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Abstract Source term information is required for to reconstruct a device used in a dispersed radiological dispersal device. Simulating a radioactive environment to train and exercise sampling and sample characterization methods with suitable sample materials is a continued challenge. The Idaho National Laboratory has developed and permitted a radioactive response training range (RRTR), an 800 acre test range that is approved for open air dispersal of activated KBr, for training first responders in the entry and exit from radioactively contaminated areas, and testing protocols for environmental sampling and field characterization. Members from the Department of Defense, Law Enforcement, and the Department of Energy participated in the first contamination exercise that was conducted at the RRTR in the July 2011. The range was contaminated using a short lived radioactive ^{82}Br isotope (activated KBr). Soil samples contaminated with KBr (dispersed as a solution) and glass particles containing activated potassium bromide that emulated dispersed radioactive materials (such as ceramic-based sealed source materials) were collected to assess environmental sampling and characterization techniques. This presentation summarizes the performance of a radioactive materials surrogate for use as a training aide for nuclear forensics.

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Information about the material source term of a detonated radiological dispersal device (RDD) is required to reconstruct the device in order to determine its origin, potential legitimate use and possibly identifying the point of loss of control. An important part of the reconstruction effort is to collect samples and data that provide information about the source or device. Doing this requires the development of suitable collection techniques with known sampling efficiencies in order to accurately estimate the material source term. Simulating a radioactive environment with suitable surrogate materials to test sampling and characterization methods is a challenge.

To date, most RDD training and exercise venues have used ^{99}Mo or ^{67}Ga as a source of short lived radionuclides. These isotopes have limited applicability as radioactive surrogates because they cannot be used in solid form, and thus cannot adequately emulate exploded, dispersed solid material for sampling. Liquids are absorbed into soils and porous surfaces that prohibit accurate sampling of solely the source material. INL has developed radioactive surrogate materials for salt and glass/ceramic forms of radioactive source materials using neutron activated potassium bromide (KBr). This paper summarizes the performance of the radioactive materials surrogate for use as a training aide for nuclear forensics.

Introduction

Field sampling is an important aspect of reconstructing a detonated radioactive dispersal device (RDD) to determine the initial material activity and the number of sources and

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Fig. 1 Photograph (*left*) and micrograph (*right*) of the 25 % by mass KBr containing sol–gel

Table 1 Calculated isotope activities (Ci) at various cooling times after discharge for 1 g KBr irradiated for 1 h

Decay (curies)										
Isotope	Initial	1.0 h	8.0 h	16.0 h	1 day	3 days	1 week	2 weeks	3 weeks	4 weeks
^{33}P	1.03E−13	1.03E−13	1.02E−13	1.01E−13	9.99E−14	9.46E−14	8.48E−14	7.00E−14	5.78E−14	4.77E−14
^{36}Cl	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11	1.68E−11
^{38}Cl	1.24E−05	4.06E−06	1.63E−09	2.15E−13	2.83E−17	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{39}Ar	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07	1.10E−07
^{41}Ar	6.48E−05	4.43E−05	3.12E−06	1.50E−07	7.20E−09	8.87E−17	1.35E−32	0.00E+00	0.00E+00	0.00E+00
^{40}K	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10	2.83E−10
^{42}K	7.78E−03	7.35E−03	4.97E−03	3.17E−03	2.03E−03	1.37E−04	6.30E−07	5.11E−11	4.19E−15	5.07E−17
^{43}K	1.39E−10	1.34E−10	1.08E−10	8.43E−11	6.57E−11	1.48E−11	7.49E−13	4.05E−15	2.19E−17	1.18E−19
^{81}Se	2.27E−09	4.09E−10	1.35E−12	4.03E−15	1.21E−17	8.67E−33	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{81m}Se	3.05E−10	1.48E−10	9.13E−13	2.73E−15	8.18E−18	5.88E−33	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{80}Br	1.19E+00	2.87E−01	6.54E−02	1.86E−02	5.31E−03	2.86E−06	8.26E−13	2.98E−24	1.49E−35	0.00E+00
^{80m}Br	2.14E−01	1.83E−01	6.10E−02	1.74E−02	4.96E−03	2.67E−06	7.71E−13	2.78E−24	9.94E−36	0.00E+00
^{82}Br	4.74E−02	4.72E−02	4.12E−02	3.52E−02	3.01E−02	1.17E−02	1.78E−03	6.58E−05	2.43E−06	8.98E−08
^{82m}Br	2.87E−01	3.24E−04	7.68E−25	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00
^{83}Br	1.46E−07	1.10E−07	1.45E−08	1.44E−09	1.43E−10	1.36E−16	1.24E−28	0.00E+00	0.00E+00	0.00E+00
^{79}Kr	9.61E−13	9.42E−13	8.20E−13	7.00E−13	5.97E−13	2.31E−13	3.46E−14	1.24E−15	4.48E−17	1.61E−18
^{83m}Kr	1.00E−07	1.08E−07	3.62E−08	4.86E−09	5.43E−10	5.73E−16	5.22E−28	0.00E+00	0.00E+00	0.00E+00
Total	1.74E+00	5.25E−01	1.72E−01	7.44E−02	4.24E−02	1.19E−02	1.78E−03	6.59E−05	2.54E−06	2.00E−07

types that hypothetically were used in an RDD event. Of particular importance to the forensics community are ^{137}Cs and ^{90}Sr sources. The chemical forms of these isotopes are either pressed salt pellets (CsCl), glass silicates such as cesium or strontium silicates (Cs_2SiO_3, $SrSiO_3$) or ceramics such as cesium pollucite ($Cs_2Al_2Si_4O_{12}{\cdot}12H_2O$) or strontium titanate ($SrTiO_3$). INL has developed radioactive surrogate materials for salt and glass/ceramic forms of radioactive source materials using neutron activated KBr. The KBr has similar properties (i.e. melting point, structure) to CsCl and has also been incorporated into high purity sol–gel glasses to simulate Cs silicate and ceramic forms. KBr has similar crystal and physical properties to CsCl [1]. Each compound is found as a cubic lattice with similar heats of formation ($\Delta H_f = -103.5$ and -93.7 kcal mol^{-1}). The melting point and boiling points of KBr are 1,007 and 1,708 K, respectively; the melting and boiling points of CsCl are 918 and 1,563 K, respectively. Both compounds are highly soluble in water. Hence, KBr should be a good surrogate for CsCl for a variety of dispersion mechanisms (i.e. spray or explosion) and thus should provide realistic dispersions for training.

With respect to the glass matrix, it is synthesized using a sol–gel technique that can be readily doped with upwards of 25 % KBr by mass. The glasses are high purity which is beneficial for irradiation to avoid unwanted activation products. The particle size can be varied from micron up to millimeters in size and the density is similar to other glasses used in radioactive sources.

Potassium has three natural occurring isotopes, ^{39}K (93.3 %), ^{40}K (0.012 %) and ^{41}K (6.73 %). The primary radioactive potassium isotope formed is ^{42}K ($T_{1/2} =$ 12.4 h) that decays to stable ^{42}Ca by beta emission. The thermal capture cross section of ^{41}K is 1.5 barns [2]. The

Table 2 Sample masses and calculated ^{82}Br activities at sample collection times

Irradiation sample	Irradiation rabbit	Sample mass	Estimated ^{82}Br activity (MBq) decay corrected to 13:00 7/25/2011	Estimated ^{82}Br activity (MBq) decay corrected to 13:00 7/26/2011
1	1	3.495	31	20
2	1	3.616	32	20
3	2	2.153	34	22
4	2	2.097	33	21
5	3	0.8787	20	13
6	3	0.9584	22	14
7	3	0.9663	22	14
8	4	0.983	26	16
9	4	1.024	27	17
10	4	0.988	26	16

Table 3 Measured sample dose rates and activities ^{82}Br from collections

Field collection sample	Sample dose rate (mRem/h @ 5 cm)	Measured sample activity (MBq)
13	15	1.4
14	40	2.3
15	60	3.0
16	50	2.0
17	10	1.0
18	60	2.7

anthropogenic production of ^{40}K is insignificant relative to the natural quantity of ^{40}K found in the salt. Bromine has two naturally occurring isotopes ^{79}Br (50.7 %) and ^{81}Br (49.3 %) that activate by (n, γ) reactions to form ^{80}Br ($T_{1/2} = 17$ min) and ^{82}Br ($T_{1/2} = 35.3$ h) [3]. Each Br isotope decays to stable Kr isotopes. After 72 h from end of irradiation, ^{82}Br is the primary radionuclide contributing to gamma emission and radiation dose rate; it has major gamma emission rays at 776 keV (83.5 %) and 554 keV (70.8 %) [4]. From the perspective of a short lived training stimulant, the irradiated KBr has no long-lived detrimental radioisotope; traces of Se are formed but in insignificant quantities, the calcium and krypton decay produces stable isotopes.

INL has built the radioactive response test range (RRTR), an 800 acre training site that is permitted to release 37 GBq of irradiated KBr activation products. An exercise was conducted that used irradiated glass containing 25 % by mass KBr as a radioactive sample surrogate to estimate sampling efficiency. This paper presents the result of the use of the surrogate KBr glasses obtained from a field collection exercise.

Experimental

Preparation of glasses using sol–gel synthesis

Potassium bromide containing glasses were prepared using a sol–gel synthesis [5] in 50 g batches that result in a variety of particle sizes ranging from micron to millimeter in size. The silica matrix was formed by the acidic hydrolysis of tetraethyl orthosilicate (Sigma 99.999 % trace metals basis) for 4 h at a temperature of less than 313 K. High purity (99.999 %) "Spectrograde" KBr powder (Crystal Labs) was dissolved into the mixture. The silica was cross linked by the addition of high purity KOH (Sigma 99.99 % semiconductor grade) to adjust the pH of the solution to 8.6. The solidified silicate was calcined for 3 h at 873 K resulting in a dense glass particulate; at temperatures above 873 K vaporization and loss of the bromine was observed. A photograph of the batch produced sol–gel silicate glass is shown in Fig. 1. KBr was incorporated into separate glass batches at concentration levels of 25 % by mass.

Neutron irradiation and sample preparation

ORIGEN [6] calculations were used to estimate the sample activity. For the purposes of this calculation, 1 g KBr was modeled in a thermal neutron flux of 2.5×10^{12} n cm^{-2} s^{-1} for an irradiation time of 1 h. The total activity of the activated KBr at discharge was estimated to be 0.22 GBq g^{-1} h^{-1} and decayed accordingly for a variety of cooling times. The resulting activity calculated at discharge out to 4 weeks is provided in Table 1. The activities reported in Table 1 are reported in Curies, the output of ORIGEN code.

Polyethylene containers were pre-cleaned with high purity 2-propanol then rinsed with 18 Mohm water. Ten glass samples containing KBr at 25 % by mass ranging from 1 to 4 g were loaded into polyethylene containers and irradiated in aluminum rabbits for 1 h. Gold wires were irradiated to confirm the reactor flux. Sample sizes, irradiation times and cooling times were adjusted in order to irradiate sufficient samples for a minimum of ten sample collection operations. The sample masses and calculated activity of the samples at the time of shipment from the irradiation facility is provided in Table 2. The time of shipment was used because the samples were irradiated on different days due to handling limitations in the reactor and the time of shipment is within a few hours of deposition and the onset of the experiment. The estimated activity

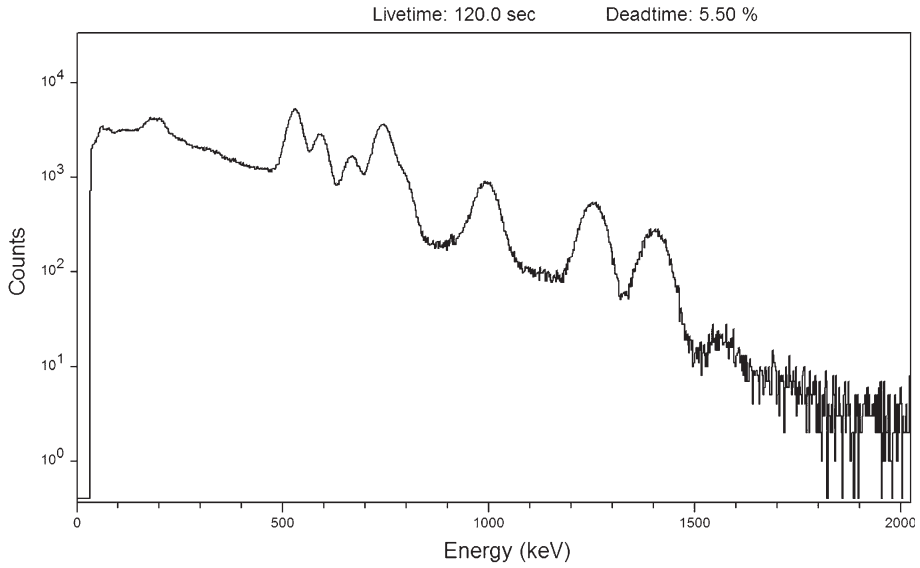

Fig. 2 NaI spectrum irradiated KBr containing sol–gel glass

ranged between 20 and 34 MBq. Gamma spectrometry results confirmed the purity of the glass after activation and the activity of the ^{82}Br. The measured activity matched the calculated activity within 10 %.

Field sample set-up

Sampling platforms were constructed from 4 × 4 ft wood pallets that were coated with 1 inch of asphalt or concrete. The pallets were placed in 10 × 12 ft canvas tents equipped with snap in canvas floors. This provided a containment that was required to contain the irradiated silicate glass and permitted unhindered entry. The irradiated glass particulate was deposited manually onto the sampling surfaces. Samples were collected manually or using portable vacuum samplers. Ten sampling stations were established in the RRTR gravel pit. Fireplace ash, indigenous soil and gravel were sprinkled on the surfaces with the KBr to provide a matrix that would emulate ash from fires and debris from collapsed structures. The dose rates for samples measured at contact (2 inches) deposited at the sampling stations is provided in Table 3. The radiation levels of the sol–gel samples at use did not present a handling hazard and provided a measurable dose rate at distances out to 1 m. The 35 h half-life of the ^{82}B provided sufficient activity for the 3 days use.

Samples numbered 13, 14 and 22 were collected at stations where 1 g of irradiated KBr doped glass was deposited; samples 17, 18, 19, and 20 were collected at stations that contained approximately 3.5 g of the irradiated KBr doped glass. Field processing consisted of measuring the dose rates at contact (5 cm) from each sample and obtaining the sample spectrum and activity with a NaI or HpGe detector. One sample, number 13, was processed by sieving onto three sieves with mesh sizes greater than 2 mm, greater than 850 microns, and greater than 500 microns were used to process the sample. This process was evaluated to provide a sample that had minimal environmental contamination (i.e. dirt) that confounds the determination of the age since material purification. Sample activities were measured using a sodium iodide detector.

Results and discussion

Several processes were evaluated in this process. Adequate samples with sufficient activities were collected and processed. An important step was sample sieving. For this process, the glass was robust and could survive sample sieving that removed as much soil and ash as possible. The greater than 850 μm sieve contained 58 % of the sample mass and 65 % of the sample activity. The greater than 500 μm sieve contained 25 % of the sample mass and 29 % of the sample activity. The less than 850 μm sieve fraction contained 17 % of the sample mass and only 7 % of the sample activity. This indicates a significant fraction of the ash and soil could be removed by sieving for this particle size distribution. It is to be determined if sample sieving could be beneficial to an explosively dispersed RDD for salt, glass or ceramic materials.

A second important test was the ability of field teams to correctly identify and assay the samples using gamma spectrometry. A screen capture of a representative spectrum obtained from field measurements of the irradiated sol–gel KBr containing glass obtained using a NaI detector is provided in Fig. 2. Analogously, a screen capture of a higher resolution gamma spectrum obtained with an Ortec Detective high purity germanium detector in the field is

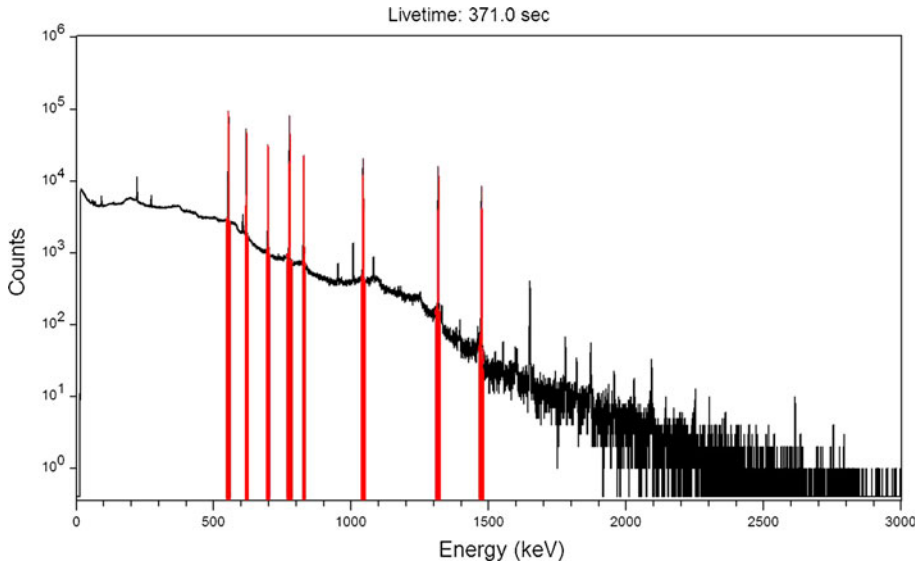

Fig. 3 HpGe spectrum irradiated KBr containing sol–gel glass

shown in Fig. 3. The ^{82}Br spectrum provides an adequate number of gamma emission lines for training in peak and species identification and quantification. The activity in each sample was determined using spectra obtained from collected samples. The total ^{82}Br activity in the samples ranged from 0.96 to 4.1 MBq. The manual dispersion process for depositing the glass samples resulted in a heterogeneous distribution of the samples on the sampling platforms. This made the direct assessment of the sampling efficiency difficult. Collectively, the measured sample activities were approximately 1/10–1/5th of the activity placed on the sample pods based on the deposited and measured sample activities. This represents 10–20 % of the activity deposited for collection. This is the first attempt to assess the sample collection efficiency using radioactive samples. To obtain a better estimate, the homogeneity of the sample dispersion needs to be improved to more accurately assess sampling efficiency.

Conclusions

Irradiated samples of glass containing 25 wt% KBr were used at INL's RRTR to test sample collection efficiency. The collection efficiency was estimated to range from 10 to 20 %. The glass samples provided a medium to test sample collection and measurement techniques. Activated KBr contained in glass particulate was dispersed to provide radioactive samples for collection. The samples provided a realistic medium for sample collection of controlled surfaces with in-field characterization.

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