

SEGREGATION OF LOW-LEVEL DRY ACTIVE WASTE

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ABSTRACT

A program has been carried out to characterize the Dry Active Waste (DAW) stream from a typical PWR power plant in order to determine the usefulness of large-volume DAW monitors for segregating such waste in order to dispose of it in appropriate facilities.

A waste monitor using plastic scintillation counters was used for measuring the waste. The monitor had a volume of about 300 liters and an overall efficiency of about 12% for a typical fission product mixture. It provides automatic compensation for background radioactivity and can measure a bag of waste in less than a minute, including background measurements.

Six hundred consecutively generated bags of DAW were measured. These had a total activity of about one millicurie and an average specific activity of about 540 nanocuries per kilogram. About half of the bags contained less than 1000 nanocuries and had specific activities of less than 100 nanocuries per kilogram.

Based on simplified preliminary calculations, it appears that an evaluation of the risks of disposal of bags such as these in a landfill other than a low-level waste disposal facility could be carried out that would demonstrate that such disposal of half or more of these bags would not result in any substantial hazard, either short or long term.

INTRODUCTION

This paper presents the major findings of an Electric Power Research Institute (EPRI) study RP1557-9. The study was directed at evaluating Dry Active Waste (DAW) monitoring techniques for effective segregation of uncontaminated wastes or wastes that are only slightly contaminated. Four objectives were defined for the project:

1. To characterize the distribution of the measured activity (nCi/kg) in DAW from an operating commercial nuclear plant;
2. To establish the reliability of large-volume monitoring, compared to hand methods;
3. To demonstrate calibration techniques for large-volume DAW monitors; and
4. To quantify the effectiveness of segregation procedures to reduce the volume of DAW at an operating nuclear power plant.

The first objective has been achieved as it relates to a particular plant during normal power operation. The reliability of large-volume monitoring has been established, although it is not yet possible to devise a suitable index for comparing its reliability to that of hand methods. A calibration technique has been developed and demonstrated. The final objective has been achieved to the extent that all of the necessary data are in hand to quantify the effectiveness of segregation procedures at this plant, once an appropriate set of segregation criteria has been selected. A set of criteria must be developed as a separate project, based on calculated health effects and legal requirements. To become of practical value, such a set of criteria would have to be acceptable to and accepted by the Nuclear Regulatory Commission and, in some cases, appropriate State or local authorities. Given such criteria and the data developed in this project, the effects of segregation can easily be quantified.

The instrument used for large-volume monitoring

was the WCM-10 Waste Curie Monitor manufactured by National Nuclear Corporation. It was set up in the Auxiliary Building of the Sequoyah Nuclear Plant of the Tennessee Valley Authority. A group of bags of contaminated Dry Active Waste collected during April and the first half of May, 1983 constituted the sample tested.

Each bag was measured with a hand-held detector in accordance with the normal procedure and then measured in the WCM-10. Most bags were then opened on a frisking table and the contents hand-frisked. If individual items having high activity were identified, these were removed and the bags remeasured. This was effective in only a few cases, because of the relatively uniform contamination of the material in most bags. The results from the six hundred sample bags were analyzed in three sequential groups of 200 bags each to give a measure of consistency. A record was kept of which bags went into each of about a dozen "test drums" and the total measured activity put into each drum compared to the estimated activity measured in the manner normally used to obtain shipping information for shipment to the waste disposal site.

The remaining sections of the paper will discuss the background of the project, the equipment used, the calibration method developed, the data collected, and the conclusions reached.

BACKGROUND

Disposal of low-level radioactive waste is becoming increasingly burdensome to nuclear power plant operators. The number of active burial sites has been reduced to two and the operation of new sites is still fairly far in the future. Both shipping and disposal costs are increasing at an unprecedented rate. These impacts have been compounded by additional restrictions on shipping and disposal, both by the States and the NRC, as well as by the disposal site operators. These factors all impose additional costs and workloads on the power plant operators.

In addition to the burdens identified above, it has long been recognized that the space available at disposal sites is limited and should be used for material requiring application of the conditions currently imposed on disposal of radioactive waste and not for disposal of material that could safely be disposed of more simply. The NRC reinforced this point in October, 1981, in its policy statement on low-level waste volume reduction.

The DAW stream in a nuclear power plant contains a large volume of low level waste. The majority of this material is of extremely low level, but it frequently is mixed with some material of somewhat greater activity. Some plants attempt to provide a system for the separate collection of noncontaminated material from controlled areas, but this has seldom been very successful and the material usually ends up commingled with the contaminated material. In addition, where the segregation has been carried out, there is often reluctance, for public relations reasons, to release the material locally. Even if this is not a problem, licensees properly will not release the material without a check to assure that it is in fact uncontaminated. Because of the increasingly sensitive measuring equipment becoming available, this, in turn, leads into questions regarding the level at which one considers material to be non-contaminated and how that level is to be measured.

In view of the above considerations, in early 1982 National Nuclear Corporation (NNC) proposed to the Electric Power Research Institute (EPRI) a program to test its Waste Curie Monitor, Model WCM-10, in a realistic plant environment to achieve the objectives defined in the Introduction. It was NNC's belief that the proposed study, together with a subsequent safety analysis based on the data obtained, could be used to prepare a license amendment request to the NRC to justify disposal of a large fraction of this relatively innocuous waste stream either in a utility-owned land-fill within the site boundary or in a local public land-fill.

In June, 1982, EPRI contracted with NNC to carry out the program. The Tennessee Valley Authority (TVA) was contacted by NNC and agreed to support the program by providing the use of TVA facilities and the manpower necessary to carry out the on-site activities. The work was originally planned for the Browns Ferry site, but operational considerations later required a move to Sequoyah. Various plant contingencies delayed the start of full scale operations until the beginning of April, although calibration work was performed in the meanwhile. During a six week period ending May 13, 1983, 600 bags of waste were generated and measured.

EQUIPMENT DESCRIPTION

The monitor used for measuring the radioactive material content of the bags is the National Nuclear Corporation Model WCM-10 Waste Curie Monitor. This is a large-volume high-sensitivity monitor with a counting volume measuring about 24" wide by 27" high by 29" deep. It uses six 25" by 19" by 1-1/2" plastic scintillators (including one in the door) as detectors. The detectors and counting volume are surrounded by a 1-1/2" lead shield to reduce background. The scintillations are detected by photomultipliers, amplified, and counted. The number of counts accumulated during the counting interval (normally 10 seconds with an option of 100 seconds) is divided by the setting of a calibration control and each count out of the driver gives a single count on the digital read-out. Proper selection of the calibration control setting makes the digital read-out direct reading in nanocuries. An

alarm is provided that can be set at any desired nanocurie level to light a red or green signal at the end of each measurement, providing guidance to unskilled operators.

A background count is taken before each sample count is made. The circuitry automatically subtracts this count from the subsequent sample count. The difference, which appears on the digital read-out, represents the true radioactivity of the sample with continuous updating of a changing background. This method of operation permits use of the monitor in areas having background radiation levels typical of those to be found where DAW measurements are likely to take place. Care must be taken, however, to avoid making measurements when high-activity material (e.g., drums containing curie quantities of waste) are being moved in the immediate vicinity of the monitor. The background level will, however, affect the sensitivity and accuracy of the monitor.

The monitor is quite sensitive and has an overall efficiency of about 12% for a typical fission product mixture. It is not, however, very energy dependent and cannot be used for isotopic discrimination.

CALIBRATION

The first topic we consider as part of the calibration procedure is the measurement of the radiation background and analysis of its impact. Following that we will discuss the preparation and measurement of calibration samples, and, finally, the selection of an appropriate calibration setting for the monitor.

Background Measurements

As mentioned above, background measurements are made before each sample measurement. Two aspects of the background are of interest. First is the amount of background and its effect on accuracy and sensitivity, and the second is changes between the times of the background and sample measurements.

The amount of background radiation penetrating the shielding and striking the detectors basically determines the minimum amount of activity that can be measured. A measurement sequence consists of first measuring the background by going through a counting cycle with the monitor empty and then putting in the sample and measuring sample-plus-background counts. The background number is then subtracted from the sample-plus-background. The uncertainty in this can be shown to be proportional to the square root of the sample counts plus twice the background counts. The proportionality factor turns out to be one over the square root of the calibration factor (the calibration factor is the number of counts detected in ten seconds for a source of one nanocurie or, for single gamma disintegrations, 370 times the efficiency).

Several series of measurements, totalling a few thousand seconds, were carried out over two or three days. These showed that the levels in the location selected for the monitor were the equivalent of about 100-120 nanocuries. From this we can calculate that the uncertainty is about two nanocuries if we are measuring a very small sample, about 3% of the measured value for a sample of 100 or so nanocuries, one-half percent for a one microcurie sample and less for larger samples.

The other factor of interest in background measurement is temporal changes. The first of the background measurement series used 100-second counts and was spread over several hours. The uncertainty calculated from these measurements was about 2-1/2 nano-

curies, whereas the uncertainty calculated on a statistical basis was about 1/2 nanocurie. This indicates that over this period of time there were background changes equivalent to several nanocuries. Since it was apparent by that time that 10-second counts would be used for the final measurements, the remaining background series were run with that interval. Analyses of these measurements showed that during time periods of the order of five minutes there were no discernible changes in background, except when identifiable activities, such as the movement of nearby waste drums, were taking place. On this basis, it was concluded that the monitor's background compensation capability was acceptable and that the measurement accuracy, as it relates to statistical and background effects, was 4 nanocuries for small samples, about 5 nanocuries for a one microcuries sample and increased as the square root of the sample size for larger samples.

Calibration Sample Preparation

In order to obtain accurate measurements, the WCM-10 should be calibrated with samples having isotopic distributions similar to the material to be measured after calibration. Although the instrument is not very energy-dependent, there is a slight dependence because at very low energies (e.g., under 100 kev) gamma rays are attenuated by the 1/16th-inch stainless steel liner and at relatively high energies fewer of the gammas lose their energy in the plastic detectors. Since most of the isotopes of interest have gammas between about 0.2 and 1.5 mev, these variations are not very significant. More important is the fact that some isotopes have two gammas per disintegration. The only one of these of interest is Cobalt-60, but this is a very important isotope as far as the makeup of DAW is concerned.

At Sequoyah it was known from previous experience that the isotopic distribution of the DAW stream (and most of the other contamination in the plant) is very similar to that of the water entering the clean-up system. This is basically reactor primary system water that has decayed a few weeks. This was selected as a suitable source of materials for preparing samples.

The first samples measured were one-liter samples prepared by using full strength or diluted water taken from the clean-up system before the demineralizers. Isotopic distributions and specific activities were measured in the laboratory. The samples were then measured in the WCM-10 and new calibration control settings calculated. Some samples taken after demineralization were also measured. Although the isotopic distributions were somewhat different from the before-demineralizer samples, as would be expected, the calculated calibration factors were essentially the same. This would be expected since the change in the abundance of Cobalt-60 relatively small. The absolute value of the calibration factor, however, was significantly lower than expected. Since it was known from the laboratory analysis that the samples contained significant amounts of Xenon-133 (a very low energy gamma emitter), it was concluded that, although neither the WCM-10 or the laboratory detector saw much of it (because of self-absorption in the sample), the algorithm used in the laboratory for abundance calculations made a correction for this, whereas the WCM-10 did not. Subsequent samples were more thoroughly de-aerated to eliminate part of this problem, but it was also decided that a measuring geometry with less self-absorption should be used.

It was decided that the most desirable configuration for the calibration sample would be a bag of rags, geometrically similar to the bags of trash,

with the liquid sample dispersed in the rags. A new set of samples was prepared in this way. Before preparing the bags, however, an 800-ml sample was measured in a Marinelli beaker, both in the laboratory and in the WCM-10. As expected, the calibration factor calculated from the Marinelli beaker measurements was higher than that calculated from the 1-liter bottle measurements and the factor calculated from the measurements of the dispersed samples was higher yet. The final number determined to be the "best value" for the calibration control setting was 66. Since the sample contained about 34% Cobalt-60, the efficiency calculated from this measurement was 13%, not inconsistent with previous estimates.

The five samples finally used contained from 14 to 1100 nanocuries.

Calibration Setting Selection

The "best value" of the calibration setting, as noted above, was found to be 66. In the interest of conservatism, it was decided to increase the sensitivity (decrease the calibration setting) to make allowance for two factors - geometric changes and changes in isotopic composition.

When making up the first dispersed sample, the liquid was inadvertently concentrated in one part of the rag bundle, with the result that the monitor reading was affected by whether that section of the bag was adjacent to the wall of the monitor or not. The opportunity was taken to make a series of measurements to determine how large an effect to anticipate as a result of asymmetries in the samples. Based on these measurements, it was decided to make about a 10% allowance for geometric effects.

The most significant effect of changes in isotopic composition on calibration comes from the varying number of gammas per disintegration resulting from varying fractions of Cobalt-60. As mentioned earlier, the measured amount of Cobalt-60 was about 34% of the total activity. This results in an average of about 1.34 gammas per disintegration. If this abundance should drop to 20%, the number of gammas would drop about 10% to 1.20. Even if there were no Cobalt-60 at all, a most unlikely occurrence, the drop in number of gammas would only be 25%. Since this is the non-conservative direction, it was decided to make an additional allowance for this effect that would fully take care of a drop to 20% Cobalt-60, which is about all that could be expected, and provide a reasonable margin for worse situations. With respect to increases in Cobalt-60 fraction, these would cause the results to err in the conservative direction. Actually, the effect of Cobalt-60 on the calibration is somewhat less than calculated here, because the detector is slightly less sensitive to Cobalt-60 gammas than to the lower energy gammas that predominate the other common isotopes.

As a consequence of these considerations and the desire to provide some margin for statistical effects, it was decided to apply an additional 10% sensitivity increase. The final calibration setting selected was 54.

This sort of application of conservatism factors does not lend itself to a simple expression of probable error. We concluded, however, that the measured value was unlikely to be low by more than 3 nanocuries, even for samples of low radioactivity, and unlikely to be high by more than 20%. If the "best value" setting had been used, all of the activity data discussed below would be reduced by 22%.

DATA AND ANALYSIS

Data were collected on 600 consecutive bags of DAW. The data taken on each included bag number, maximum external dose rate, WCM-10 reading, and weight. If the dose rate was low enough (less than 2 mrem/hr), the bag was opened on the frisking table and a general description of the contents recorded. If the contents were such that it seemed likely that much of the radioactivity was concentrated on a few items, these were removed and the bag remeasured. This was only done on a few bags, as in most cases the contamination was widely distributed or the contents were wet.

The data were handled in three batches of 200 bags each. The specific activity of each bag (in nCi/kg) was calculated and lists were prepared ranking the bags by both total and specific activity. The two lists for the first batch are shown in Tables I and II. The data on each bag includes a figure (in the column headed "Fraction Below") showing the fraction of the activity in the batch that is contained in the bags lower on the list. For example, the bag ranked 117th in total activity contained 485 nanocuries and the 83 bags having less activity contained, in total, only 1.23% of the activity in the batch. This allows us to select any cut-off point and readily determine the fraction of the bags and the total activity falling below that cut-off.

The first batch, for which data are shown, contained about 20% more activity than the average for all of the batches, but was otherwise typical. It had a measured content of 0.990 millicuries and weighed 1442.6 kilograms. The average specific activity was 687 nCi/kg. For the 600 bags, the total activity was 2.35 millicuries, the weight was 4366 kilograms and the average concentration was 538 nCi/kg.

Depending on the method of analysis selected, either total or specific activity may be the parameter of interest in calculating radiation exposure. In the first category, for example, one might select 100, 1000, and 5000 nanocuries as cut-off points. Table I shows that the first would include 39 (19-1/2%) of the bags in the first batch, the second would include 106 bags, and the 5000 nCi cut-off would include 147, or almost 3/4 of the bags. The concentration, or specific activity, data can be handled in the same way. One can also calculate the number meeting a combined set of criteria, such as 1000 nanocuries of total activity and 200 nCi/kg. This, for the first batch, would include 103 bags. There are seven more that meet the specific activity criterion only and three that meet the total activity criterion only.

Figure 1 shows the percentage of the bags having less than a given total activity per bag and the percentage having less than a given specific activity. Figure 2 shows the fraction of the total activity as a function of the number of bags with the bags ranked by total content. This curve is close to logarithmic. Both figures include the data for all 600 bags.

There is no way to make a simple graphic presentation of the relationship between dose rate measurements and radioactivity content. It is obvious, of course, that there is a general correlation between the two, but there are many exceptions. This is illustrated, for example, by looking in Table I at the 22 bags measuring over 20 microcuries. Dose rates range from less than 0.5 to 40 mrem/hr. The sequence and values of the numbers are almost entirely unrelated. With respect to the dose rates for the low activity bags, the readings are pretty much a function of the background at the moment and of the care taken by the technician. Unfortunately it was impossible to

have a single technician assigned to this program continuously. Many readings, particularly in the first batch, were simply recorded as "less than" some amount rather than as a specific reading. Close consideration of these data make clear that at the low levels we are concerned with, dose rates cannot be used as a criterion for determining a disposal mode.

CORRELATION OF WCM-10 AND SHIPPING MEASUREMENTS

An attempt was made to correlate the measurements made in this program with the normal shipping measurements. One hundred and thirty-five of the bags were packed into 13 identified drums. From 6 to 18 bags went into each. The total activity in these drums was then determined by the method normally used at Sequoyah in preparing shipping information.

This method consists of making dose rate measurements with a hand-held instrument at a distance of three feet from the hottest point on the drum surface. This value (in mrem/hr) is then multiplied by an empirical constant (which varies with drum weight) to give the radioactivity content in millicuries.

The data produced for shipment yielded results several orders of magnitude higher than the numbers obtained by adding the measured values of the individual bags. For example, one drum, by WCM-10 measurements contained 10.6 microcuries, but by the shipping papers contained 2.0 millicuries. Inquiry showed that the empirical constant for this drum weight (less than 250 pounds) was 20 and the dose rate recorded was "less than 0.1 mrem/hr", presumably the bottom of the scale on the instrument used. The number 0.1 was then used in the calculation with the result noted. The formulae used for heavier drums have larger constants, with the amusing result that a 501 pound drum of clean sand, measured with an instrument that could measure down to natural background levels, would be shipped as containing over 4 millicuries. If measured with the instrument actually used, it would be shipped as containing 30 millicuries!

The equations used at Sequoyah are very conservative. This probably is appropriate for the purpose for which it was intended, but makes them useless for confirming the measurements made in this program.

CONCLUSIONS

The data show that for the conditions existing at Sequoyah during the measurements, most bags of waste had very low levels of radioactivity. Ten percent of them contained about 60% of the total activity and the top 30% contained 90% of the total. Forty percent of the bags contained negligible amounts, totalling about 1% of the entire activity.

The distributions found, however, could not have been found by dose rate measurements with a hand-held instrument. A monitor measuring total activity with a good geometrical efficiency was necessary.

Calibration techniques were demonstrated that show good agreement with laboratory measurements of samples.

If an acceptable set of disposal criteria are developed, a method is at hand for applying the criteria to the DAW stream from a plant. The activity distribution developed for the Sequoyah plant, which clearly shows that a substantial fraction of the DAW is only slightly contaminated, takes on added importance when considered with the fact that DAW comprises approximately 50 % of nuclear plant wastes. The annual disposal cost of DAW is in the range of \$500,000 per

year per unit. Utilities could effect a substantial cost reduction in this area through segregation of waste materials. EPRI intends to support the reexamination of disposal of DAW to determine if all of the present practices are necessary to protect public health and safety.

1. NATIONAL NUCLEAR CORPORATION, "Segregation of Un-

contaminated Dry Active Waste," EPRI NP3299, Electric Power Research Institute (1983).

2. GILBERT ASSOCIATES, INC., "Identification of Radwaste Sources and Reduction Techniques. Volume 1: Implementation Handbook" EPRI NP3370, Electric Power Research Institute (1984).

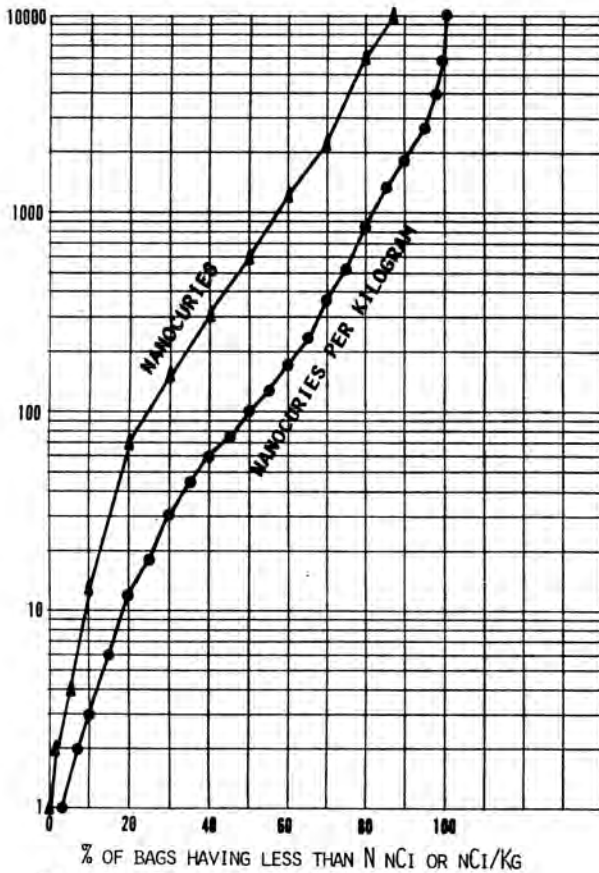


FIG. 1. TOTAL OR SPECIFIC ACTIVITY VS. PERCENTAGE OF BAGS.

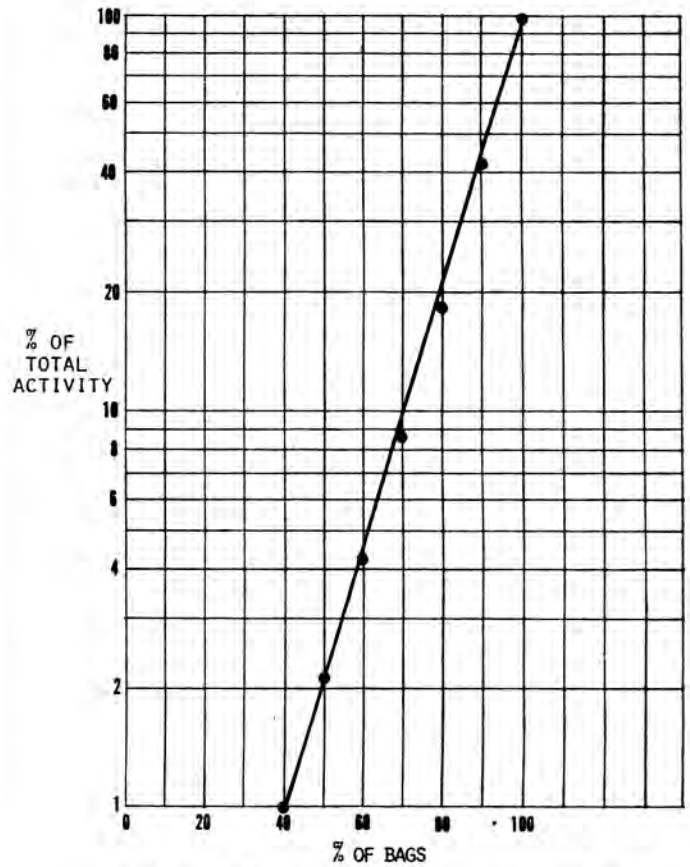


FIG. 2. PERCENTAGE OF TOTAL ACTIVITY VS. PERCENTAGE OF BAGS.