylene mixture were generated and trapped in an aluminum cylinder and quartz glass vial arrangement similar to that shown in Figure 1. Two fuel discs, about one centimeter thick and 4.5 centimeters in diameter, were placed inside an aluminum cylinder. Each fuel disc is a mixture of 4 grams of UO_2, with 20% enrichment in U^{235}, and 100 grams of polyethylene. The cylinder is sealed at one end by a neoprine O-ring seal and a bellows valve is attached to the other end. The cylinder is then evacuated to a pressure of less than 1 micron. The glass vials were then attached by means of a standard taper joint to the valve and the valve then opened. This introduced a small quantity of air into the cylinder and was useful in diluting the sample for counting purposes. The pressure inside the cylinder and glass vials was found to be approximately 200 microns. The cylinder and the glass vials were then encased in carbon blocks and placed in the access port of the AGN-201 Reactor near but outside of the core. The reactor was operated at a power of 50 watts for 15 minutes subjecting the fuel elements to a thermal neutron flux of approximately 10^9 neutrons per square centimeter per second. Zero time for decay counting purposes was defined as the time at which the reactor was shut down, however, it must be pointed out that the composite age of the gas formed in the

cylinder was from five to twenty minutes old at the start of counting. Even so this method produces a more definite sample of gas than was obtained by previous workers, Bredderman and Laskaris. Their methods of extracting gas from the core tank of the reactor through a copper tubing and manifold system resulted in a sample which contained gas evolved from different operations of the reactor. This gas could not be said to have a specific age as the gas composition would have varied from sample to sample.

B. FRACTIONATION AND EXTRACTION

Upon shut down of the reactor the cylinder was removed and the bottom vial was placed in a cooling bath composed of a slurry of dry ice and trichorolethylene. After one minute of cooling the vial was sealed and removed to the scintillation counter. The upper vial was then placed in a liquid nitrogen cooling bath and cooled for several minutes before being sealed and counted. Several runs were made later in which the gas sample vial received no cooling but was sealed as soon as possible, permitting counting within 2.5 minutes of shut down.

C. MEASUREMENT OF RADIOACTIVE SPECIES

Gas samples were placed in the well of a 3" by 3" sodium iodide scintillation crystal which is coupled to a photomultiplier tube. The resulting impulses of electrical energy are fed into a 512 channel analyzer. The multichannel analyzer was calibrated by utilizing the

prominant photopeaks of Cesium 137 (0.032 MEV and 0.662 MEV), Gold 198 (0.069 MEV and 0.411 MEV), Manganese 54 (0.840 MEV), Cobalt 60 (1.17 MEV and 1.32 MEV), and Sodium 24 (1.370 MEV and 2.753 MEV). The electrical pulses of varying heights which correspond to various energies, are then counted and stored in the memory unit of the multichannel analyzer. The associated equipment with this analyzer includes an oscilloscope for the visual display of stored data, a teletypewriter for printout of total number of counts in each channel, a mechanical plotter for plotting the gamma ray spectrum and a tape punch mechanism for permanent recording of the data. This tape can be fed back into the multichannel analyzer at will for spectrum comparison studies.

Two single channel analyzers were employed to follow the decay of prominent gamma photopeaks. The signal from the photomultiplier tube is passed through a preamplifier and attenuator circuit before reaching the discriminator circuit of the scaler. The discriminator can be adjusted so that an energy window is formed to allow specific pulse height signals to pass to the counter. The single channel analyzers were calibrated in the following manner. First, appropriate calibration of the 512 channel analyzer was made utilizing the standard test samples. Then a spectrum from the previous experiment was fed into the memory core of the 512 channel analyzer. An appropriate energy window was selected from the display on the oscilloscope. A signal from a variable height pulse generator was fed into the multichannel analyzer at the

same point that the signal from the photomultiplier tube enters. The pulse generator is then adjusted so that its signal is counted on the channel corresponding to one edge of the photopeak window. This signal, equaling a specific gamma ray energy, is fed into the pre-amplifier and attenuated to allow counting in the best operating region of the single channel scalers. The appropriate discriminator is adjusted to count this pulse. As a result pulses whose heights fall within the photopeak energy window are counted. Counting intervals with the single channel analyzer varied from 30 seconds to 10 minutes as appropriate to the half lives of the isotopes estimated in the photo-peak. The length of the counting period or experiment usually ran for 300 minutes after shut down of the reactor, although counts were taken as late as 1000 to 1500 minutes after shut down, in order to establish a long term decay component. The photopeaks that were followed by this method were the 0.157, 0.201, 0.224, 0.267, 0.410, 0.455, 0.585, and 1.745 MEV as these were the photopeaks that were strong and showed some indication of short half life components of less than 60 minutes.

D. CHEMICAL ABSORPTION TECHNIQUES

A chemical separation was attempted by bubbling the reactor gas through a solution of sodium sulfite to absorb and insure reduction of the iodine and bromine into their lowest oxidation states. The gas was pumped from the cylinder, located inside the reactor, through three

traps connected in series. The entire system was evacuated initially. A solution containing 140 milliliters of 1M sodium sulfite and 10 milliliters of 1M hydrochloric acid was placed in the first trap. The second and third traps were surrounded by dewar flasks containing liquid nitrogen. The reactor was run at 50 watts for 15 minutes. After shut down the gas was allowed to bubble through the sodium sulfite solution by action of the vacuum pump for 10 to 15 minutes. A quick check with a geiger counter indicated most of the activity had been trapped in the middle trap containing liquid nitrogen. A very small amount of radiation was detectable in the last trap. The sulfite solution was placed in a beaker and counted. The high activity species in the second trap was transferred into an evacuated glass container and also counted.

III. RESULTS

In general the results of this experiment are best given in tabular form or in reproductions of the gamma ray spectra. Reactor gas was produced on twelve separate runs. Three of these runs were for the purpose of obtaining gamma ray spectra only and no significant half life data were measured. Eight successful runs were made for half life determination of photopeaks. One attempt was made to follow the decay of the 0.032 MEV photopeak but results were inconclusive since numerous isotopes have minor gamma and x-ray energies in this region and their resolution is poor. Readings were erratic because of the highly complex decay, growth of daughter products, and Compton scattering from higher energy photons. Gamma ray spectra were also taken during these runs and in all some 174 spectra were obtained. Listed in Table I are the photopeaks and their respective energies. In general the spectra obtained were very similar to those reported by Lindsay with some variations of the energy values. Energies reported by Bredderman are from gas of an indeterminate age which includes daughter products. Energies reported by Lindsay are from gas produced in a similar method described in this report and also includes daughter products. It was found that the multichannel analyzer did not maintain a linear ratio of energy per channel throughout all the channels. Instability in the calibration by as much as 2 channels per 24 hour period was seen. It was observed that the calibration of the multichannel

analyzer holds quite well for a 3 or 4 hour period during the day, however at night drifting occurred. The single channel analyzers did not maintain their calibration as well as hoped. At the completion of runs the same calibrated signals from the pulse generator were fed back into the scalers and it appeared that the discriminators had shifted. The pulse signal generator held its calibration quite well. When a signal was fed from it into the multichannel analyzer no detectable deviation of that signal into adjacent channels could be observed. This was true at the beginning and at the end of a run.

A. EARLY GAMMA RAY SPECTRUM ANALYSIS

Shown at Figure 2 are comparison spectra, 0 to 1 MEV range, of the dry ice cooled vial and liquid nitrogen cooled vial. It was hoped that cooling with dry ice would give a partial fractionation or enrichment of the iodine and bromine isotopes with a resulting smaller amount of krypton and xenon isotopes. Cooling at liquid nitrogen temperature freezes out all of the gaseous isotopes. The significant difference between the dry ice cooled and the liquid nitrogen cooled samples is evident in the 0.455 MEV photopeak which is slightly higher in the liquid nitrogen cooled spectrum. Figure 3 are comparison spectra, 0 to 2 MEV range, between a vial that was cooled with dry ice and a vial that received no cooling at all. No significant differences were observed in comparable spectra between dry ice cooled gas samples and the gas samples which received no cooling. Comparison

spectra between a dry ice cooled gas sample and the chemical absorbed sample are shown in Figure 4. Of significance here is the absence of the prominent photopeaks at 0.155, 0.201, 0.263, 0.432 MEV. A significant change had occurred about an hour later in the doublet photopeak 0.416 and 0.470 MEV. The spectrum taken of the activity trapped in the liquid nitrogen bath was essentially the same as corresponding spectra from a dry ice cooled vial. Figures 5 and 6 are 0 to 2 and 0 to 4 MEV range spectra taken during the 9th run. The decay of the prominent photopeaks is readily apparent and will be discussed in the next paragraph. The growth of certain photopeaks is also noticeable and will be commented upon later.

B. HALF LIFE ANALYSIS

The decays of nine separate photopeaks were followed by the single channel analyzers and used as input data for the "FRANTIC" program. The 0.032 MEV photopeak showed so much change, both increasing and decreasing, that no useful half life measurement could be made. Other decay measurements are summarized in Table II for each photopeak. The "FRANTIC" program corrects the observed data for background, dead time of the counting system, and the decay from zero time. The program then goes through a least squares fit of the data to obtain the decay curve. Included as input parameters to the computer program are the decay constants for the suspected isotopes in the photopeak. These decay constants are not held fixed by the computer

program but are allowed to vary by a factor of three per iteration to obtain the best fit of the curve. Up to fifty iterations are allowed to find the best fit. Calculations were made to fit each curve with a minimum of two components and as many as six components. Various combinations of input decay constants were tried to determine if certain combinations fit better than others. The "FRANTIC" program is designed to accommodate up to 10 components in a decay curve, however, it seems to have difficulty separating components whose half lives differ by less than a factor of three. Several attempts were made to compute decay constants graphically from the corrected data. These decay constants were then used as input into "FRANTIC" to determine a more accurately fitted curve to the set of data points. In some cases the same fit was obtained but with fewer iterations. In other cases a poorer fit was obtained and in some cases a better fit to the curve was obtained. The "FRANTIC" program computes a standard deviation for each data point from the calculated decay curve. The total number of data points used for the calculation and the number of data points deviating more than two standard deviations are indicated in Table II. The weighted mean variance for each curve is also given. The smaller the weighted mean variance the better is the fit of the curve. A weighted mean variance of one is considered a very good fit to a set of experimental data.

GAMMA PHOTOPEAKS IN EARLY SPECTRA OF REACTOR FUEL GAS

Photopeak Number	Energy (MEV)	Previously Reported
1	0.032 ± 0.008	[1] [4]
2	0.155 ± 0.008	[1] [4]
3	0.201 ± 0.008	[4]
4	0.224 ± 0.008	
5	0.263 ± 0.008	[4]
6	0.316 ± 0.016	[1] [4]
7	0.408 ± 0.008	[1] [4]
8	0.432 ± 0.016	[4]
9	0.455 ± 0.008	[4]
10	0.524 ± 0.008	[4]
11	0.539 ± 0.008	[4]
12	0.585 ± 0.008	
13	0.613 ± 0.016	[4]
14	0.654 ± 0.016	[4]
15	0.854 ± 0.008	[1] [4]
16	1.023 ± 0.008	[4]
17	1.231 ± 0.008	[1] [4]
18	1.392 ± 0.016	
19	1.423 ± 0.016	[1] [4]
20	1.596 ± 0.032	
21	1.745 ± 0.008	[4]
22	1.841 ± 0.016	[1] [4]
23	1.999 ± 0.016	[4]
24	2.203 ± 0.016	[4]
25	2.344 ± 0.016	[4]
26	2.392 ± 0.016	[1] [4]

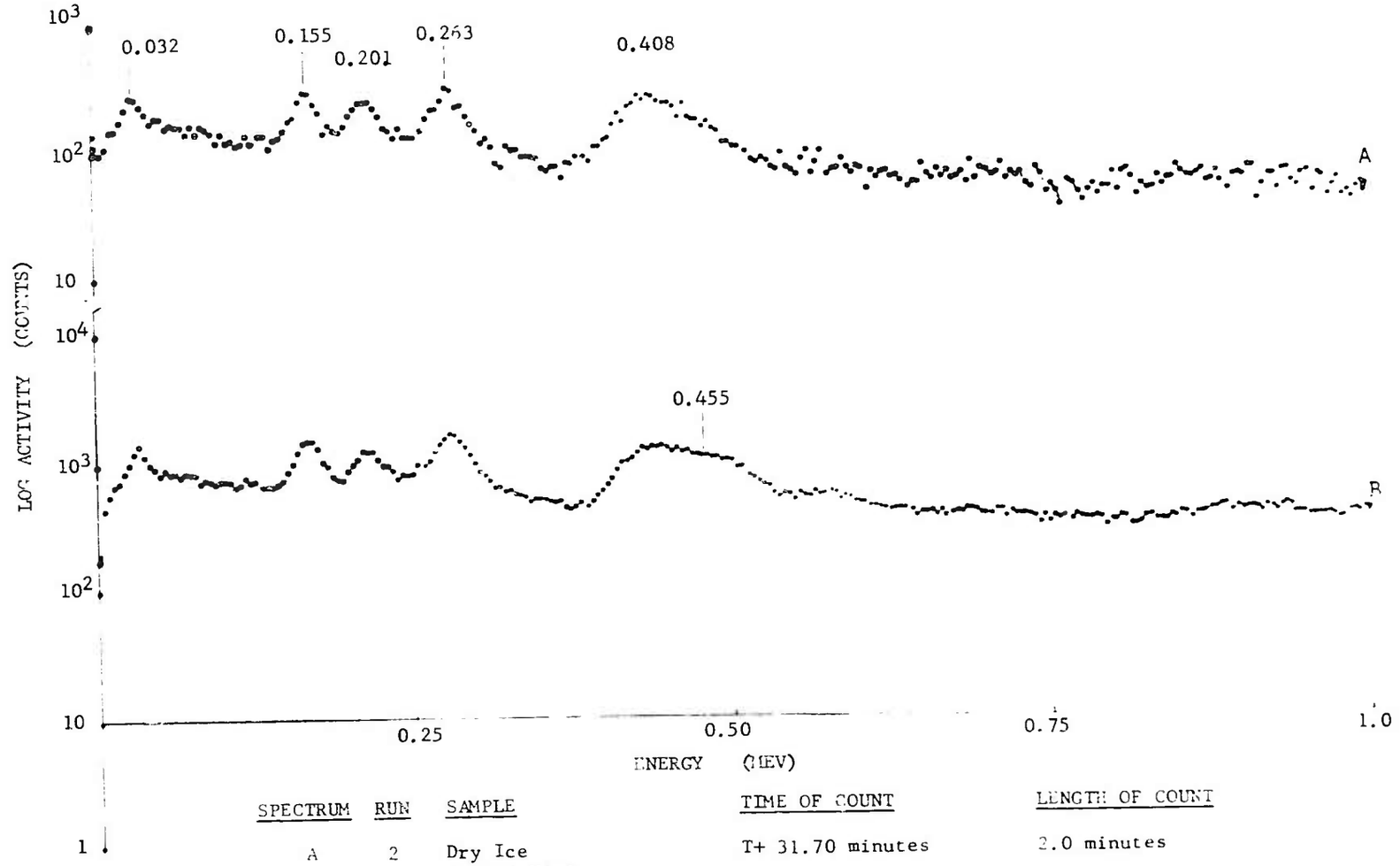
TABLE I

HALF LIVES OBSERVED

Observed Half Life in Minutes

Observed Photopeak	Run	1st Component	2nd Component	3rd Component	Weighted Variance of Fit	Number of Points	Points Deviating 2	Number of Iterations
0.155 MEV	D	2.83	18.2	227.4	0.811	105	3	9
0.201 MEV	E	0.56	5.13	127.0	1.154	100	6	26
0.224 MEV	J	3.39	22.1	131.5	2.799	160	32	12
0.263 MEV	C	2.38	17.6	-----	1.098	34	2	10
	D	3.49	15.2	140.2	1.065	125	3	15
	G-1	4.6	15.0	160.0	0.878	110	2	31
	G-2	3.29	15.0	215.0	0.670	110	3	22
0.408 MEV	E	2.70	14.3	75.1	1.03	105	5	13
0.455 MEV	H-1	2.49	25.6	123.6	2.147	110	5	10
	H-2	2.63	29.9	129.4	1.549	110	5	15
0.585 MEV	I	2.56	13.1	62.3	1.104	90	5	16
1.745 MEV	K-1	2.56	81.5	-----	1.56	130	15	10
	K-2	2.74	109.0	-----	2.167	140	11	26

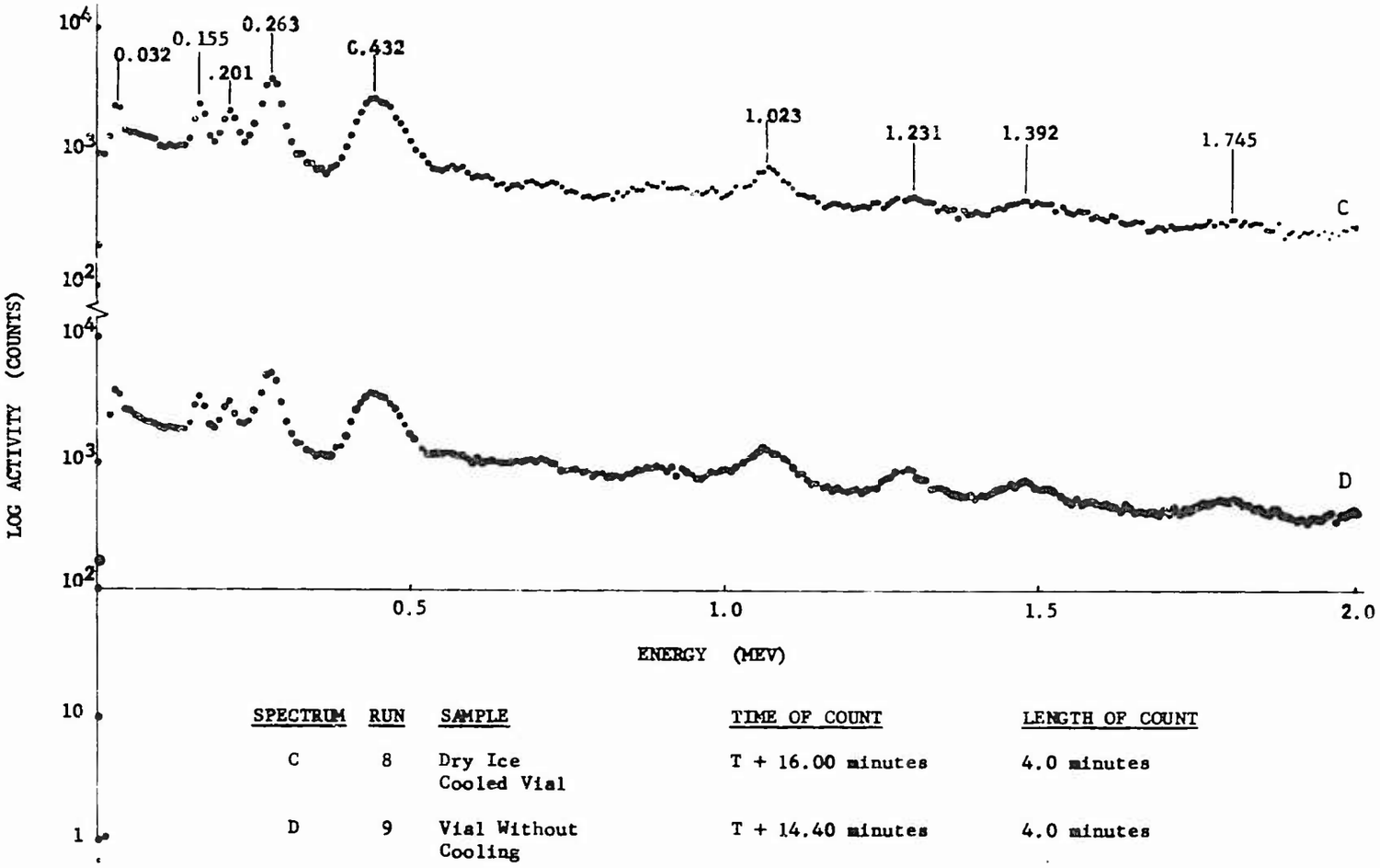
TABLE II



SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
A	2	Dry Ice Cooled Vial	T+ 31.70 minutes	2.0 minutes
B	2	Liquid Nitrogen Cooled Vial	T+ 35.50 minutes	2.0 minutes

Comparison Spectra of Dry Ice and Liquid Nitrogen Cooled Gas Samples.

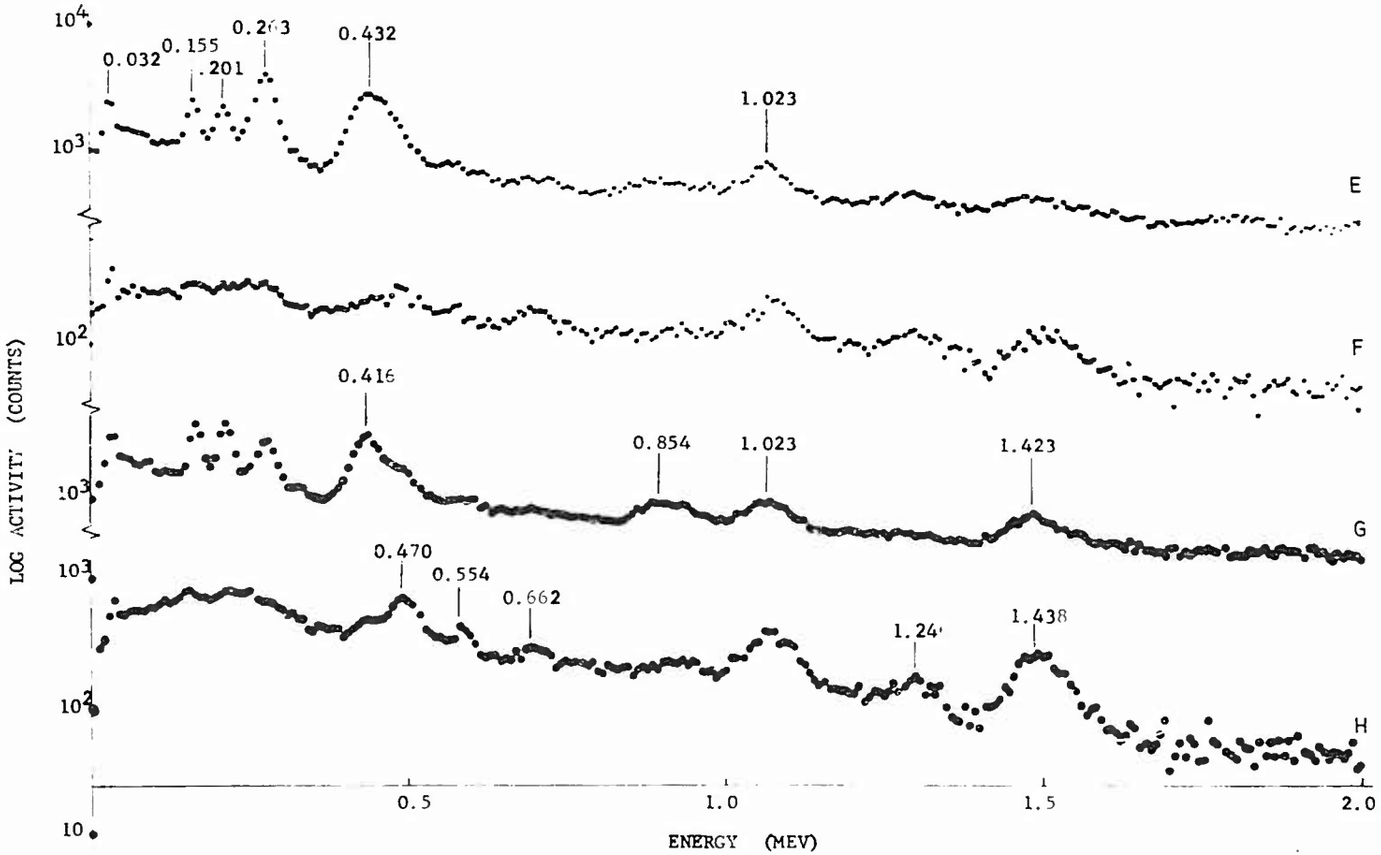
Figure 2



SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
C	8	Dry Ice Cooled Vial	T + 16.00 minutes	4.0 minutes
D	9	Vial Without Cooling	T + 14.40 minutes	4.0 minutes

Comparisou Spectra of Dry Ice Cooled Gas Sample and Sample Without Cooling.

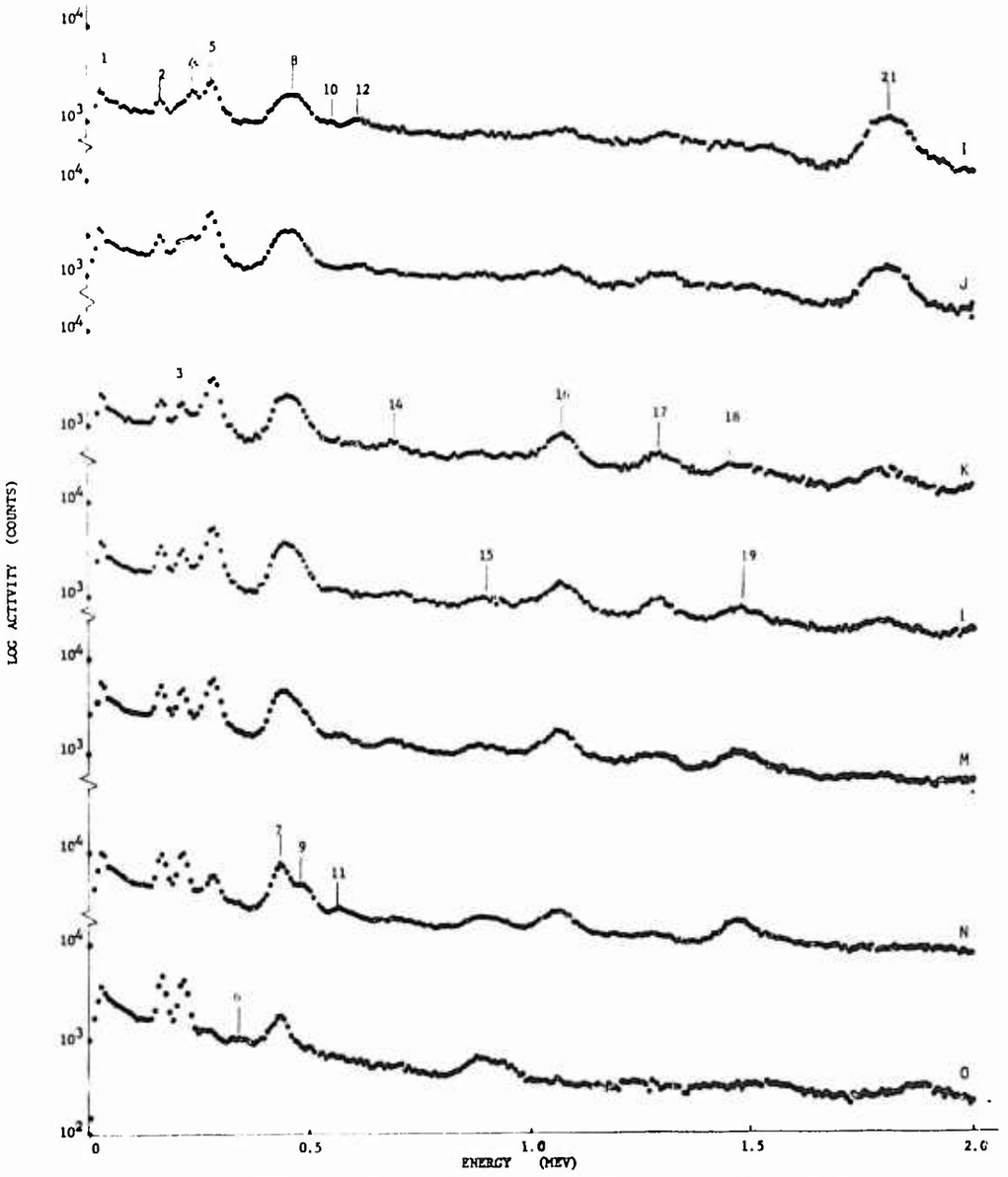
Figure 3



SPECTRUM	RUN	SAMPLE	TIME OF COUNT	LENGTH OF COUNT
E	8	Dry Ice Cooled Vial	T + 16.00 minutes	4.0 minutes
F	12	Absorbed in Solution	T + 18.50 minutes	20.0 minutes
G	8	Dry Ice Cooled Vial	T + 50.00 minutes	10.0 minutes
H	12	Absorbed in Solution	T + 47.50 minutes	20.0 minutes

Comparison Spectra of Dry Ice Cooled Gas Sample and Chemically Absorbed Sample.

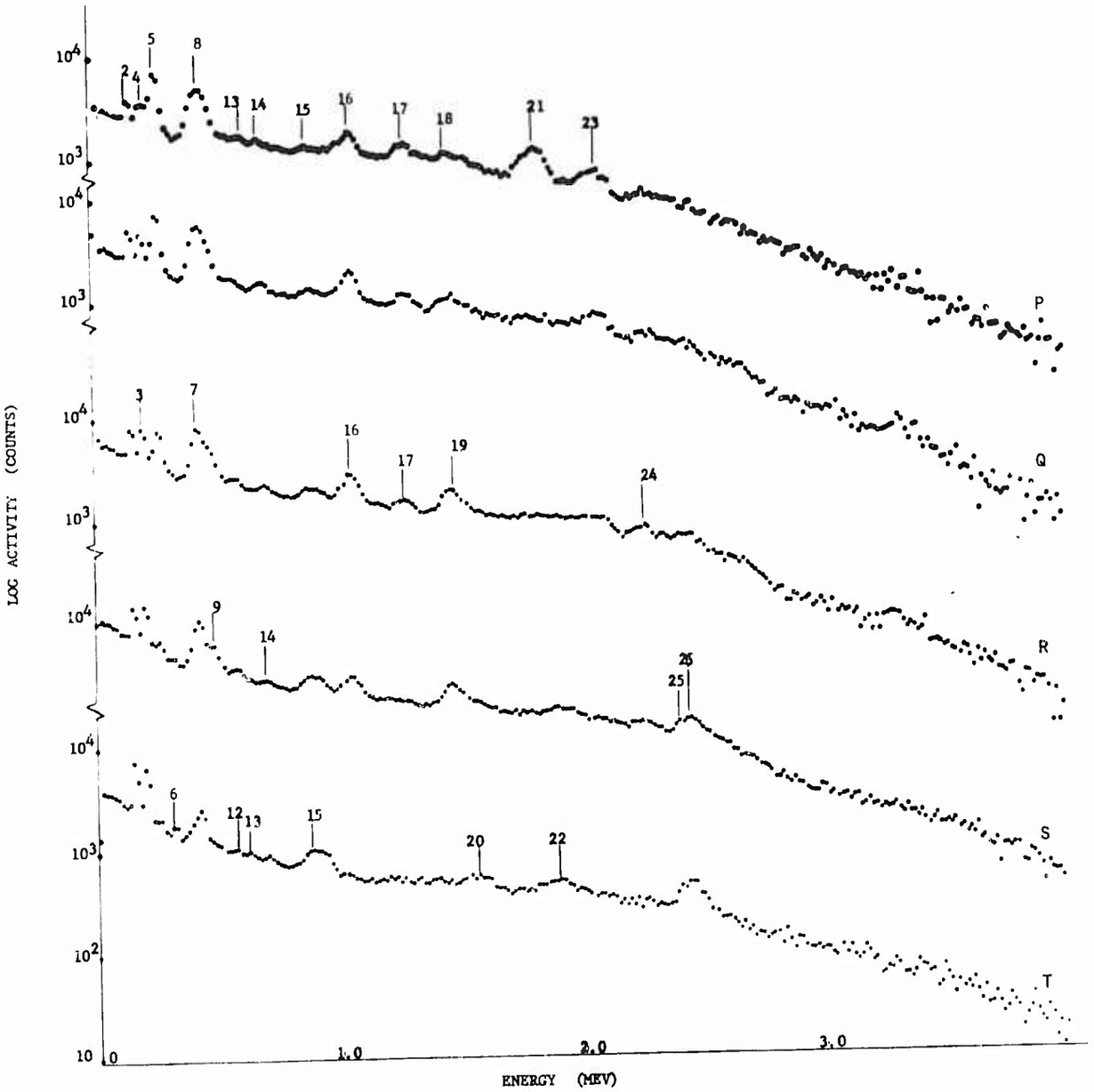
Figure 4



SPECTRUM	RUN	TIME OF COUNT	LENGTH OF COUNT
I	9	T + 2.40 minutes	1.0 minute
J	9	T + 4.75 minutes	2.0 minutes
K	9	T + 11.20 minutes	2.0 minutes
L	9	T + 14.40 minutes	4.0 minutes
M	9	T + 25.00 minutes	8.0 minutes
N	9	T + 50.00 minutes	20.0 minutes
O	9	T + 200.00 minutes	20.0 minutes

Spectrum Decay 0 to 2 MEV Range

Figure 5



SPECTRUM	RUN	TIME OF COUNT	LENGTH OF COUNT
P	9	T + 8.05 minutes	2.0 minutes
Q	9	T + 19.75 minutes	4.0 minutes
R	9	T + 36.10 minutes	8.0 minutes
S	9	T + 75.00 minutes	20.0 minutes
T	9	T + 240.00 minutes	20.0 minutes

Spectrum Decay 0 to 4 MEV Range

Figure 6

IV. DISCUSSION

It became apparent through the course of this experiment that selenium and tellurium isotopes might exist in the radioactive gas products. Selenium and tellurium could possible be present in the form of a hydride such as selenium hydride and tellurium hydride which are gases. Listed in Table III are brief descriptions of each of the selenium, bromine, krypton, tellurium, iodine, and xenon fission products that could possibly be identifiable in the reactor gas. A notation as to photopeak number, and identification by previous workers is provided. Those fission products of the above elements which are stable or have a half life that is so long as to make a half life study impractical are Se^{77}, Se^{78}, Se^{79}, Se^{80}, Se^{82}, Br^{79}, Br^{81}, Kr^{83}, Kr^{84}, Kr^{85}, Kr^{86}, Te^{125m}, Te^{125}, Te^{126}, Te^{127m}, Te^{128}, Te^{129m}, Te^{130}, Te^{131m}, Te^{132}, I^{127}, I^{129}, Xe^{129}, Xe^{131m}, Xe^{131}, Xe^{132}, Xe^{134}, and Xe^{136}. Those fission products whose half life is so short that indication of their presence is not expected are Se^{77m}, Se^{85}, Se^{87}, Br^{88}, Br^{89}, Br^{90}, Kr^{90}, Kr^{91}, Kr^{92}, Kr^{93}, Kr^{94}, Kr^{95}, Te^{135}, I^{137}, I^{138}, I^{139}, Xe^{140}, Xe^{141}, Xe^{142}, Xe^{143}, and Xe^{144}. Also included are comments regarding the daughter products such as rubidium and cesium that may be present. The gamma ray energies and half life of the isotopes listed below are taken from literature values found in works by Goldman and Roesser [7], and Der Mateosian and McKeown [8] who have compiled excellent listings of gamma rays emitted by

radioactive nuclides. The Radiological Health Handbook, published by the U. S. Department of Health, Education and Welfare also contains very useful information on radioactive nuclei [9]. The energies of observed photopeaks may vary somewhat from literature values because of the lack of resolution by the scintillation crystal of two or more gamma rays of similar energies. All of these isotopes decay by beta particle and subsequent gamma emission.

No significance should be made of the third component (See Table II) observed in photopeak 5 (0.263 MEV), because Xe^{135} emits a delayed gamma at 0.25 MEV. This nuclide has been identified by previous workers. The rapid decay of photopeak 21 is caused by the activation of Al^{27} by thermal neutrons to form Al^{28} which then decays by emitting a beta particle and a gamma ray of 1.78 MEV. The half life of Al^{28} is 2.30 minutes, and accounts for the 2.6 minute half life observed in photopeak 21. In a compilation of gamma ray spectra observed in scintillation spectrometry by Heath [10] photopeaks are shown for Al^{28} at 1.78, 1.35, 0.95, 0.40, 0.032 MEV. A gas sample was obtained in which all normal procedures were followed with the exception that the fissionable fuel elements were not placed in the aluminum cylinder. The resulting spectrum showed a strong photopeak at 1.75 MEV which decayed away within twenty minures after reactor shut down. The chemical form of the aluminum in the gas is probably an aerosol suspension of aluminum oxide particles.

POSSIBLE NUCLIDES DETECTABLE IN EARLY GAS SPECTRA

Isotope	Half Life	Detectable Photopeaks Principle	Secondary	Observable in Early Spectra *	Observed Half Life **	Previous Identification
Se^{79m}	3.9 min	0.096		None		
Se^{81m}	56.8 min	0.103		None		
Se^{81}	18 min	0.28		5	15.6 min	
Se^{83m}	69 sec	1.01	2.02	None		
Se^{83}	25 min	0.173	0.368	3 & 4	22.1 min	
Se^{81}	3 min	No Gamma Energies Reported				
Br^{83}	2.4 hrs	0.051		None		
Br^{84m}	6 min	0.88	1.46	None		
Br^{84}	31.8 min	0.879	1.90	None		
Br^{85}	3.0 min	0.91	0.92	None		
Br^{86}	54 sec	1.6	1.4	None		
Br^{87}	55 sec	1.4	2.6	None		
Kr^{85m}	4.36 hr	0.1495	0.304	2 & 6	3.79 hr	[1], [3], [4]
Kr^{87}	76 min	0.403	1.75	7	75.1 min	[1], [3], [4]
				21	81.5 min	
Kr^{88}	2.77 hr	0.191		3 & 4	2.14 hr	[1], [3], [4]
Kr^{89}	3.2 min	0.60	0.22	12	2.56 min	[4] ***
				4	3.39 min	#
Rb^{88}	17.8 min	1.853	0.906	22 & 15	Daughter of Kr^{88}	[3] ***, [4] (See Fig. 5)
Rb^{89}	15.4 min	1.05		16	Daughter of Kr^{89}	[3] ***, [1] *** (See Fig. 5)
Te^{127}	9.3 hr	0.418	0.360	7		
Te^{129}	67.3 min	0.0258	0.475	1 & 9		
Te^{131}	24.8 min	0.15	0.448	2	18.2 min	
				9	27.7 min	
Te^{133m}	50 min	0.334		None		
Te^{133}	2 min	0.6	1.0	None		
Te^{134}	42 min	0.030		1	##	
I^{131}	8.05 day	0.364		None		[3], [4]
I^{132}	2.26 hr	0.677	0.778	None		
I^{133}	20.8 hr	0.54		None		[3] ***, [4] ***

Isotope	Half Life	Detectable Photopeaks Principle	Secondary	Observable in Early Spectra *	Observed Half Life **	Previous Identification
I^{134}	52.5 min	0.86		None		[4] ***
I^{135}	6.7 hr	1.14	1.275	None		[3] *** [4] ***
I^{136}	83 sec	1.32		None		[4] ***
Xe^{133m}	2.35 day	0.233				[1] [2] [4]
Xe^{133}	5.27 day	0.0809				[1] [2] [3]
Xe^{135m}	15.3 min	0.53		12	15.1 min	[3] ***
Xe^{135}	9.2 hr	0.249		5		[1], [3] [4]
Xe^{137}	4.2 min	0.44		7 & 8	2.71 min	[3] ***
				8 & 9	2.56 min	
Xe^{138}	17 min	0.42		7	14.3 min	[3] [4]
Xe^{139}	41 sec	0.22		3	33.6 sec	
Cs^{137}	30 yr	0.662			Daughter of Xe^{137}	***
Cs^{138}	32.2 min	1.436		19	Daughter of Xe^{138}	[3], [4]
Cs^{139}	9.5 min	1.28	0.63	17 & 14	Daughter of Xe^{139}	

* Photopeak numbers from Table I and Figures 4 and 5.

** Half Life values from Table II and averaged where appropriate.

*** Photopeak identification with no supporting half life data.

5.13 min half life in photopeak 3 may be caused by interference from this isotope.

This isotope is probably the major contributor to photopeak 1. Other contributions from Te^{129} and from low energy gamma rays and x-rays from other isotopes are likely.

This isotope is not observable in early spectra but is detectable in the background spectra at the reactor facility. This species has probably built up through out the laboratory by long term leakage from the reactor control rods.

TABLE III

V. CONCLUSIONS

Radioactive nuclides present in the gaseous discharge from the uranium fuel of the AGN-201 reactor at the Naval Postgraduate School are summarized in Table IV. The technique employed is adequate to obtain a gas sample for the determination of half lives between 0.5 and 20 minutes. The absence of the strongest photopeaks for bromine and iodine gases in the initial spectra suggests the bromine and iodine fission products are largely trapped in the matrix of the uranium-polyethylene fuel. While there may be trace quantities of these halogens present in the gas, they cannot be identified.

NUCLIDES IDENTIFIED

Initial Volatile Nuclides:

Te^{131}

Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{135}, Xe^{137}, Xe^{138}, Xe^{139}

Se^{81}, Se^{83}

Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, Kr^{89}

Active Daughters:

Cs^{137}, Cs^{138}, Cs^{139}

Rb^{88}, Rb^{89}

I^{131}

Nuclides Possibly Present*

I^{132}, I^{133}, I^{134}, I^{135}, I^{136}

Te^{127}, Te^{129}, Te^{134}

Br^{83}, Br^{84m}, Br^{84}, Br^{85}, Br^{86}, Br^{87}

*Based on photopeak analysis with no supporting half life data.

TABLE IV

APPENDIX A

EQUIPMENT

1. Counting Chamber: A 2-inch thick lead cube, 22 inches square by 28 inches high, lined with 1/8-inch aluminum and 1/4-inch high copper content brass.

2. Scintillation Detector: Harshaw 12SW12-W3, 3-inch by 3-inch cylindrical well type NaI (Th) crystal with matched photomultiplier tube.

3. 512 channel multichannel analyzer, consisting of the following components:

   a. Nuclear Data, Inc. ND-180F Analog to Digital Converter

   b. Nuclear Data, Inc. ND-180M 512 Channel Memory Unit

   c. Nuclear Data, Inc. ND-180R Read out Control Unit

   d. Textronix, Inc. Rm503 Oscilloscope

   e. Tally Corp. Model 1506 Reader/Perforator

   f. Teletype Corp. Teletypewriter

   g. Hewlett-Packard, Co., Mosley 7590 CR Automatic Plotting System

4. Single channel analyzer (2 ea): Hewlett-Packard Model 5201L Scaler-Timer

5. Mercury Switch Pulser: Hamner Model NP - 10.

6. Vacuum System: A glass vacuum system with a CENCO HYVAC 2 pump and an oil diffusion pump. Pressure measurement: Thermocouple gauge calibrated to 1 micron. (Pressures actually used were lower than this and were estimated to be 10^{-4} mm when the meter was "pegged").

7. Reaction Vessel: A cylindrical aluminum contained approximately 2 x 5 inches fitted with a vacuum stainless steel valve.

8. Fuel: 8 grams of UO_2, 20% enrichment in U^{235}, in approximately 200 grams of polyethylene.

BIBLIOGRAPHY

1. Bredderman, R. T., Radioactive Gaseous Effluents from Core of the AGN-201 Reactor at the United States Naval Postgraduate School, NPS Thesis, 1966.

2. Laskaris, G. C. A., Simultaneous Gas Chromatographic and Activity Analysis of the Effluent Gases from the AGN-201 Reactor at the Naval Postgraduate School, NPS Thesis, 1967.

3. Cole, L. W., Analysis of Volatile Reactor Fuel Decomposition Products by Gas Chromatographic Methods, NPS Thesis, 1968.

4. Lindsay, L. A., Analysis of Volatile Reactor Fuel Decomposition Products by Spectral Analysis, NPS Thesis, 1968.

5. Katcoff, S., Nucleonics, 18, 201, 1960.

6. Rogers, P. C., FRANTIC Program for Analysis of Exponential Growth and Decay Curves, Massachusetts Institute of Technology, Technical Report No. 76, 1962.

7. Knolls Atomic Power Laboratory, The Chart of the Nuclides, 9th Edition revised 1966.

8. Der Mateosian, E. and M. McKeown, Table of Gamma-Rays Emitted by Radioactive Nuclei, Brookhaven National Laboratory, 1960.

9. U. S. Department of Health, Education, and Welfare, Radiological Health Handbook, 1960.

10. Heath, R. L., Scintillation Spectrometry Gamma-Ray Spectrum Catalogue, Atomic Energy Commission Research and Development Report IDO-16408, 1957.

11. Williams, E. T. and C. D. Coryell, Journal Inorganic & Nuclear Chemistry, 28, 682, 1966.

Security Classification

DOCUMENT CONTROL DATA - R & D

Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)	2a. REPORT SECURITY CLASSIFICATION
Naval Postgraduate School Monterey, California	Unclassified 2b. GROUP

3. REPORT TITLE

Search for Short Lived Isotopes in Volatile Reactor Fuel Products

4. DESCRIPTIVE NOTES (Type of report and, inclusive dates)

Master's Thesis; June 1969

5. AUTHOR(S) (First name, middle initial, last name)

Ward Duane Fagan

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
June 1969	39	11

8a. CONTRACT OR GRANT NO	9a. ORIGINATOR'S REPORT NUMBER(S)
b. PROJECT NO	
c.	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
d.	

10. DISTRIBUTION STATEMENT

~~Distribution of this document is unlimited~~

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	Naval Postgraduate School Monterey, California 93940

13. ABSTRACT

The volatile radioactive nuclides produced by thermal neutron fissioning of uranium oxide-polyethylene fuel of the AGN-201 Reactor were collected in an evacuated aluminum cylinder. The following constituent isotopes, identified by gamma ray spectrometry, present in 20 minute old gas are; Te^{131}, Xe^{131m}, Xe^{133m}, Xe^{133}, Xe^{135m}, Xe^{137}, Xe^{138}, Xe^{139}, Se^{81}, Se^{83}, Kr^{85m}, Kr^{85}, Kr^{87}, Kr^{88}, and Kr^{89}. Radioactive daughter products identified are Cs^{137}, Cs^{138}, Cs^{139}, Rb^{88}, Rb^{89}, and I^{131}. Other isotopes of iodine, tellurium, and bromine may be present. Spectra were taken with a NaI (Th) scintillation crystal.

DD FORM 1 NOV 65 1473 (PAGE 1)

S/N 0101-807-6811

Security Classification

A-31408

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
gamma-ray spectrum						
fission products						
gas						
selenium						
bromine						
krypton						
tellurium						
iodine						
xenon						
isotopes						