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INSTALLATION AND TESTING
OF A SPENT FUEL AGE MONITOR
AT NPD

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INSTALLATION AND TESTING OF A
SPENT FUEL AGE MONITOR AT NPD

By

A. D. Vaughn
D. E. Christensen

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PACIFIC NORTHWEST LABORATORIES
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A. D. Vaughn and D. E. Christensen

ABSTRACT

A spent fuel age monitor has been installed in the spent fuel bay at the NPD Reactor, Rolphton, Ontario, Canada, for the purpose of determining cooling time of NPD irradiated fuel assemblies from gamma-ray measurements. The operation of the monitor is a safeguards experiment planned by the Joint Canada/USA Working Group on Safeguards Instrumentation as part of the Canada/USA Tamper Resistant Unattended Safeguards Techniques Program.

The details of shipping, installation, calibration and the initial testing results are presented. A significant result of the calibration measurements was that an operator acquainted with the decay characteristics of fission-product gamma-ray spectra estimated the cooling time with fair accuracy by visual inspection of the spectra. The initial testing results indicated that the cooling time measurements can be obtained with the same techniques demonstrated during the cooling time measurements of KE Reactor fuels and as reported in BNWL-1575.

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INSTALLATION AND TESTING OF A SPENT FUEL AGE* MONITOR AT NPD

A. D. Vaughn and D. E. Christensen

INTRODUCTION

A spent fuel age monitor has been installed at the Nuclear Power Demonstration (NPD) Reactor at Rolphton, Ontario, Canada, as part of the Canada/USA Tamper Resistant Unattended Safeguards Techniques Program (TRUST). The monitor was installed in the NPD spent fuel bay and its operation is a safeguards experiment planned by the Joint Canada/USA Working Group on Safeguards Instrumentation to determine the age of spent fuel irradiated at the NPD Reactor.

The spent fuel monitor, which is primarily a gamma-ray spectrometer, was designed, fabricated and tested by Battelle Northwest to demonstrate the age determination of spent fuels. The demonstration was performed under contract with the U. S. Atomic Energy Commission and testing of the monitor was done⁽¹⁾ at Hanford's KE Reactor spent fuel bay utilizing KE Reactor irradiated fuels. The monitor was removed from the spent fuel bay in early April, 1971, after having been tested since December, 1970. The equipment was shipped from the Hanford plant at Richland, Washington, on April 7, 1971, and arrived at NPD on April 21, 1971, for installation into the spent fuel bay at NPD. The installation was completed on April 23, 1971, and a calibration and testing program followed.

This report presents a discussion of the principles of age determination as well as a summary of the results from the KE Reactor demonstration experiment, the details of the installation of the spent fuel monitor and results from the

* Age of spent fuel is defined as the time in days since the irradiated fuel was discharged from the reactor.

initial testing program. The initial results are also presented from a controlled measurement program which was started at NPD using six irradiated natural-uranium fuel assemblies to derive the variation in fission-product decay rates with respect to the age of the assemblies. The controlled measurement program is still in progress and this report should be considered as only the initial documentation of the measurement results.

PRINCIPLE OF THE MEASUREMENT AND RESULTS FROM KE REACTOR

Intense gamma-ray activity due to radioactive fission products is an identifying property of irradiated nuclear fuels. The age of discharged fuel is also an identifying characteristic and the age can be obtained from measurement of the gamma-ray activity associated with irradiated fuel. Due to the variations in half-lives of the radioactive fission products, the gamma-ray intensities emitted at various energies decay differently as a function of cooling time. The characteristics of fission-product gamma-ray spectra i.e., the number and relative height of photopeaks and the shape and relative height compared to photopeak height of the continuum between photopeaks, are continually changing as short-lived isotopes disappear and long-lived isotopes begin to appear. Fission-product spectra obtained with a sodium iodide detector are presented in Figure 1 as a function of cooling time up to 100 days. The fission products whose contributions to the gamma-ray spectra are most evident in Figure 1 are listed in Table 1. The changing characteristics of such spectra are readily recognizable to the point that an operator acquainted with fission-product spectra can make a good estimate of the age by visual inspection of the spectra.

FIGURE 1. Fission-Product Gamma-Ray Spectra as a Function of Cooling Time Obtained with a Sodium Iodide Detector. Single-channel analyzer settings used for measurements are indicated.

TABLE 1. Radioactive Fission-Product Isotopes Which Contribute to Prominent Photopeaks Evident in Sodium Iodide Spectra up to a Cooling Time of 100 days.

KeV ⁽²⁾	Fission Product*	Half-Life	Region (Figure 1)
487	$^{140}\text{Ba} - ^{140}\text{La}$	12.8 days - 40.2 hours	} Peak 1
496	^{103}Ru	39.8 days	
724	^{95}Zr	65 days	} Peak 2
756	^{95}Zr	65 days	
764	$^{95}\text{Zr} - ^{95}\text{Nb}$	65 days - 35 days	
1600	$^{140}\text{Ba} - ^{140}\text{La}$	12.8 days - 40.2 hours	Peak 3
2186	$^{144}\text{Ce} - ^{144}\text{Pr}$	284 days - 17.3 minutes	

* Gamma rays from $^{106}\text{Ru} - ^{106}\text{Rh}$ (367 days-30 seconds) and ^{137}Cs (30.0 years) do not become evident in sodium iodide spectra until after 100 days cooling time.

The determination of age of discharged fuel thus takes advantage of the differences in decay rates of fission products and is based on forming ratios of the measured activities from the different fission products. These principles are easily implemented with a sodium iodide detector and an appropriate collimating system even though the energy resolution of sodium iodide is low and individual photopeaks present in fission-product gamma-ray spectra are not usually separated. Single-channel analyzers in conjunction with scalers are used and set so that they indicate a given energy response of the detector. Various energy regions useful during measurement are indicated in Figure 1 and from the measurements, ratios such as Peak 3 to Peak 2 (that is, $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$), Plateau 2 to Plateau 1 and Peak 1 to Peak 2 ($^{140}\text{Ba} - ^{140}\text{La} + ^{103}\text{Ru}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$) are formed. The ratios are an age index which, when

compared to a standard, indicate the number of days that have elapsed since the fuel was discharged. A basic requirement of the technique is that the standard, that is, a graph of the activity ratios as a function of cooling time in days (see Figure 2) or a meter scale, needs to be derived from a controlled measurement program.

In order for the standard to be applicable to fuel of different exposures, irradiation histories, design and cooling times, a prime consideration during the controlled measurement program is to select fuel assemblies which will span the variations that may affect the ratio values. For the activity ratios to be a true age index for all irradiated fuel from a given reactor, they should be insensitive to all variables except cooling time. Using first weekly and then monthly measurement results, a standard can be derived that will be applicable to all fuel irradiated in a given core. Useful, also, to support age determination measurements is a theoretical calculation of the gamma-ray intensities and ratios which can be expected from a given fuel based on its irradiation history. Particularly useful are calculations of how the intensities decay as a function of cooling time and which ratios are the most appropriate to use. A theoretical curve of the $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$ ratio is also included in Figure 2 for comparison to the experimental curve.

Initially the slopes of both the experimental and calculated curves shown in Figure 2 are the same for the $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$ ratio. However, the calculation does not take into consideration any Compton, electron or other continuum on which the various photopeaks are superimposed. The Compton spectra arise from gamma-ray interactions within the detector which do not result in total energy loss of the gamma rays. Thus, partial energy loss of gamma rays of higher energy than

FIGURE 2. Measured and Calculated Activity Ratios for KE Reactor Fuel. The measured values resulted from the Controlled Measurement Program and serve as the standard for age determinations for KE Reactor fuel.

the 1600 keV gamma ray of $^{140}\text{Ba} - ^{140}\text{La}$, for example, the 2186 keV gamma rays from $^{144}\text{Ce} - ^{144}\text{Pr}$, contributes to a Compton spectrum which the 1600 keV photopeak is superimposed upon. The Compton spectrum decays slower and is still present when the 1600 keV gamma ray from $^{140}\text{Ba} - ^{140}\text{La}$ has completely decayed to zero. The presence of the Compton spectrum is the reason that the experimental curve begins to flatten as the cooling time approaches 100 days since the single-channel analyzer makes no distinction between 1600 keV gamma rays from $^{140}\text{Ba} - ^{140}\text{La}$ and 1600 keV Compton contributions from $^{144}\text{Ce} - ^{144}\text{Pr}$.

For cooling times greater than 100 days and less than 150 days, the $^{140}\text{Ba} - ^{140}\text{La}$ photopeak will become weaker and eventually disappears. The strategy then, which is supported by calculations, is to ratio longer-lived fission product contributions, for example, $^{95}\text{Zr} - ^{95}\text{Nb}$ to $^{144}\text{Ce} - ^{144}\text{Pr}$, in order to obtain an age index at cooling times greater than shown in Figure 2.

The data shown in Figures 1 and 2 resulted from the experimental measurements and the theoretical calculations for KE Reactor fuel.⁽¹⁾ During the application of the above principles at the KE Reactor using a sodium iodide system, eight fuel pieces of the same design and known irradiation history were used for the controlled measurement program. The exposures of the fuel pieces varied by a factor of eight from minimum to maximum exposure. The $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$ ratio proved to be relatively insensitive to exposure thus providing a useful age index. The remaining ratios, Plateau 2 to Plateau 1 and $^{140}\text{Ba} - ^{140}\text{La} + ^{103}\text{Ru}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$, were observed to vary approximately 4% with exposure. These ratios were nevertheless useful when used concurrently with the $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$ ratio to determine age. As a result of the measurements, a standard in the form of a graph of the

activity ratios as a function of cooling time was derived for cooling times between ten and ninety days. By use of the standard it was possible to make age determinations to within a statistical uncertainty of plus or minus three days.⁽¹⁾ The demonstration experiment at KE Reactor was thus successful and was the basis to proceed with a like demonstration using a sodium iodide detector at NPD. As a result of the experience gained, an operating manual for the experimental program to be carried out at NPD was written to aid installation and operation of the monitor at NPD.

In addition to the sodium iodide data obtained from KE Reactor irradiated fuels, gamma-ray spectra were collected using a high resolution germanium-lithium-drifted detector. The results from these measurements as they apply to age determination are presented in Appendix A.

SHIPPING, INSTALLATION AND INITIAL CALIBRATION

The components which comprise the collimator assembly of the fuel age monitor are shown in Figure 3. The unit was designed for underwater measurements and special features included a variable aperture, hydraulic fuel lift and an air tube to inject air and displace water in the collimator head to insure an in-air path for gamma rays from the fuel contact to the detector, except for a 1/8 inch aluminum water seal in the collimator head. Rubber tips fashioned from rubber tubing to match fuel assembly dimensions were put on the bottom of the collimator head for contact with the irradiated fuel. These components as well as the associated electronic equipment listed in Table 2 were shipped to NPD after being removed from the spent fuel storage area at KE Reactor. A fuel handling device was also fabricated and shipped. There was no damage

FIGURE 3. Collimator Assembly - Fuel Age Monitor

to any of the monitor components during shipment. The electronic equipment arrived in excellent condition and has worked perfectly during initial testing.

TABLE 2. Components of the Fuel Age Monitor Including Associated Electronic Equipment Shipped to NPD, Rolphton, Ontario, Canada

<u>Quantity</u>	<u>Description</u>	<u>Quantity</u>	<u>Description</u>
1	Collimator Shield Assembly.	1	High Voltage Power Supply, RIDL 40-9B.
1	Collimator Tube Assembly.	1	Analog Stabilizer, Scintimatic 39-4.
1	Variable Aperture Assembly.	5	Single Channel Analyzer, TC-441.
1	Hydraulic Lifting Assembly.	5	Scaler.
1	Hydraulic Hand Pump.	1	Timer-Scaler, RIDL 54-S.
1	Fuel Handling Tool and Fuel Handling Rod.	3	BIN Power Supply.
1	Sodium Iodide Detector, 2 in. by 2 in. Harshaw laced with ^{241}Am . 3.0 Mev photopeak for Stabilizer.	1	Instrument Storage Bin.
1	Preamplifier, ORTEC 113.	1	Multi-Channel Analyzer, Nuclear Data 130 (512 channels).
1	Amplifier	1	Oscilloscope, Textronic RM 561.
		1	MCA Power Supply.
		1	Box of Miscellaneous Cables.

Installation began on April 21, and was completed on April 23. Some minor modifications to the cradle holding the collimator were necessary. In addition, two hydraulic cylinders (see Figure 3) had been purchased to replace those used at KE Reactor. The cylinder replacements were done at NPD as well as replacement of the hydraulic hand pump which was found defective after installation. Installation of the fuel age monitor

was a relatively smooth operation with only minor difficulties being encountered during installation and with operation of the hydraulic lifting tray. The final setup of all components and electronic equipment is shown in a block diagram in Figure 4.

The initial calibration of the sodium iodide system was performed by selecting one 19-rod cluster assembly at random from the available NPD irradiated fuel assemblies. It was expected that a relatively short-cooled assembly (less than 15 days cooling time) had been obtained. However, with the first spectrum taken, it was noted that the 2186 keV photopeak from $^{144}\text{Ce} - ^{144}\text{Pr}$ was visible and the 1600 keV photopeak of $^{140}\text{Ba} - ^{140}\text{La}$ had decreased even further beyond that shown in Figure 1. The operator was familiar with the decay characteristics of fission-product spectra and estimated that the assembly had been out of the reactor in excess of 100 days and less than 150 days. Records were consulted at that point and they indicated that the assembly had cooled 129 days. Thus, the calibration measurements resulted in an immediate demonstration of age determination.

INITIAL TESTS AND THE CONTROLLED MEASUREMENT PROGRAM

After the monitor and equipment were installed and operating satisfactorily, a controlled measurement program was initiated to derive the variation in fission-product decay rates as a function of the age of spent fuel from NPD Reactor. Six irradiated natural-uranium fuel assemblies were selected on the basis of providing an adequate representation of the various irradiation histories and exposures of assemblies that are discharged from the NPD core. The assemblies are identified in Table 3 and were stored in Basket 38 which was placed adjacent to the collimator. The assemblies were discharged from the reactor on April 10 through 14, 1971, and were 10 to 14 days out of the reactor when measurements were started.

FIGURE 4. Block Diagram of Sodium Iodide Detector and Instrumentation

TABLE 3. Fuel Assemblies Being Used for Controlled Measurement Program of NPD Fuel

<u>Assembly Number</u>	<u>Date Charged</u>	<u>Date Discharged</u>	<u>Reactor Location*</u>	<u>Estimated Exposure MWd/MTU</u>	<u>Storage** Location Basket 38</u>
DB035	2/3/67	4/10/71	F6	6,680	C4
6808	6/21/66	4/14/71	G10	10,150	C5
60036	2/22/66	4/11/71	E2	6,890	A5
GC002W	1/17/68	4/14/71	E8	6,580	A1
BC029W	4/1/68	4/14/71	F6	6,400	A3
HB013W	10/21/66	4/14/71	G10	9,450	A4

* The NPD Reactor is a 12 by 12 array of horizontal fuel channels. Each channel is designated by giving the vertical component a letter designator of A through M (skipping I) and a horizontal component of 1 through 12.

** Basket 38 has 25 storage locations. Each storage location is designated by giving the horizontal component a letter A through E and a vertical number component 1 through 5.

Several tests were performed using the fuel assemblies listed in Table 3. Measurements were made for the purpose of experimenting with aperture diameters with respect to fuel of different irradiation histories and exposures. It was determined that fuel assemblies discharged approximately 10 days and which were discharged from the central regions of the core can be counted using a 5/32 inch diameter aperture. Fuel assemblies irradiated in the outer regions of the reactor vessel require a larger aperture diameter of 1/4 inch. A three minute counting interval provided reasonable statistical accuracy. During these measurements, it was discovered that the fuel lifting tray was not raising properly due to a faulty hydraulic hand pump and the hand pump was therefore replaced.

Some difficulties were encountered in moving the fuel from storage to the spectrometer fuel tray. It was not possible to use either the fuel handling tool provided by Battelle Northwest

or the cylindrical tool normally used at NPD due to the fragile nature of the assemblies. The most satisfactory method developed for fuel handling was to maintain the assemblies in an aluminum can during measurement and storage. The cans were designed with a wire lifting bail at the top which permitted the can to be moved from storage and to be tipped to a horizontal position onto the fuel lifting tray. Also, tests were made with assembly 6808 to determine the counting rates when a) the assembly was removed from its aluminum container and counted directly, b) the assembly was contained in the aluminum container and counted when the collimator head made contact with the can and c) the assembly was contained in the aluminum container but now the container had a slot cut in its side (eight inches long by two and one half inches wide) to permit the collimator head to make contact with the assembly during counting. From these tests, it was decided to use aluminum containers with a slot in the side for all measurements to minimize the possible interference due to the container wall and extraneous water in the beam path. This method of fuel movement and counting maintained the integrity of each assembly although the operation of placing the assemblies on the lifting tray was more difficult than expected, requiring two men to position an assembly on the tray.

A third test was performed at the beginning of the controlled measurement program to determine the effect of a 1/4 inch thick lead filter when the filter was inserted in the beam path directly below the detector. The measurements were made using assembly 6808 and are summarized in Table 4. The count rates were satisfactory without the lead filter so that a filter was not required as long as the 5/32 inch diameter aperture was used.

TABLE 4. Gamma-Ray Activity Measurements of Assembly 6808 With and Without a 1/4 Inch Thick Lead Filter Below the Detector. Cooling time was 15 days.

Channel #1	Channel #2	Counts			Count Interval	Aperture Diameter	Date	Remarks	Ratios		
		Channel #3	Channel #4	Channel #5					4/1	2/3	5/3
838541	773526	615206	151585	231361	300 Sec.	5/32 in.	4/29	With Filter	0.1808	1.257	0.3761
912998	835824	651185	161815	248375	↓	↓	↓	Without Filter	0.1772	1.283	0.3814
816663	759499	610130	151054	231749				With Filter	0.1850	1.245	0.3798
817509	758858	609972	149872	231024				With Filter	0.1833	1.244	0.3787
907546	835553	652011	160216	249518				Without Filter	0.1765	1.282	0.3827

Channel #1 Plateau #1, Continuum and Background.
Channel #2 Peak #1, ^{140}Ba - ^{140}La and ^{103}Ru Gamma Rays.
Channel #3 Peak #2, ^{95}Zr - ^{95}Nb Gamma Rays.
Channel #4 Plateau #2, Continuum and Background.
Channel #5 Peak #3, ^{140}Ba - ^{140}La Gamma Ray.

AGE MEASUREMENT STANDARD FOR NPD FUELS

A typical fission-product gamma-ray spectrum obtained from measurements of NPD fuel assembly 6808 with a sodium iodide detector is presented in Figure 5. The cooling time of the assembly when the spectrum was obtained was 15 days and the spectrum is shown as viewed on the oscilloscope. The multi-channel pulse height analyzer was calibrated such that the ^{241}Am peak (for stabilization) was positioned near the vertical scale line at the far right of the spectrum. Except for the ^{241}Am photopeak, the spectrum is identical in detail to the short-cooled spectra seen in Figure 1. The three prominent photopeaks were identified (from left to right) as the ^{140}Ba - ^{140}La plus ^{103}Ru gamma rays at approximately 500 keV, the ^{95}Zr - ^{95}Nb gamma rays at approximately 750 keV and the ^{140}Ba - ^{140}La gamma ray at 1600 keV.

Theoretical calculations were made of the gamma-ray intensities and ratios that can be expected from NPD fuels and were reported in Reference 1. The results of the calculations indicated that the decay of the fission-product isotopes should provide adequate data with which to determine the age of the NPD irradiated fuel.

The measurements made during the initial testing also provided the initial values of the intensity ratios which are an index for the age of the NPD discharged fuel. The ratio values determined from the measurements of fuel assembly 6808 with and without a filter are given in Table 4. The values agree closely to the values obtained for KE Reactor fuels at 15 days cooling time as can be seen by plotting the NPD values on Figure 2.

FIGURE 5. Fission-Product Gamma-Ray Spectrum of Assembly 6808 After 15 Days Cooling. The spectrum was obtained using a sodium iodide detector. (No filter was used)

CONCLUSIONS

The installation of the spent fuel age monitor and its associated electronic equipment at NPD is complete and the instrument is in operation scanning spent fuel assemblies as a safeguards experiment. The installation and testing phase progressed smoothly and the instrument was brought on line with very few problems. At present it is being operated by a recent University graduate with no previous gamma-scanning experience.⁽³⁾ This was possible as a result of the use of the operating manual and some limited training.

The calibration and initial testing measurements of gamma-ray intensities emitted from NPD irradiated fuel assemblies indicated that techniques similar to the methods used for KE Reactor fuels are applicable to NPD spent fuels to determine a standard for age measurements. Based on the close agreement

of the activity ratios obtained from measurements after 15 days cooling time from both NPD and KE Reactor spent fuels, it is expected that a standard involving curves very similar to those shown in Figure 2 will be the final result of the controlled measurement program at NPD. A significant result of the calibration measurements was that an operator acquainted with the decay characteristics of sodium iodide spectra can estimate the age of discharged fuel with fair accuracy by visual inspection of the fission-product spectra.

The controlled measurement program is continuing and final documentation will be done upon completion of the experiment. Interim results from measurements through June 23, 1971, are given in Appendix B.

ACKNOWLEDGEMENTS

Many individuals were involved in shipping the monitor from Richland and installing it at NPD. Personnel of Douglas United Nuclear and ITT Federal Support Services were efficient in preparing and crating the collimator for shipping and in packaging the electronic equipment. The equipment was delivered to Mr. Ron Cecil and Mr. Emerson Schilke, maintainer at NPD, became knowledgeable with the mechanical components and performed an excellent task in adapting the components to the NPD spent fuel bay. Mr. Bob Taggert, operator in training, made the necessary fuel assembly movements from storage to the assembly holding tray of the monitor. His careful assistance was appreciated.

A number of other people were involved in providing NPD operating specifications, operating histories and physics parameters which were necessary to successfully install the monitor and to calculate fission-product intensities of NPD fuel. In addition to Mr. Ron Cecil mentioned previously, the cooperation of Mr. J. B. Ogg, Mr. J. Griffiths and Mr. W. H. Walker of AECL

and Mr. W. D. Smythe of AECB was appreciated. In particular, the many discussions with Mr. J. G. McManus, Associate Scientific Advisor - Safeguards, AECB, were invaluable.

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APPENDIX A

APPENDIX A

The collimator assembly of the fuel age monitor was designed such that both sodium iodide and germanium-lithium-drifted, Ge(Li), detectors could be utilized to collect data. High resolution gamma-ray spectra were therefore collected as a function of cooling time from KE Reactor spent fuels in addition to the sodium iodide spectra presented. The results from four of the Ge(Li) spectra have been summarized and are presented with reference to age determination measurements.

The four spectra represent gamma-ray data from one KE Reactor fuel piece as a function of cooling time. Thirty minute spectra were collected. The spectra are shown in Figure A-1 and the distinguishing feature of the Ge(Li) spectra is the large number of photopeaks evident as compared to the sodium iodide spectra. The photopeaks listed in Table 1 of the main text are identified. The greater detail of the Ge(Li) spectra provides additional decay characteristics that can be followed to give an age index. An obvious visual age index for the cooling times listed in Figure A-1 is the relative heights of the photopeaks at 487 keV and 496 keV, ^{140}Ba - ^{140}La and ^{103}Ru , respectively, since the relative heights vary markedly from spectrum to spectrum.

The areas of certain of the photopeaks seen in Figure A-1 were determined through least squares analysis with a computer code GSSLRN-II.⁽¹⁾ The areas for photopeaks from ^{140}Ba - ^{140}La , ^{95}Nb and ^{103}Ru are summarized in Table A-1. Ten photopeaks from ^{140}Ba - ^{140}La are listed as an example of the greater detail of Ge(Li) spectra. In order to ascertain how accurate the area values are, they have been used to determine the quantity $\exp(-\lambda t)$ where t in this case is the number of days elapsed from spectrum number 10 to each successive spectrum and which is listed in Table A-1. This was done by dividing

FIGURE A-1. Fission-Product Gamma-Ray Spectra
Collected with a Ge(Li) Detector
as a Function of Cooling Time

TABLE A-1. Photopeak Areas for ^{140}Ba - ^{140}La , ^{95}Zr - ^{95}Nb and ^{103}Ru as well as Ratios of Certain Photopeak Areas

Spectrum Number	Cooling Time (days)	¹⁴⁰ Ba - ¹⁴⁰ La Photopeak Areas										
		304.9 keV	328.6 keV	486.7 keV	537.3 keV	574* keV	815 keV	868 keV	923 keV	1500* keV	1596 keV	
10	15	13,751	78,112	199,582	90,089	46,024	100,725	24,033	10,524	9,044	324,519	
35	22	9,739	52,604	135,876	58,914	28,920	67,091	15,069	--	5,610	213,314	
76	42	4,404	19,366	50,956	20,288	--	23,735	5,019	--	1,908	76,055	
118	87	--	--	4,905	--	--	2,077	357	228	190	6,993	
	t	Decay of Each Photopeak										e ^{-λt}
10	0.00	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
35	6.91	0.708	0.673	0.680	0.654	0.628	0.666	0.627	--	0.620	0.657	0.688
76	26.90	0.320	0.248	0.255	0.225	--	0.236	0.209	--	0.211	0.234	0.238
118	71.95	--	--	0.0246	--	--	0.0206	0.0149	0.0217	0.0210	0.0215	0.0203

Spectrum Number	Cooling Time (days)	⁹⁵ Zr - ⁹⁵ Nb Photopeak Areas			¹⁰³ Ru Photopeak Areas	
		724 keV	756 keV	764 keV	496 keV	
10	15	208,210	255,136	386,144	291,674	
35	22	181,600	232,336	371,761	251,283	
76	42	153,116	191,319	391,837	193,969	
118	87	101,865	132,089	336,001	89,117	
	t	e ^{-λt}			e ^{-λt}	e ^{-λt}
10	0.00	1.000	1.000	1.000	1.000	1.000
35	6.91	0.872	0.911	0.929	0.963	1.050
76	26.90	0.735	0.750	0.751	1.015	1.098
118	71.95	0.489	0.518	0.464	0.870	0.950

Spectrum Number	Cooling Time (days)	Ratio 1596/724		Ratio 1596/756		Ratio 496/724	
		Measured	Calculation Normalized	Measured	Calculation Normalized	Measured	Calculation Normalized
10	15	1.559	1.559	1.272	1.272	1.401	1.401
35	22	1.175	1.161	0.918	0.947	1.384	1.335
76	42	0.497	0.484	0.397	0.395	1.267	1.153
118	87	0.0686	0.0697	0.0529	0.0568	0.875	0.837

the first area of a column (from spectrum Number 10) into the subsequent areas in the same column. These values are compared to the calculated values of $\exp(-\lambda t)$ using published half-lives and the calculated values are also shown in Table A-1. The same has been done for ^{95}Zr - ^{95}Nb and ^{103}Ru photopeak areas. It will be noted that, generally, the decay values for spectrum number 35 are low and it was determined that the resolution of spectrum 35 was poorer for an unknown reason resulting in lower photopeak areas than were expected. Also the ^{95}Nb photopeak area in spectrum 10 appears high, possibly due to a second undetected gamma ray hidden under the main photopeak.

The ratios of photopeak areas useful for an age index were not greatly affected by the poorer resolution of spectrum 35 as can be seen by the ratio values listed at the bottom of Table A-1. The measured values are compared to calculated values where the calculated values have been normalized to the measured values at 15 days cooling time. The ratios are also shown in Figure A-2. Only the 1596 keV photopeak of ^{140}Ba - ^{140}La has been used to form ratios with both the ^{95}Zr photopeaks. The result of ratioing the remaining ^{140}Ba - ^{140}La areas would be curves of the same slope but displaced vertically as are the two curves involving the 1596 keV photopeak in Figure A-2. It should be noted that in the case of the present analysis, the curves do not flatten because the photopeak area determination was done by subtracting the continuum away.

Taking advantage of the higher energy resolution, the 496 keV photopeak of ^{103}Ru was ratioed to the 724 keV photopeak of ^{95}Zr as a possible candidate for age determinations of cooling times greater than 100 days. The half-lives of the two isotopes differ by a factor which is less than 2 (39.8 to 65 days). It would appear that the most satisfactory method of choosing ratios to represent an age index is to select photopeaks whose half-lives differ by a factor of 5 or more as in

FIGURE A-2. Gamma-Ray Activity Ratios as a Function of Cooling Time Which Resulted from Ge(Li) Measurements

the case of ^{140}Ba and ^{95}Zr (12.8 to 65 days, respectively). If $^{144}\text{Ce} - ^{144}\text{Pr}$ were used in place of ^{95}Zr , the resulting curve would be steeper. Longer counting times are needed to get statistically good numbers for $^{144}\text{Ce} - ^{144}\text{Pr}$ which has a half-life of 284 days. However, a calculated curve for the ^{103}Ru to $^{144}\text{Ce} - ^{144}\text{Pr}$ ratio, 496 keV/2186 keV ratio, is shown for illustration. Then, there is $^{106}\text{Ru} - ^{106}\text{Rh}$ whose half-life of 367 days provides an even steeper curve. The fission yield of ^{106}Ru in the case of KE Reactor fuels would be prohibitive but in the case of NPD fuels which attain higher burnups where appreciable plutonium fissions occur, the $^{106}\text{Ru} - ^{106}\text{Rh}$ gamma rays should be more prominent. The next fission product to consider is ^{137}Cs , 30.0 year half-life, as the denominator in any ratio, in order to obtain the steepest curve from which to determine cooling time from measurements.

Ratios which include ^{103}Ru in the numerator should be appropriate age indexes from 100 days to a year. Then ^{95}Zr in the numerator would provide an age index up to approximately 2 years. After that, ^{106}Ru in the numerator would give an age index for cooling times up to 10 years. Thus it is seen that Ge(Li) spectra offer many individual isotopes to use as an age index.

REFERENCE

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APPENDIX B

APPENDIX B

A report entitled "Interim Report on the Test Program of the Spent Fuel Monitor at NPD, Rolphton, Ontario" prepared by John Neri, AECB, was received July 6, 1971. The data contained in the report has prompted the writing of Appendix B in order to discuss certain results of importance to the program. The following information was contained in the report:

- a) Photographs of gamma ray spectra from assembly 6808 as a function of cooling time up to 68 days from date of discharge.
- b) Photographs of gamma ray spectra from the five other assemblies indicated in Table 3 of the main text and assembly LB-084 which has also been included in the controlled measurement program. The spectra were obtained 69 to 74 days from date of discharge of the assemblies.
- c) Plots of the activity ratios for six of the assemblies as a function of time.
- d) Irradiation histories for each assembly which included core positions and residence time in each position, estimates of fast flux and slow flux in all positions and a graph of the flux history. The total burnup in MWd/tonne U for each assembly, see Table 3, was also given in the report.

An observation made after looking at the photographs of the gamma-ray spectra was that it is necessary to redefine the meaning of age and cooling time in the case of NPD spent fuels. The $^{140}\text{Ba} - ^{140}\text{La}$ photopeak at 1600 keV had completely disappeared from the spectra of assemblies GC002W, LB084 and DB35, indicating that these assemblies had been out of a neutron flux 150 days or longer. As it turned out, GC002W had

been out 146 days, LB084 176 days and DB35 195 days as listed in Table B-1. These "total cooling" times were determined using the dates on which the assemblies were charged into a zero flux position. Thus, age and cooling time in the case of NPD fuels should include the time an assembly has resided in a zero flux position as well as the time from assembly discharge from the reactor.

The data given for the various activity ratios in the interim report were plotted keeping in mind the redefinition of cooling time, see Figure B-1. Each individual plotting symbol used represents data from a given assembly as listed in Table B-1. The redefinition extends the cooling time to nearly 200 days and curves have been drawn in by inspection to connect the data points. A second observation was that the shape of the curves shown actually depicts reality. For example, what one is observing by the data in the case of the Plateau 2 to Plateau 1 ratio, #4/#1, is the differences in the decay rates of the 1072 - 1233 keV Compton, as monitored by single-channel analyzer #4, and the 214 - 429 keV Compton, as monitored by single-channel analyzer #1. The higher energy Compton, #4, decays with a half-life nearly that of 12.8 days (from ^{140}Ba - ^{140}La) until the ^{140}Ba - ^{140}La has decayed through approximately eight half-lives and then the decay rate changes gradually during the next three half-lives until it decays at a rate more nearly that of 284 days from ^{144}Ce - ^{144}Pr because the ^{140}Ba - ^{140}La completely disappears. The lower energy Compton, #1, although affected by ^{140}Ba - ^{140}La , decays at a rate more nearly equal to a half-life of 65 days (^{95}Zr) and will continue to do so for approximately two years. Therefore the curve representing the #4/#1 ratio turns around and begins to increase after 100 days cooling time as seen in Figure B-1 because, after 100 days, the larger half-life is now in the numerator and the denominator is decreasing faster than the

B-4

FIGURE B-1. Ratio Values as a Function of Redefined Cooling Time

numerator. Thus, it is appropriate to change this ratio to #1/#4 after 100 days. It is also appropriate to increase the energy region monitored by single-channel analyzer #4 in order to obtain good counting statistics in a reasonable time, 1072 keV to 1600 keV. The ratios formed from the readings of single-channel analyzers #1 and #4 are thus fairly sensitive as an age standard over a period of approximately one and a half years except during the transition period after 100 days.

Changes to the effective half-life determining the decay rate of the $^{140}\text{Ba} - ^{140}\text{La} + ^{103}\text{Ru}$ peak, single-channel analyzer #2, also explains why the curve representing the #2 to #3 ratio turns around and begins to increase after 100 days where the $^{95}\text{Zr} - ^{95}\text{Nb}$ is represented by single-channel analyzer #3. In this case, the $^{140}\text{Ba} - ^{140}\text{La} + ^{103}\text{Ru}$ peak is super-imposed on a ^{95}Zr Compton (half-life 65 days) while the peak itself is decaying with an effective half-life dependent on the 12.8 day half-life of $^{140}\text{Ba} - ^{140}\text{La}$ at 487 keV and the 39.8 day half-life of ^{103}Ru at 496 keV. As the cooling time passes 100 days, $^{106}\text{Ru} - ^{106}\text{Rh}$ at 512 keV grows in with an accompanying half-life of 368 days and the effective half-life gradually changes from one which is less than 65 days to one which is much greater than the 65 day half-life of the denominator, $^{95}\text{Zr} - ^{95}\text{Nb}$. It is also appropriate to invert this ratio to a #3/#2 ratio after 130 days. The ratios involving #2 and #3 do not provide as sensitive an age standard as the ratios involving #1 and #4 do.

The #5/#3 ratio is shown plotted in Figure B-2 and the ratio is very sensitive to age up to 100 days cooling time. After 100 days, single-channel analyzer #5 which monitors the $^{140}\text{Ba} - ^{140}\text{La}$ peak at 1600 keV should be changed to monitor the 2186 keV peak of $^{144}\text{Ce} - ^{144}\text{Pr}$ and the ratio inverted to a #3/#5 ratio (the longer half-life data in the denominator).

FIGURE B-2. Ratio Values of the #5/#3 Ratio for Cooling Times Less than 100 Days

The #5 single-channel analyzer should also be set to include a large energy region, say 1600 keV to 2300 keV in order to obtain good counting statistics in a reasonable time.

As can be seen from Figures B-1 and B-2 the scatter in the data is a cause for concern since the resulting curves are not as definitive as desired for accurate age measurements. The initial data from spectrum 1 through spectrum 9 (12 and 13 days cooling time) were questionable since a recalibration was done after spectrum 9. The accuracy of a number of the initial data points are in doubt because the calibration may not have been correct for at least some of the measurements for spectra 1 through 9. Factors which may have caused scatter in the later data were apparently present due to the fact that the stabilizer began to malfunction after a few weeks operation. It was disconnected from the system and care has since been taken to assure that electronic drift is kept at a minimum.

Other factors⁽¹⁾ that may have had an influence on the scatter include the following:

- a) Several of the fuel bundles involved have come apart during the course of the experiment due to the fracture of the bundle end plates. It would therefore seem likely that the motion of the can as it is transferred between the measurement tray and the storage basket resulted in a rearrangement of fuel pencils and constant geometry between detector and source has not been maintained with the possibility that extraneous water has been introduced in the beam path.
- b) Additional decay in the stabilization of certain of the electronic equipment has been unresolved. For example, single-channel analyzers #1 and #2 have not exhibited as

sharp a threshold cut off as the other single-channel analyzers. Checking is currently underway to determine what scatter this unstable situation has introduced.

- c) Background activity due to some collection of crud activity at the collimator head despite the air bubbling technique. Readings are also being taken to determine how the background may vary over a period of time.

In addition to the scatter, apparently there are biases in the data sets, see Figure B-2. The data from BC029W plots lower everywhere and a possible correlation may be in the residence time of the last flux position where the last flux position differs by a factor of 2 (smaller) from the flux in the previous position, see Table B-1. If the residence time is less than 200 days, the saturation level of ^{95}Zr in the last position may be affected and be high compared to the saturation level of $^{140}\text{Ba} - ^{140}\text{La}$. At least this is consistent with what is seen in Figure 2 for assemblies 60036 and BC029W although the scatter in the data does complicate reaching a definite conclusion.

A recommended basis of proceeding in order to define an age measurement standard with less scatter is to first find the cause of the scatter in the data. Once the cause is understood or eliminated then it would be appropriate to obtain newly discharged assemblies selected according to residence time in their last position and verify the present curves. The curves should include data measurements as a function of total cooling time of 200-300 days, 100 days for the time after discharge and 100 to 200 days for possible residence time in a zero flux position. The curves would thus include a redefinition of cooling time as well as the inversion of all the ratios (based on different settings of certain of the single-channel analyzers once the total cooling time exceeds 100 days).

It should be pointed out that even with the scatter evident from the data, the curves drawn by inspection in Figure 2 for KE Reactor fuel and in Figure B-2 for NPD Reactor fuels in the case of the $^{140}\text{Ba} - ^{140}\text{La}$ to $^{95}\text{Zr} - ^{95}\text{Nb}$ ratio, #5/#3, are nearly identical between 10 and 90 days cooling time. Thus by identifying the cause for the scatter and understanding mechanisms for possible biases, a reliable standard for NPD fuels is expected.

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