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Decontamination of select infrastructure materials after a radiological incident using a water-based formulation

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ABSTRACT

This paper summarizes the results of the decontamination of the infrastructure materials concrete, limestone, brick and asphalt contaminated with ^{60}Co, ^{85}Sr, ^{137}Cs and ^{241}Am. The paper focuses on the effect of differences in substrate properties and of the pH of the radionuclide solution used for surface contamination on adsorption or ion exchange of the radionuclides and how these factors affect the decontamination effectiveness. A six-component chemical formulation was used and a process effectiveness of up to 76% was obtained depending on the substrate and radionuclide. Asphalt was the easiest material to decontaminate because of its more hydrophobic nature. Concrete and limestone (and to some extent brick) were less effectively decontaminated as their porous surfaces allowed penetration of radionuclides into water-filled pores in the substrate facilitating adsorption or ion exchange and making them difficult to remove. Brick was the most difficult material to decontaminate because the major component of brick is clay which retains most mono- and divalent ions. The removal of ^{60}Co, ^{85}Sr and ^{137}Cs from the surfaces of concrete, limestone and brick increased when the pH of the radionuclide solutions was moderately acidic to neutral compared to when they were highly acidic. The variability in the test results was similar to that observed in other studies using other decontamination methods, attributed to the inhomogeneity of the substrates used and considered representative of real infrastructure materials.

1. Introduction

There is significant interest in decontamination of critical infrastructure contaminated in a radiological nuclear (RN) incident, including accidental releases such as at Chernobyl in 1986, and the Fukushima Daiichi Nuclear Power Plant (FDNPP) in 2011, or intentional releases involving the use of a Radiological Dispersal Device (RDD) or an Improvised Nuclear Device (IND) (Samuleev et al., 2013). Decontamination and remediation of evacuated areas such as residential houses, hospitals, power plants, schools, roads, farmland, forests, etc., after the FDNPP incident highlighted the need for highly effective processes due to the scale of the accident and types of contaminated materials and radionuclides involved (IAEA, 2011; Yao et al., 2014; Semmler et al., 2014). Of particular interest are so called "low-tech" decontamination processes which must be effective for removal of contamination from a variety of materials such as concrete, limestone, brick, asphalt, etc., under a variety of climate conditions, are easily scalable, rapidly deployable, cost effective, commercially available, non-destructive and environmentally friendly.

An RDD (also known as dirty bomb) is any weapon that is designed to spread radioactive material with the intent to kill and cause disruption. Dirty bombs would use conventional explosives to spread radioactive material, such as the spent fuel from NPPs or radioactive medical waste, and could therefore contain alpha-, beta-, and gamma-emitting radionuclides. Radioisotopes that pose the greatest security risk are created in NPPs and include ^{60}Co, ^{90}Sr, ^{137}Cs, ^{192}Ir, ^{226}Ra, ^{238}Pu, ^{241}Am, and ^{252}Cf (Holbrook, 2005). For INDs, initial (detonation) hazard will be from the heat, overpressure (shrapnel), and radiation, and later hazards will result from the fallout; the chemical composition of the fallout will depend on the location of the detonation.[1] RDDs can generate terror but cause few fatalities while an IND is a crude nuclear device that has the potential to cause building collapse and generate high radiation doses, heat and fire and result in significant loss of life.

In Canada, Defense Research and Development Canada (DRDC),

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[1] INDs usually explode at ground level, rather than at an altitude which may cause irradiation of materials on the ground.

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while supporting work in other laboratories, has also built facilities to conduct tests with radiological materials. DRDC laboratories have the capability to perform tests simulating an RN environment using MBq quantities of short half-life radioactive materials such as ^{24}Na (surrogate for ^{137}Cs with $t_{1/2}$ = 0.63 d), ^{85}Sr, ^{140}La, ^{192}Ir (surrogate for ^{60}Co with $t_{1/2}$ = 74 d) and ^{225}Ac, simulating RDD contamination and IND fallout under different environmental conditions (wet and dry deposition) with different particle sizes, on targets ranging from 10 cm × 10 cm coupon surfaces to larger equipment such as light armoured vehicles. Recently, DRDC and the US Environmental Protection Agency (EPA) tested the removal of ^{24}Na, ^{85}Sr, and ^{140}La from the surfaces of concrete and asphalt by a vacuuming technique; the study simulated fallout from an IND using different particle sizes (20–800 μm) (Desrosiers, 2012).

In the US, the National Homeland Security Research Center (NHSRC) was established to conduct research and deliver products that improve the capability of the agency to carry out these responsibilities. NHSRC created the Technology Testing and Evaluation Program where independent and quality assured testing (EPA, 2011a) of the performance of commercially available technologies are carried out to supplement vendor-provided information. The EPA's NHSRC has evaluated many commercial products such as Rad-Release I (RRI) and Rad-Release II (RRII) developed by Environmental Alternatives Inc. (EPA, 2011b), Argonne SuperGel (EPA, 2011c), CBI Polymers DeconGel (EPA, 2011d), NTEK ND-75 and NTEK ND-600 (EPA, 2011e), RDS 2000 (EPA, 2011f), and a modified product based on Allen-Vanguard's Surface Decontamination Foam (SDF™), known as Universal Decontamination Formulation (UDF)[2] (EPA, 2013a). Evaluation of four technologies including SDF™, UDF, ASG and RRII was carried out on surfaces with new and aged ^{137}Cs (EPA, 2013b), and the work was expanded to include the removal of ^{243}Am from the surfaces of granite in addition to concrete (EPA, 2013c). The effectiveness of DeconGel, RRII, ASG, INTEK Technologies LH-21 and RDS 2000 for decontamination of surfaces of materials including limestone, marble, granite, asphalt and concrete contaminated with ^{60}Co, ^{85}Sr, ^{137}Cs, and ^{243}Am were assessed (EPA, 2013c). Although the composition of the chemicals and foams used in these evaluations were not specified due to their proprietary nature, the results obtained can be compared with the results obtained in this work in terms of ease of deployment, requirements and conditions, decontamination effectiveness and waste volumes generated.

The purpose of this work was to test the effectiveness of a water-based chemical decontamination formulation developed and tested at Environment and Climate Change Canada (ECCC) (Volcheck et al., 2018). To this end extensive preparation for experimental work was initiated at Chalk River Laboratories (CRL) in mid-2015. Two temporary ventilated enclosures (TVEs) were erected to place equipment and conduct the tests, three vertical test platforms were designed and constructed, each holding 15 large coupons (surface areas of 225 cm^2) to allow exposure of the coupon surfaces to radionuclides in a vertical position simulating an RN accident. Four test materials, concrete, limestone, brick and asphalt, and four radionuclides, including ^{60}Co, ^{90}Sr, ^{137}Cs and ^{241}Am, were selected for testing. A portable gamma-spectrometer with an air-cooled detector was used for in-situ measurements of surface activities before and after decontamination. Strontium-90, a beta emitter, was replaced with ^{85}Sr to allow the detection of the four radionuclides using the gamma-spectrometer. This paper summarizes work carried out to prepare for the tests, and results from tests evaluating the effect of several test parameters in order to determine their impact on mitigation/decontamination process effectiveness.

2. Experimental

2.1. Test materials

Coupons of concrete, limestone, brick and asphalt were cut to dimension 15 cm × 15 cm; the concrete, limestone and asphalt coupons were 5 cm thick, while the brick coupons were 3 cm thick (Fig. 1). Concrete blocks (90 cm × 15 cm x 5 cm) were purchased from Central Precast Inc. (Ottawa, ON, Canada). Aged asphalt (more than 10 years old) was obtained from Tomlinson Construction (Ottawa, ON, Canada). Brick and limestone were purchased from Merkley Supply Ltd. (Ottawa, ON, Canada). Brick coupons were made from Clay Flue Liners manufactured by Clay Superior Corporation (Uhrichsville, OH, USA) and the limestone was made by Arriscraft International (Cambridge, ON, Canada). All materials were cut to dimension (15 cm × 15 cm) at Arban Stoneworks Ltd. (Ottawa, ON, Canada), and the coupon surfaces were rinsed with deionized (DI) water, dried,[3] and wrapped individually prior to use.

The coupons cut from the four materials tested in this work each had a surface area of 225 cm^2 and visual examination of the surfaces indicated variations in porosity and surface roughness; occasionally small minor scratches and dents were identified. Prior to testing, coupon surfaces were visually examined and coupons were rejected if they appeared to have surface defects.

Elemental composition analyses and measurements of carbonate (CO_3^{2-}) concentration and total carbon content for samples of concrete, brick and limestone were carried out using Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES), ion chromatography (IC) and a carbon and sulphur analyzer (CS230), respectively. For elemental analysis, approximately 0.2 g of each material were crushed into a powder and digested in a mixture of concentrated acids[4] before elemental analyses. In each sample, some residue was left undissolved and required additional digestion prior to analysis.[5] For carbonate analyses, approximately 0.2 g of each material were leached in 10 mL of DI water overnight and the leachate was analysed by IC. The estimated uncertainty for elemental analyses was ± 10% and for carbonate analysis was ± 25%. For total carbon content, samples were analysed as received. The elemental composition of the materials are summarized in Table 1.

Concrete is made of cement (mainly Portland cement), aggregate, and water. The major components of concrete are calcium silicate hydrate (C-S-H) and calcium hydroxide (C-H) with smaller concentrations of tri-calcium aluminate and tetra-calcium alumino ferrite. The major elements in the concrete sample used in this work were calcium (Ca) (205 mg/g) and silicon (Si) (105 mg/g).[6] The calcium to silicon ratio (Ca/Si) of approximately 2 suggests a mixture of calcium silicate hydrate and calcium hydroxide as the major components (Cong and Kirkpatrick, 1996). In addition, smaller amounts of magnesium (Mg) (48 mg/g), aluminum (Al) (24 mg/g), iron (Fe) (15 mg/g), potassium (K) (9 mg/g), and sodium (Na) (6 mg/g) were measured.

The major elements in limestone were Ca (245 mg/g) and Mg (154 mg/g) with smaller concentrations of silicon (Si). Based on the elemental composition of the limestone sample, it appears that the limestone used in these tests is calcitic dolomite (Geological and Land Survey, 2011). The two major elements in the brick sample were Si (251 mg/g) and aluminum (Al) (116 mg/g) with smaller concentrations

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[2] The UDF process was developed by Environment Canada, renamed Environment and Climate Change Canada in 2015.

[3] The cleaning and drying step after the coupons were cut was only to remove any debris from cutting.

[4] A mixture of nitric acid, hydrochloric acid, and hydrofluoric acid was used for digestion.

[5] The undissolved residue was heated to ~900 °C and the melted residue dissolved in nitric acid first followed by dissolution in concentrated sodium hydroxide before analysis.

[6] Average values of two measurements (see Table 1).