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IRRADIATED FUEL AGE DETERMINATION STUDY

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By

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Facilities Engineering Section
IRRADIATION PROCESSING DEPARTMENT

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IRRADIATED FUEL AGE DETERMINATION STUDY

INTRODUCTION

From time to time a quantity of incompletely aged irradiated fuel has been sent by mistake from the reactors to the processing areas. The release of uncontrollable quantities of harmful fission products such as I_{131} upon processing this fuel creates a need for a method to prevent its shipment to the processing areas. At present this is done by procedural techniques using cards, filing techniques, and cross checks. These methods are entirely independent of the characteristics of the fuel itself and are subject to failure from human error. The occasional shipments of fresh fuel attest to the weakness of the procedural method. It is therefore advantageous to find a method of determining the age of irradiated fuel after discharge from the properties of the fuel. Such a method, with its relative immunity to human error, would be very useful in checking shipments as well as for such jobs as determining the correct age of fuel being stored for accumulation of desirable fission product daughters. It was the purpose of this effort to make a study of possible methods of direct fuel age determination, formulate and perform a test of the most promising method, and analyze the results in terms of practicability here at Hanford.

SUMMARY AND CONCLUSIONS

The results of this test indicate that the age of irradiated fuel after discharge may be accurately determined by the ratio of two prominent gamma emission peak amplitudes. The decay of La^{140} and $Zr^{95}-Nb^{95}$ produces gamma emissions at 1.58 Mev and .76 Mev respectively whose amplitude ratio as seen by the equipment used in this test is a very strong function of time and is only slightly effected by irradiation power level and total exposure. At a fuel age of approximately 120 days, the nominal value of the ratio indicates a fuel age within ± 6 days with 95% confidence, showing the commendable accuracy of the method.

The cost of a prototype of this device is approximately \$10,000, installed in a reactor fuel storage area, with a fair chance of reduction if more than one unit is built. Although fewer units would be required if they were installed at the separations areas rather than at the Reactors, the danger of melt down during shipment indicates that reactor area installation would be more desirable.

DISCUSSION

The measurement of gross gamma activity to determine the cooling time of the fuel discharge does not appear to be a suitable solution. The gross gamma activity of irradiated fuel decreases by a factor of about 10 during the period from 10 to 120 days after discharge. The position of the fuel in the reactors may cause the initial activity of the fuel to vary by as much as 5:1. This could cause a bucket of fringe zone irradiated fuel to appear 120 days old when its actual age was 65 days. This high error is obviously intolerable. The heating rate of water surrounding the fuel depends on the wattage output of the radioactivity of the fuel. The heating rate has been found to decrease at a rate approximately equal to that of the gross gamma activity and therefore would be subject to the same uncertainty in the age of the fuel being monitored. This last method could be used at Hanford as a check on the fuel after it had been loaded into the tank

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cars. An excessive heating of the tank car water would indicate a shipment of fuel that was unquestionably too green. The lack of a high heating rate would not necessarily indicate adequate fuel aging time however, and the heating rate itself would not be a good measure of the age of the fuel.

The apparent decay rate of the gross gamma activity may be increased by shielding the detector. The shielding filters out the low energy gamma radiation relative to the high energy gamma, leaving a flux at the detector which is weighted in favor of the high energy gamma. Since high energy gamma is predominantly a feature of short half life isotopes, the shielded gross gamma activity will decrease at a faster rate than the unshielded gamma. Behind two inches of lead shielding at a distance of 17 feet the gamma activity was found to decrease by a factor of about 70 during the period from 6 to 124 days after discharge. The variation of initial activity of individual elements was found to be as high as 24:1 with the same shielding conditions. Therefore, the accuracy of this method is not significantly better than that of the unshielded gamma technique.

The methods that appeared to show the most promise were those involving the monitoring of one or two specific gamma emission energies. It was thought that perhaps a prominent gamma could be found that would decay at such a rate that the variation in initial activity of the fuel would be insignificant when compared with the decay rate and could be neglected. If this did not turn out to be the case, then it was felt that perhaps two prominent gamma energies could be found from isotopes of significantly different half lives that the ratio of the amplitudes of the two peaks would be an indication of the age of the fuel, relatively independent of its initial activity. It was with this in mind that a test was formulated to monitor the gamma decay of six randomly selected buckets of fuel and six individual irradiated fuel elements chosen to represent the anticipated extremes in initial activity.

The apparatus used is shown schematically in Figure 1. The stream of gamma photons was brought to the surface of the storage basin through an air column, two inches of lead shielding, and eight inches of $\frac{1}{4}$ " diameter collimation. It then impinged on a sodium iodide scintillation crystal and was counted by a photomultiplier tube. This signal was sent to a 256 channel analyzer which read out the spectra on an oscilloscope or punch tape for analysis. The collimator was portable and could be moved from point to point for accurate positioning on the fuel samples. Readings were taken in two positions on each bucket giving a total of twelve readings from the six randomly selected buckets of fuel.

The six elements chosen to represent the extremes in activity were from each of three positions in a fringe zone and a central zone process tube. The most inactive front element, the most inactive rear element, and the most active element from each of these two tubes were chosen. These six elements were monitored for comparison to the data from the randomly selected fuel in the buckets.

The data resulting from this test are all in the form of gamma spectra. Figure 2 shows the series of gamma spectra taken in a particular position on one of the six randomly selected buckets of fuel over a period of 153 days after discharge from the reactor. The ordinate of the figure is the counts received in a four minute

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live count in each of the 256 energy channels which constitute the abscissa. The highest curve was taken 15 days after discharge and the lowest 153 days after discharge. As can be seen from Figure 2 there is a greater amplitude changeover time in the prominent energy peaks than in the amplitude of gross activity.

Figure 3 shows the two most widely varying spectra from the six individual elements selected. The highest is from the hottest central zone element and the bottom is from the extreme rear element from the fringe zone tube. These were both taken six days after discharge of the fuel.

Only the lowest two energy peaks on the spectra appear to be readily discernable over the full 150 day period (see Figure 2). In order to identify them and to examine their characteristics, their amplitudes were plotted as a function of time in Figure 4 and 5. From Figure 2 the lowest energy peak is at .76 Mev and the next higher energy peak is at 1.58 Mev. The high yield of the Zr^{95} isotope in the fission product makeup, the abundance of the .76 Mev gamma emissions in the Zr^{95} - Nb^{95} - Mo^{95} decay scheme, and the proximity of the apparent half life of this peak to that of 65 day Zr^{95} lead to the conclusion that this peak is from the Zr^{95} decay system. Although the half life cannot be determined accurately by the plot of peak amplitude vs. time as shown in Figure 4, it is close enough to 65 day Zr^{95} to be substantiating, and there are no other high abundance fission product isotopes with a gamma emission and a half life in this range.

The 1.58 Mev peak is from La^{140} , the daughter of the very abundant Ba^{140} . Although the La^{140} has a half life of 40 hours, it can disintegrate no faster than it is produced by the 12.8 day Ba^{140} . This peak follows the 12.8 day half life line very closely as can be seen in Figure 5. The departure of the data from the 12.8 day half life line at the high fuel ages can be explained by the fact that the peak is sinking into the lower intensity background gamma. Figure 2 shows that as the fuel ages, the 1.58 Mev peak becomes less distinct from the background. Since this background is from longer half lived isotopes, the rate of descent of the peak decreases at high fuel ages. A similar effect is seen in Figure 4 at low fuel ages where the .76 Mev peak descends more rapidly than the 65 day half life due to the high background of short half lived isotopes (see Figure 2). As the background recedes, the .76 Mev peak is better defined, and it settles down to the 65 day decay rate that would be expected.

A careful examination of Figures 4 and 5 will reveal the fact that the readings taken at fuel ages of 49, 63, and 76 days are lower than would be expected. It can be shown that the percentage error between the 12.8 day half life line and these data points for the 1.58 Mev peak is very nearly the same as the error between the 65 day half life line and these points for the .76 Mev peak. This leads to the conclusion that this is a function of machine error rather than a part of the fuel decay behavior. To illustrate this, Figure 6 shows the decay curve for the .76 Mev peak after having been "corrected" so that the 1.58 Mev peak decay curve coincides with the 12.8 half life line in its central region (49 to 76 days).

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Figures 7 and 8 show the data taken from the six individual elements superimposed on the data taken from the buckets of fuel shown in Figures 4 and 5. These data were taken on the same days as the data on the buckets were taken but the discharge date of the individual elements is one month later than that of the buckets. Therefore, the first point shown for the individual elements was taken on the same day as the third reading for the buckets. The important point is that although the individual element curves show the same lowering as the bucket data curves, it is a function of the day the data were taken and not of the time after discharge. The element and bucket data taken on April 14 and 27 and May 11, 1964 are lower than would be expected, but the two sets of elements had been discharged on different days. This proves that the lowering of these three data points was caused by a machine failure and not by an irregularity in the decay process.

Figures 7 and 8 also show the wide variation possible in initial activity of the fuel and how this continues through the decay process. In the case of the 1.58 Mev peak, this variation can be as great as 30:1 when the extreme elements are considered. The same circumstances applied to the .76 Mev peak yield an initial variation as great as 10:1. The better of the two peaks to be monitored as an indication of fuel age is that of the 1.58 Mev gamma which varies by a factor of 470 from discharge to 120 days after discharge. The .76 Mev gamma varies by a factor of only about 10:1 over the same period. Using the 1.58 Mev peak alone and the data from the randomly selected buckets of fuel, a possible error of 40 days at a fuel age of 120 could be realized. That is to say, in the case of the 1.58 Mev peak at 120 days cooling where the permissible peak amplitude may vary from 68 to 280 days old (Figure 5) the fuel could be as little as 95 days old and as much as 162 days old. If the deliberately selected extremes represented by the individual elements are included, the error is many times worse as can be seen by inspecting Figure 8.

In order to minimize the effect of initial activity, the ratios of the .76 Mev and 1.58 Mev peaks were taken at the various cooling times for the data taken from the randomly selected buckets of fuel and plotted in Figure 9. A straight line approximation of the averages of the data sets may be drawn and follows the line described by $r = .18 e^{-.0385t}$, where r = the ratio of the amplitude of the .76 Mev peak to that of the 1.58 Mev peak, and t = the time after discharge of the fuel. The departures of the actual average curve from the straight line approximation at 30 to 130 can be explained by the burying of one or the other of the peaks in background gamma of different half lives. This is the same reason as that for the curved ends of the peak magnitude curves as discussed above.

As can be seen by comparing Figure 9 with Figures 4 and 5, the deviation from the mean of the ratio curve is very much less than for either of the peak amplitude curves. As a result of this, the peak amplitude ratio is the best measure of the age of the fuel. The effects of the machine failure, noticeable in the peak amplitude curves, cannot be distinguished in the ratio curve. The accuracy of the bucket data at 120 days indicates that a bucket of fuel yielding a ratio within the range of those expected at 120 days could have a maximum age of 131 days and a minimum of 109 days.

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The superposition of the extreme data gathered from the individual chosen elements are shown on Figure 10. An inspection of Figure 10 will reveal that for the most part the deviations of these data from the random bucket data are slight with the exception of the coldest rear element from the fringe zone process tube. The average of the data other than those taken of this element fall within the deviation found for the random bucket data. Therefore, excluding this one element, the accuracy is the same as that given for the bucket data above, but if the coldest rear fringe tube element is included, the minimum age that can be mistaken for 120 days is 74 days. This is still far better than the accuracy obtainable by the 1.58 Mev peak alone.

The question arose as to the significance of the comparatively great disparity of the fringe tube rear element data from the rest of the data taken in this study. The behavior of this element is as though it were 30 days older than its discharge date would indicate. Barring any mistakes in selecting the fuel for the test, the most likely explanation is that there was a "cold spot" in that region of the reactor for a period before the element was discharged. This would allow the fission products to decay while the fuel was still in the reactor. Due to the random nature in which buckets of irradiated fuel are loaded it is extremely unlikely that an entire bucket would be charged with fuel from such a "cold spot". If this did happen, the ratio would indicate that the bucket was ready for processing, whereas the discharge date would indicate that it was not sufficiently aged. Calculations indicate that such fuel could be processed safely.

The small diameter of the air column (Figure 1) caused the spectra to be dependent upon the element on which the column rested. To determine the error introduced by placing the collimator on a "cold" element in a "hot" bucket of fuel, the following test was run. A normal element from a 120 day old bucket of fuel was placed in the two positions in the top layer of an 84 day old bucket of fuel and the collimator placed on it for readings. The resulting ratio indicated that the bucket was between 95 and 106 days old. The ratio taken on the 84 day old bucket before the cold element was inserted indicated an age of between 79 and 92 days. The cold element itself indicated an age of between 115 and 125 days. Thus it can be seen that a reading taken directly over a very cold element in a hot bucket will produce an error in the ratio indicating a greater age than actually is the case, but the error is not intolerably large. The possibility of this occurring is very slight due to the great rarity and small size of "cold spots" in the reactor. The error may be reduced by increasing the diameter of the air column and the probability of its occurring halved by taking two readings per bucket.

In general, it may be said from the data collected in this study that fuel from lower power (fringe) tubes will yield higher ratios than higher power (central) tubes. The central zone tube elements yielded ratios very close to the average found from the random bucket data, which would be expected due to the high preponderance of central over fringe zone tubes.

Enriched fuel was not tested in this program, but there is no reason to believe that the same methods would not work as well to determine its age after discharge.

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Prototype Design

The electronic apparatus used in these tests is very much more expensive and complicated than that which would be required for a prototype. The collimator, shielding, and detector would remain unchanged except for the addition of a source to be used in calibrating the counter to compensate for photomultiplier tube drift. The first prototype could use the existing detector shielding, signal attenuator, and air column assembly, modified slightly to install a calibration source. The cost of these items is \$1500.

The electronics of the monitor can be assembled using, primarily, commercially available modules and using components which have been developed by Nucleonic Instrumentation, HL for other instrument systems. The monitor will consist of a single scintillation detector (NaI (tl) with the output of the detector providing both the pulse information for age determination and dc current information to monitor for possible overloading of the detector by a fresh bucket with extremely high activity. This overload activity will initiate visual and aural alarms. The pulse output from the detector is amplified by a linear pulse amplifier and then divided into three channels of pulse height information. One of the channels will be used to monitor the calibration source for the calibration of the monitor prior to using it. The "window" of this channel will be centered on the photopeak energy of the calibration source. The readout will be on a count rate meter mounted inside a locked cabinet.

Of the other two channels, one will have its window set to monitor the 0.76 Mev photopeak from Zr^{95} - Nb^{95} and the other channel will monitor the 1.58 Mev photopeak of La^{140} . The output of the La^{140} channel will drive a 6-decade preset count scaler and the output of the Zr^{95} - Nb^{95} channel will drive a 6-decade scaler. The ratio of the Zr^{95} - Nb^{95} to the La^{140} is determined by making the preset count scaler stop both scalers when the preset count is reached. The output of the decade scaler can be decoded into analog information and displayed on a panel meter marked in days or weeks of fuel age, as desired. The meter can have a red line section, and a separate alarm (visual and/or aural) can be actuated. This will allow the high voltage supply, the amplifiers, the single channel pulse height analyzers, and the scalers to be mounted inside a locked cabinet.

During calibration, the calibration source will be moved into position and a visual alarm on both the front panel and the interior instrument panel will indicate the source position. A circuit can also be activated which will remove the fuel age indication from the front panel meter as a further precaution during calibration. This could be done by an instrument technician once every two weeks or so depending on the drift of the photomultiplier tube. As presently conceived, the bucket readings could be taken by an operator who would position the detector, activate the counter, and await a go or no go signal from the visual or aural alarm. This would take three or four minutes per reading.

A line filter and isolation transformer, with two voltage taps, will be included to alleviate line noise problems and to provide some adjustment for high or low line voltage. A line voltage regulator could be used except that those acceptable for pulse counting systems are generally quite expensive.

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The estimated cost for the first prototype is as follows:

Commercially Available Electronics Components	-	\$6340
Onsite Electronics Fabrication and Services	-	1560
Source Installation in Detector Shielding	-	<u>200</u>
Subtotal	-	\$8100
10% Contingency	-	<u>800</u>
Total Cost, First Prototype	-	\$8900

If more than one system is to be built, the estimated electronics cost per system would be lower due to cost reductions in purchasing several items at one time, and the elimination of the one system engineering cost such as general system design, preparing specifications and bid review, and circuit development. The fabrication costs of the developed circuits would have to be included. Any prototype other than the first will require a new detector shielding and collimator assembly which will add \$1300 to the above costs, making the estimated total cost \$10,200 per unit.

It may be possible to find a vendor who would bid on the completed assemblies in which case the costs would probably be less. It would be necessary to prepare a detailed specification describing the complete system requirements.

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FIGURE 1
SCHEMATIC REPRESENTATION
OF APPARATUS

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FIGURE 4
.76 Mev PEAK AMPLITUDE
VS
TIME AFTER DISCHARGE
(BUCKET DATA ONLY)

MAX
 ○ AVE (12 POINTS)
 MIN

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FIGURE 6
.76 Mcv. PEAK AMPLITUDE
VS.
TIME AFTER DISCHARGE
(BUCKET DATA ONLY)
READINGS 4,5,6 CORRECTED

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FIGURE 7
.76 Mev PEAK AMPLITUDE
VS.
TIME AFTER DISCHARGE

KEY (XY)

X=TUBE

Y=POSITION

F=FRINGE

F=FRONT

C=CENTRAL

C=CENTER

R=REAR

DATE

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