Journal of Environmental Radioactivity 148 (2015) 183–195

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Spatially-resolved analyses of aerodynamic fallout from a uranium-fueled nuclear test

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ARTICLE INFO

Article history:
Received 15 January 2015
Received in revised form
10 April 2015
Accepted 11 April 2015
Available online 28 July 2015

Keywords:
SIMS
Trinitite
Fallout
Device debris
Nuclear forensics

ABSTRACT

Five silicate fallout glass spherules produced in a uranium-fueled, near-surface nuclear test were characterized by secondary ion mass spectrometry, electron probe microanalysis, autoradiography, scanning electron microscopy, and energy-dispersive x-ray spectroscopy. Several samples display compositional heterogeneity suggestive of incomplete mixing between major elements and natural U ($^{238}U/^{235}U = 0.00725$) and enriched U. Samples exhibit extreme spatial heterogeneity in U isotopic composition with $0.02 < {}^{235}U/{}^{238}U < 11.84$ among all five spherules and $0.02 < {}^{235}U/{}^{238}U < 7.41$ within a single spherule. In two spherules, the $^{235}U/^{238}U$ ratio is correlated with changes in major element composition, suggesting the agglomeration of chemically and isotopically distinct molten precursors. Two samples are nearly homogenous with respect to major element and uranium isotopic composition, suggesting extensive mixing possibly due to experiencing higher temperatures or residing longer in the fireball. Linear correlations between $^{234}U/^{238}U$, $^{235}U/^{238}U$, and $^{236}U/^{238}U$ ratios are consistent with a two-component mixing model, which is used to illustrate the extent of mixing between natural and enriched U end members.

1. Introduction

There is renewed interest in post-detonation glassy fallout formed during surface and near-surface nuclear explosions (Parekh et al., 2006; Eby et al., 2010; Fahey et al., 2010; Belloni et al., 2011; Bellucci and Simonetti, 2012; Cassata et al., 2014; Eppich et al., 2014; Sharp et al., 2014). During the era of above ground nuclear testing, fallout analyses focused on understanding device performance and how radioactivity spread post-detonation. Techniques for analyzing fallout on small spatial scales were generally limited to methods such as autoradiography to image the distributions of fission and activation products (Adams et al., 1960). Today, using modern microanalytical techniques, spatially-resolved analyses of post-detonation fallout from historic nuclear tests can provide much more information on the distribution of stable and long-lived radioactive isotopes with the goal of a more comprehensive understanding of fallout formation, such as improved constraints on formation mechanisms, timescales, and temperatures (Cassata et al., 2014; Eppich et al., 2014).

Glassy fallout forms when a nuclear device is detonated on or near the Earth's surface. Surface detonations melt large masses of environmental material (e.g., soil), which may remain in place or be swept into the nuclear fireball. In both cases, the molten material may interact with device components, fission and activation products, and unfissioned fuel (collectively 'device debris'; Brode, 1968; Glasstone and Dolan, 1977). As the fireball cools below the melting point of the surrounding material, device debris becomes trapped within a fused glassy matrix and rapidly cools, forming glassy fallout. Because the mass of melted environmental material can be much greater than the mass of the device in near surface events, the major element composition of these glasses can predominantly reflect the local geology and/or emplacement environment, and device debris is generally present only in trace quantities (Adams and O'Connor, 1957; Miller, 1964).

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http://dx.doi.org/10.1016/j.jenvrad.2015.04.006
0265-931X/

There are two main morphological classes of glassy fallout, referred to herein as ground glass and aerodynamic glassy fallout. Ground glass usually exhibits a smooth, glassy surface with compositional variations often transitioning to unmelted soil (Eby et al., 2010). This is consistent with either in-situ melting of soil and/or solid material being swept into the fireball and being melted before raining back down onto the soil below (Hermes and Strickfaden, 2005). In contrast, aerodynamic glassy fallout (also known as fallout beads or spherules) usually appears glassy throughout and often exhibits spheroidal, near-spheroidal, or dumbbell shapes, consistent with fusing and quenching while still aloft (Miller, 1960; Crocker et al., 1965; Tompkins et al., 1970, Fig. 1). Aerodynamic glasses often have smaller glassy spheres attached to their surfaces, consistent with growth through the collision and mixing of molten droplets within the fireball (Miller, 1964; Stewart, 1956). For aerodynamic fallout, the cooling timescale depends on device yield, but is estimated (Glasstone and Dolan, 1977; Izrael, 2002) and measured (Cassata et al., 2014) to be on the order of seconds. Few previous studies have focused specifically on aerodynamic fallout glasses, but these objects are of particular interest due to their elevated concentrations of residual fuel and fission and activation products (Mackin et al., 1958; Eppich et al., 2014). How direct device components distribute themselves within aerodynamic fallout remains unaddressed.

Ground glass from the Trinity test, popularly referred to as 'trinitite', has been extensively studied over the past decade (Parekh et al., 2006; Eby et al., 2010; Fahey et al., 2010; Belloni et al., 2011; Bellucci and Simonetti, 2012, 2013; Wallace et al., 2013; Bellucci et al., 2014; Sharp et al., 2014). These studies demonstrate that trinitite is a compositionally heterogenous mixture of completely melted, partially melted, and unmelted minerals found in the local geology, with the glassy and eddied portions of the trinitite containing most of the unfissioned Pu fuel and fission products. Comparisons of trinitite with observations of aerodynamic fallout from Trinity (trinitite spherules), or between aerodynamic and ground glass from other nuclear tests have yet to be reported.

In contrast to recent studies characterizing ground glass from the Pu-fueled Trinity test, this study characterizes aerodynamic fallout glasses from a U-fueled device. Bulk studies (analyses of dissolved entire spherules) of these materials have been recently reported (Eppich et al., 2014). Here, we characterize the spatial distribution of unfissioned U fuel within individual glassy fallout spherules using secondary ion mass spectrometry (SIMS) combined with scanning electron microscopy/energy-dispersive x-ray spectroscopy (SEM/EDS), electron probe microanalysis (EPMA), and autoradiography. For each spherule, we measured the extent of uranium isotopic heterogeneity, major element composition, and radioactivity, and we correlate these observations with spatial locations within individual spherules. The extent of mixing is quantified by assuming a model of two-component mixing between uranium from the soil and/or device with enriched uranium from the device, and then calculating the relative contribution of each end member to each U isotopic measurement. We use our results to discuss the role of different mechanisms in fallout formation.

Table 1
Fallout spherule samples, masses, and average diameters.

Sample	Mass (mg)	Avg. Diameter (mm)
U1A	3.7	1.6
U1B	2.5	1.4
U2	4.5	1.6
U3	14.8	2.3
U4	14.4	2.2

2. Materials and methods

2.1. Fallout spherules

Five millimeter-size fallout spherules were selected from soil collected at the site of a historic, uranium-fueled, near-surface nuclear test. The soil samples were collected about 120 m from ground zero along the path of the fallout plume. Glassy fallout is easily identifiable in soil collections due to its smooth surface morphology and aerodynamic shape (Fig. 1). The spherules were isolated from sieved soils by visual inspection and handpicked under an optical microscope, then were weighed and photographed. Each spherule was mounted in epoxy and polished to expose an interior surface close to the mid-plane of the object. Samples were coated with ~10–20 nm of carbon to prevent charging and then characterized by SEM/EDS, EPMA, autoradiography, and SIMS.

The selected aerodynamic fallout samples range in average diameter from 1.3 to 2.3 mm and in mass from 2.5 to 14.8 mg (Table 1). They appear light to dark green in color and are optically translucent and glossy. Vesicles are visible within the volume of some of the glasses.

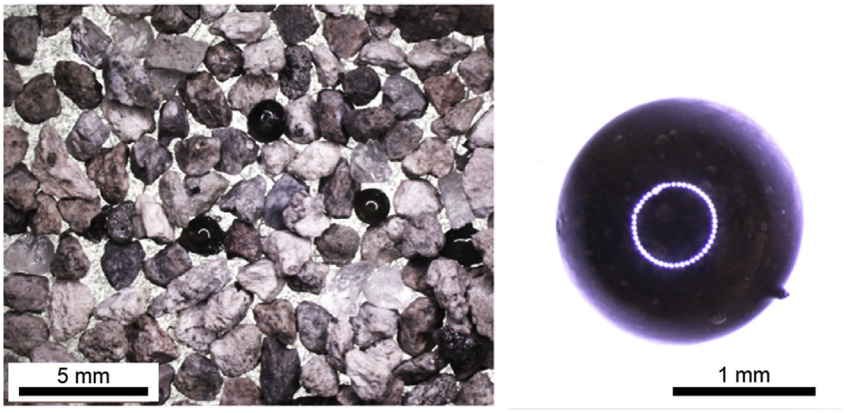

Fig. 1. Optical images of size-sorted fallout (left) showing distinct dark glassy aerodynamic shapes mixed with soil fragments, and an isolated piece of aerodynamic fallout glass (right). The bright artifact in the center of the isolated aerodynamic fallout glass is the ring light from the optical microscope, which also highlights the high degree of symmetry in these objects.