Investigation of Emulsion Characteristics by Neutron Activation

Charles A. Sparrow, Mississippi State University
Donna M. Rogers, Mississippi State University
Victor F. Medina, U.S. Army Corps of Engineers

Introduction

Understanding the effects of a Radiological Dispersal Device (RDD), or “dirty bomb,” requires engineering assessments. One approach for mitigation of effects of a “dirty bomb” explosion is to cover the affected area by a fixative that prevents radioactive materials from migrating. The asphalt/water emulsion is one fixative material that has been studied, with some promising results (1,2). Actual laboratory or field testing with radionuclides normally associated with an RDD, however, is difficult since it is undesirable to spread radiation sources over previously uncontaminated areas. Non-radioactive materials have been used to simulate actual RDD materials, but these may have important differences in terms of solubility, hydroscopic properties, *etc.* If a study is focused on environmental transport, then detectable radiation can be a superb tool to reveal directly the distribution of materials following an explosion, weathering, and treatment application. One method for using the convenient property of detectability is neutron activation. Some stable nuclei are good absorbers of neutrons and yield an activation product of short half-life that can be identified through conventional spectrometry. An inert material, such as a dust sample taken during a field test, can be activated in a subcritical assembly (SCA).

Isotopes of interest for analysis of RDD effects are ^{137}Cs and ^{90}Sr. Direct use of these isotopes in field testing is generally unacceptable. In both cases, a benign material can be used for a test, and the material subsequently irradiated to produce a radioactive material having a half-life of less than one day. The stable isotope of cesium, ^{133}Cs, can be transformed to ^{134m}Cs, which emits a photon of energy 127.5 keV in 13 % of its disintegrations. Using this approach, ordinary non-radioactive CsCl, which has the same chemical properties of the radioactive isotope, may be used in a field test. Samples may then be recovered from key locations, irradiated, and counted using conventional sodium iodide detection equipment. ^{134m}Cs may easily be identified by the presence of the 127.5-keV photopeak.

Charged particle interactions can be studied by means of an interrogating β-particle source. One such source may be produced by neutron absorption in NaCl. The activation product ^{24}Na can be a surrogate for a study of the effects of ^{90}Sr and ^{90}Y, which emit only β-particles. ^{24}Na has a half-life of 14.9590 hours. Hence, radioactive waste disposal measures are unnecessary, since the nuclide decays to the stable species in a few days.

Laboratory measurements using CsCl and NaCl in a subcritical assembly have provided data to indicate that irradiation producing low-activity samples can be successfully used to investigate some of the important questions surrounding transport of radioactive materials as well as the efficacy of treatment approaches.

Nuclear Data

Data for certain nuclides of interest that can be activated in the low-flux assembly are given in Table 1. Cross sections σ are normally presented at the energy corresponding to a speed of 2200 m/s, the speed corresponding to the most probable energy of a neutron at room temperature. The unit of cross section is the barn (b), equal to 10^{-24} cm^2. In any assembly and at any instant of time, neutrons will be characterized by a spectrum of kinetic energies. As a consequence, the average cross section is lower than this reference value. All nuclear data are derived from the "Table of Radioactive Isotopes" (3).

Table 1. Properties of Activation Targets.

Parent Nuclide	Activation Product	Half-life of Activation Product (h)	Gamma Energy (keV)	2200 m/s Cross section for Activation, σ (b)
^{23}Na	^{24}Na	14.959	1368.6	0.530
^{55}Mn	^{56}Mn	2.579	846.77	13.10
^{75}As	^{76}As	25.867	559.10	2.00
^{133}Cs	^{134m}Cs	2.903	127.5	0.310

Application 1—Secondary Radiation Production

Charged particles passing near a nucleus undergo a change in direction and therefore a change in velocity. This process is called bremsstrahlung, which means "braking radiation," and results in emission of photons. Bremsstrahlung photons exhibit a continuous energy distribution. The upper limit of the distribution is equal to the energy of the charged particle, while the minimum energy is zero.

For a β-particle, the bremsstrahlung contribution is described by the following equation (4):

$$f = 3.5 \times 10^{-4} Z E_{max}$$

where

f is the fraction of incident β-particle energy converted into photons,
Z is the atomic number of the absorber, and
E_{max} is the maximum β-particle energy, in MeV.

Emulsion-based fixatives such as those used in McGehee *et al.* (1) and in Fox *et al.* (2) use water as the solvent. Therefore, a value of 8 for Z is appropriate for estimating bremsstrahlung energy production. For ^{90}Sr as the damaging material in an RDD, ^{90}Sr will be in transient equilibrium with ^{90}Y. Maximum β-particle energies are 546 keV for ^{90}Sr, 2280 keV for ^{90}Y, and 1393 keV for ^{24}Na. The fraction of β-particle energy converted into photons from collisions with oxygen is estimated as follows:

^{90}Sr energy conversion fraction $f = 3.5 \times 10^{-4} \times 8 \times 0.546 = 1.53 \times 10^{-3}$
^{90}Y energy conversion fraction $f = 3.5 \times 10^{-4} \times 8 \times 2.28 = 6.38 \times 10^{-3}$
^{24}Na energy conversion fraction $f = 3.5 \times 10^{-4} \times 8 \times 1.393 = 3.90 \times 10^{-3}$.

The emulsions described in the literature are asphalt-based, which can be approximated as consisting of hydrogen, carbon, and oxygen. In such an emulsion, the energy conversion is therefore less than 1 %.

β-particle spectra for ^{90}Sr and ^{90}Y are shown in Figure 1. From the transient equilibrium formula, the activity of ^{90}Y will be 1.000254 times the activity of ^{90}Sr—equal to that of the parent ^{90}Sr within 0.03 percent.

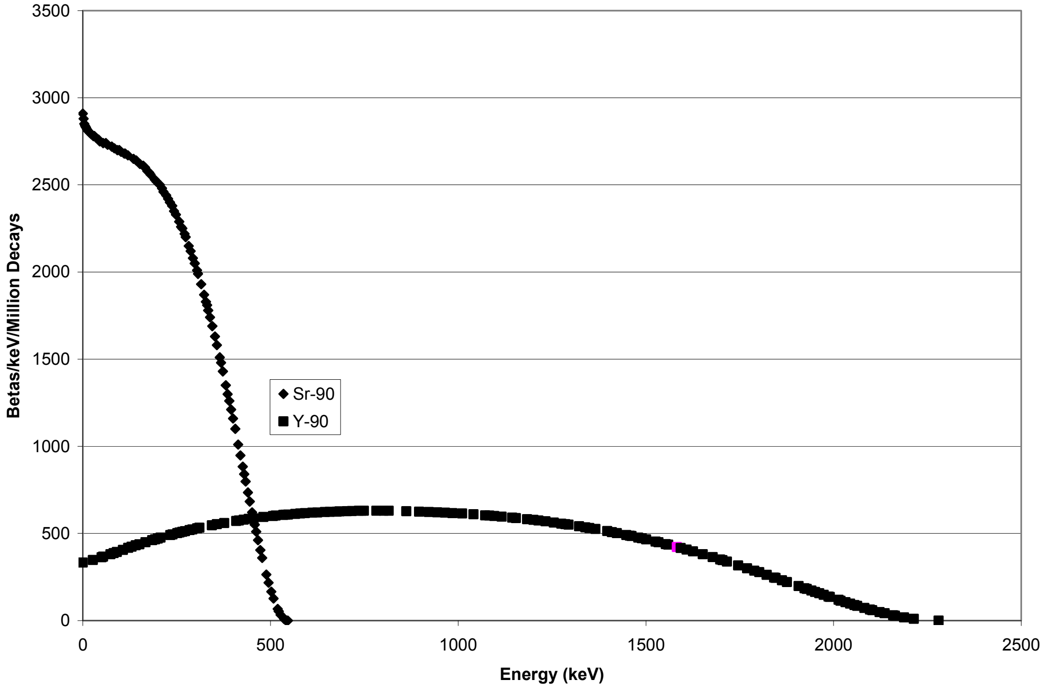

Figure 1. Energy Spectra for Pure Beta Emitters ^{90}Sr and ^{90}Y.

Figure 2 compares the spectrum for ^{24}Na with the combined spectrum of ^{90}Sr and ^{90}Y.

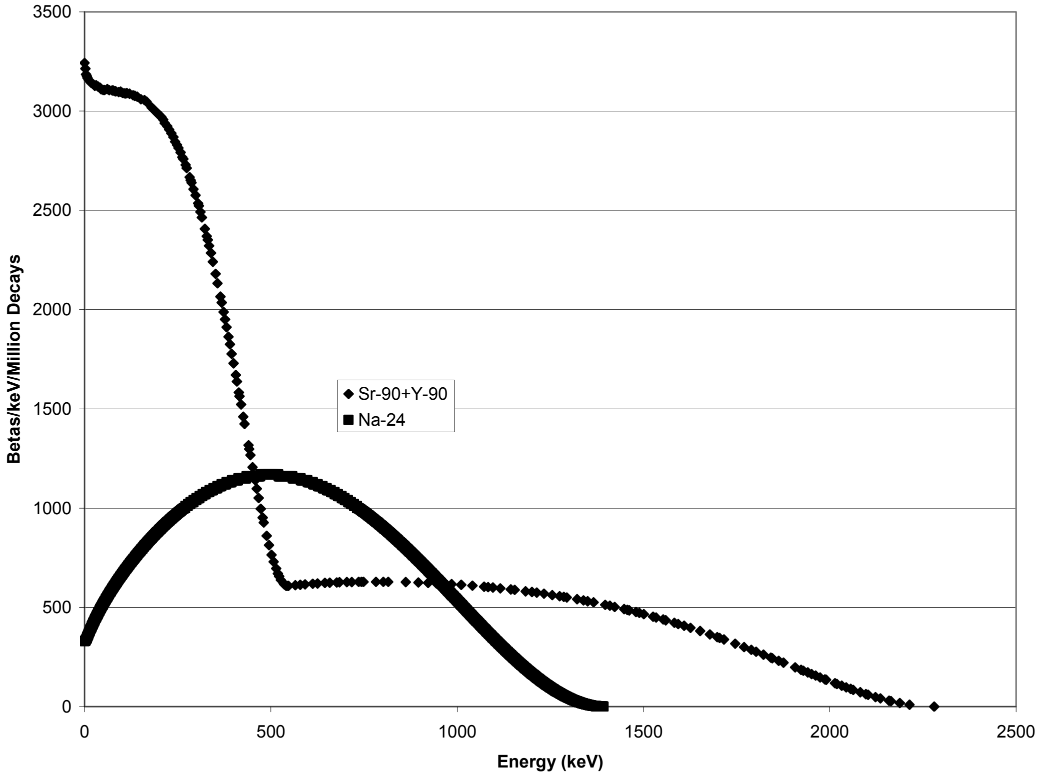

Figure 2. Beta Particle Spectra for ^{24}Na and the ^{90}Sr/^{90}Y System.

Creation of Samples

We found that Na activates easily and produces a β-particle of end-point energy 1392.91 keV in 99.944 % of its disintegrations. Decay of ^{24}Na also produces γ rays of energy 1368.633 keV in every transition and 2754.028 keV in 99.944 % of the transitions. These gamma rays can be used as internal calibration standards, enabling us to use common table salt as the activation target.

The solubility of NaCl is reported to be 36 g/100 mL water (5). If an emulsion is subsequently added to NaCl and the system is irradiated, the β-emitters should be distributed uniformly within the emulsion. Induced activity of the sodium is inferred from the photopeak count of the 1368.633-keV photon. Since the half-life of ^{24}Na is 14.9590 hours, an irradiation time of 3 half-lives, or approximately 45 hours, will provide near-maximum activation of the Na. NaCl and emulsion were placed in a 20-mL vial for irradiation and measurement.

Procedure

For counting, the vial is placed upright on the face of a shielded 3 x 3 NaI detector. Photon spectra are obtained by the following procedure:

1. Place a measured quantity of NaCl into a 20-mL vial.

2. Irradiate the vial and NaCl in a subcritical assembly at Mississippi State University for at least three half-lives of ^{24}Na, or approximately two days.

3. Remove the vial containing NaCl from the SCA and count for 12 hours to obtain a baseline spectrum.

4. Return the vial containing NaCl to the SCA for another activation.

5. Remove the vial containing NaCl from the SCA. Add a known quantity of emulsion to the vial and shake.

6. Perform a 12-hour count of the vial containing the activated salt and the emulsion.

7. From the two spectra, determine

    a. the net count under the 1369-keV peak, and

    b. the gross count in a region of interest selected to include photons from bremsstrahlung.

8. Normalize the spectrum from the vial with emulsion to the spectrum from the same vial without emulsion.

9. Subtract the gross bremsstrahlung region of interest (ROI) counts for the vial without emulsion from the corresponding bremsstrahlung ROI for the vial with emulsion.

Table 2 contains a summary of these results for the sixteen samples analyzed by this procedure. Six different emulsions were provided for assessment:

- Socorro is a high solids, slightly anionic, emulsion (50 to 55%), designed to coat contaminants. It was so named because it was developed for a field demonstration conducted at Socorro, NM.
- Longlife (LL) emulsions have a somewhat lower solids (40%) content. They were designed to have a long shelf life.
- LL + PB is the LL emulsion with a specially formulated Prussian Blue additive.
- LL + IX is the LL emulsion with ion exchange beads.
- 9.6x and 10.6x were second and third generation long life emulsions. They take advantage of proprietary blending of specially selected asphalt stocks. They also have Prussian Blue additives.

The LL series emulsions show a greater bremsstrahlung contribution than do the Socorro, the 9.6x, and the 10.6x emulsions. In this table, only the 1369-keV photon peak was used for normalization. The region of interest for bremsstrahlung radiation was arbitrarily chosen as the energy range 150-450 keV.

Table 2. Bremsstrahlung Contribution.

Vial ID	Gross Area of 150-450 keV ROI without Emulsion	Gross Area of 150-450 keV ROI with Emulsion	Normalized Gross Area of Bremsstrahlung ROI with Emulsion	Increase due to Bremsstrahlung from Emulsion	Fractional Increase due to Bremsstrahlung
Vial 1 (Socorro)	1228777	1252086	1282099	53322	0.0434
Vial 2 (LL)	1382356	1357848	1511372	129016	0.0933
Vial 3 (LL + PB)	1244872	1205638	1393681	148809	0.1195
Vial 4 (LL + IX)	1170640	1094780	1436566	265926	0.2272
Vial 5 (9.6x)	1235352	1366105	1287223	51871	0.0420
Vial 6 (10.6x)	1385157	1394868	1494333	109176	0.0788
Vial 7 (LL)	1194821	1274843	1371048	176227	0.1475
Vial 8 (LL)	1156688	1085284	1458875	302187	0.2613
Vial 9 (LL + IX)	1100485	1026204	1310199	209714	0.1906
Vial 10 (LL + IX)	1047228	1117370	1239740	192512	0.1838
Vial 11 (LL + IX)	1146521	1091089	1407650	261129	0.2278
Vial 12 (LL + IX)	1107101	1032790	1388716	281615	0.2544
Vial 13 (LL + PB)	1046000	1085340	1219646	173646	0.1660
Vial 14 (LL + PB)	1137914	1137225	1255098	117184	0.1030
Vial 15 (LL + PB)	1054538	1096501	1085493	30955	0.0294
Vial 16 (LL + PB)	1039869	1194040	1143322	103453	0.0995

Application 2: Interrogation of Polymer-based Detector Materials (6)

The use of scintillating polymers is of interest as a means of visible light production. An application is to the distribution of β-emitters such as the ^{90}Sr/^{90}Y system. More generally, spills of radioactive material on floors would be assessed if a light-producing material could be distributed over the spill.

In one concept, a powder can be broadcast over an area of suspected contamination. Some liquid will be necessary to facilitate transport of radionuclides into a microporous network of the scintillating solid. Ions can be selectively concentrated and brought into proximity of fluorophores that transduce radiation into visible light. If this visible light can be detected without the use of active electronic packages, identification of radioactive areas can be made in short times. Laboratory testing of the combinations of solvent, primary scintillators, and wave shifters can be performed using ^{24}Na as the source of β-particles.

Alternatively, fluorophores may be placed into polymer microbeads which, through the use of anion exchange extractants, selectively remove and concentrate radionuclide ions. The choice of fluorophore is dictated by the type of radiation while the choice of ion-exchange extractant is driven by the radionuclide to be concentrated.

In another approach, the fluorophore and extractant can be incorporated into a silica matrix using sol-gel chemistry. The product will be microporous and will sequester the salt of a radioactive material to bring the source in close proximity to the fluorophore. The particle size and density can be adjusted to produce a slightly translucent solid, permitting light to escape the particle.

Use of β-emitters of short half-life enables a study of combinations of fluorophore and extractant with the objective of identifying those which yield the most-easily detected photons.

Results and Conclusions

Activation in a subcritical assembly produces activities sufficient for counting by conventional equipment. Photons from β-γ emitters can be used to provide relative intensities of charged particles used for interrogation.

The significance of the method of taking difference between the ROI counts of two spectra can be analyzed by taking the difference between ROI counts of spectra for the samples containing NaCl only. Two spectra from separate vials are normalized to the integral count of the 1369-keV photon from ^{24}Na disintegration. The difference in counts from the bremsstrahlung ROI is computed. This difference may be compared to the corresponding one calculated from the vial with emulsion and the one from the same vial without emulsion. Gross areas of the bremsstrahlung region are used in the calculation. The magnitude of fractional differences ranges from ~0 to 0.092, with an average fractional difference of 0.0425. The LL, LL + PB, and LL + IX emulsions show a greater fractional difference than 0.0425, whereas the Socorro, 9.6x, and 10.6x emulsions have a smaller fractional difference.

Sensitivity analysis can be performed to determine the effect attributable to the definition of the region of interest for bremsstrahlung. Also, analytical removal of the contribution of the annihilation peak at 511 keV may be performed. However, this contribution is present in all sets of data, and differencing calculations effectively delete any effect attributable to annihilation counts.

References

1. McGehee, T., V. F. Medina, R. Martino, A Bednar, C. Weiss, and D. Abraham. 2007. “Fixation of Heavy Contaminants of a Dirty Bomb Attack: Studies with Uranium and Metal Simulants,” *Environmental Progress*, **26**(1): 94-103.
2. Fox, G. A., J. W. Fuchs, V. F Medina, and K Atapattu. 2007. “Capture of Air Particulates Using Surface Applied Emulsions: Potential for Post Detonation Dirty Bomb Cleanup,” *Journal of Environmental Engineering*, **133**(3): 255-262.
3. Ekström, L. P. and R. B. Firestone, “WWW Table of Radioactive Isotopes, database version 2/28/99 from URL http://ie.lbl.gov/toi/index.htm.”
4. Cember, Herman, **Introduction to Health Physics**, 2nd Edition, New York: Pergamon Press, 1988.
5. Dean, John A., **Lange’s Handbook of Chemistry**, 15th Edition, New York: McGraw-Hill, 1999.
6, Mark G. White, Personal Communication, July 10, 2008.

Acknowledgment

Emulsions for this study were provided by Encapco Technologies, LLC. Funding was provided by the United States Army Research, Development and Engineering Command, Armaments Research, Development and Engineering Center in Picatinny, NJ.