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# Diffusive mass transport in agglomerated glassy fallout from a near-surface nuclear test

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**Abstract**

Aerodynamically-shaped glassy fallout is formed when vapor phase constituents from the nuclear device are incorporated into molten carriers (*i.e.* fallout precursor materials derived from soil or other near-field environmental debris). The effects of speciation and diffusive transport of condensing constituents are not well defined in models of fallout formation. Previously we reported observations of diffuse micrometer scale layers enriched in Na, Fe, Ca, and $^{235}U$, and depleted in Al and Ti, at the interfaces of agglomerated fallout objects. Here, we derive the timescales of uranium mass transport in such fallout as it cools from 2500 K to 1500 K by applying a 1-dimensional planar diffusion model to the observed $^{235}U/^{30}Si$ variation at the interfaces. By modeling the thermal transport between the fireball and the carrier materials, the time of mass transport is calculated to be <0.6 s, <1 s, <2 s, and <3.5 s for fireball yields of 0.1 kt, 1 kt, 10 kt, and 100 kt respectively. Based on the calculated times of mass transport, a maximum temperature of deposition of uranium onto the carrier material of ~2200 K is inferred (1σ uncertainty of ~200 K). We also determine that the occurrence of micrometer scale layers of material enriched in relatively volatile Na-species as well as more refractory Ca-species provides evidence for an oxygen-rich fireball based on the vapor pressure of the two species under oxidizing conditions. These results represent the first application of diffusion-based modeling to derive material transport, thermal environments, and oxidation-speciation in near-surface nuclear detonation environments.


*Keywords:* Nuclear fallout; Nuclear fireball; Oxidation; Speciation; Uranium diffusion; NanoSIMS; Silicate melt

## 1. INTRODUCTION

In near-surface nuclear explosions, aerodynamically-shaped glassy fallout is produced by the mixing of vaporized device (*i.e.* bomb) and environmental materials (*e.g.* structures and dirt) into molten carriers (defined here as fallout precursor materials derived from soil and other environmental debris) in the fireball, which quench before coming in contact with the ground (Glasstone and Dolan, 1977). Vaporized materials can either condense directly onto the surface of molten carriers, or can be deposited as already condensed droplets onto the surface of carriers. The condensed material is subsequently mixed into the molten material by diffusion and mechanical mixing (Miller, 1960; Freiling et al., 1965; Freiling, 1970). The effect of the speciation on condensation and diffusion in fallout materials is poorly understood, but remains fundamental to models predicting the composition and dispersion of radioactive fallout.

Historic fallout formation theories combine physical models with empirical relationships derived from early atmospheric tests (Miller, 1960; Freiling, 1961; Norment, 1979). Refractory species (*e.g.* oxides of Al and Ca) are

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thought to volumetrically incorporate into carrier materials by mechanical mixing, while volatile species deposit onto the surface of the solidifying carriers after the fireball has cooled. These models consider physical principles such as gas-phase diffusion of species in the fireball, condensation onto and diffusion within carriers, as well as re-evaporation of condensing species under the assumption of equilibrium conditions. Such models do not, however, consider heat transfer from the fireball to the carriers, nor the effect of speciation on condensation and diffusion, both of which affect the rates of condensation onto, and diffusion through, the molten carriers. The potential importance of speciation on the condensation and mixing process is underscored by recent work analyzing the uranium isotopic composition of fallout, which found both inter- and intra-sample heterogeneity (Eppich et al., 2014; Lewis et al., 2015; Weisz et al., 2017). In the context of fallout formation models, uranium, if present in an oxide form, may condense first and mix volumetrically into molten carriers due to the refractory nature of uranium oxides (Miller, 1960; Freiling et al., 1965). Stable radiogenic xenon isotope ratios, the decay products of fissiogenic Sn, Sb, Te, measured in fallout glass provide evidence that reducing conditions attend at least some phases of fallout formation (Cassata et al., 2014), although recent findings suggest that multiple oxidation states of U and Fe may be preserved in fallout (Giuli et al., 2010; Pacold et al., 2016). The chemical and isotopic heterogeneity of uranium observed in fallout samples suggests that vapor-phase speciation due to varying redox conditions may be primarily controlled by the amount of air entrained into the fireball, and possibly buffered, to a first order, by the magnitude and speciation of iron present in the vapor phase (Pacold et al., 2016). Clearly, the oxidation state of vapor-phase constituents will impact the condensation and evaporation behaviors in an evolving fireball environment as fallout forms.

The interfaces between fused glassy fallout objects can preserve late stage condensation features, including diffusion profiles (Weisz et al., 2017). In the population of samples studied, these interfaces are characteristically enriched in sodium, iron, calcium, and uranium, and depleted in aluminum and titanium. Similar enrichments of Fe and Ca have been observed as suspected anthropogenic contributions to trinitite glass (Bellucci et al., 2017). The co-location of volatile and refractory elements at the interface implies that after initial condensation, fallout formation continues through a fractionation and diffusion process dictated by the speciation of vapor-phase constituents, and may be affected by contributions from anthropogenic materials (Weisz et al., 2017).

Understanding speciation in the vapor phase is essential to establishing a comprehensive model of fallout formation. In this study, we show that the diffuse interfaces observed between agglomerated fallout objects preserve a record of the thermal histories experienced prior to quenching in the fireball. To demonstrate and explore this record, we apply a model of one-dimensional planar diffusion to explain the Gaussian distribution of uranium concentration observed at select agglomerate interfaces. Subsequently, we estimate the time and temperature over which uranium diffusion occurred from this interface. By constraining the time and temperature of mass transport, the deposition temperature of late stage vapor species onto fallout surfaces prior to agglomeration is also constrained. We then use these insights to interpret the source and speciation of vaporized constituents, and the redox conditions of the fireball.

## 2. MODELING METHODOLOGY AND RESULTS

### 2.1. Data selection

The dataset used for diffusion modeling is derived from agglomerated fallout glasses originating from a uranium-fueled near-surface nuclear test (Weisz et al., 2017), and acquired by nano-scale secondary ion mass spectrometry (NanoSIMS). The sample set consists of mm-scale, aerodynamically-shaped, glassy objects with approximately rhyolitic major element compositions. The objects include numerous sub-mm secondary objects fused to the surface of larger glassy objects (Fig. 1a). Deposition layers at the interfaces of the fused objects were identified using backscatter electron imaging (Fig. 1b). NanoSIMS ion images were collected at these interfaces showing micro-scale compositional relationships and $^{235}U/^{30}Si$ variation (Fig. 1c and d).

Of the nine interfaces presented in Weisz et al. (2017), three of these yielded approximately Gaussian distributions of $^{235}U/^{30}Si$ profiles. In the case of a thin layer of condensed species deposited onto the surface of a molten carrier that subsequently fuses with a larger molten object, the deposited layer is expected to diffuse into both objects if there is a chemical potential gradient between the layer and the molten objects. In the idealized case, that the layer comprises a single species, is infinitesimally thin, and is between two homogeneous objects of equal composition. The concentration distribution will evolve from the layer between two objects of equal temperature over time, following the Gaussian form of

$$C(x,t) = C_0 e^{\frac{-x^2}{4Dt}} \quad (1)$$

where $C$ is concentration, $C_0$ is the initial concentration of the species in the layer, $D$ is the diffusivity, $t$ is time, and $x$ is the dimension of diffusion (Zhang, 2010). Based on these observations, three approximately Gaussian $^{235}U/^{30}Si$ profiles B1, C1, and D2 were chosen as the focus of the diffusive modeling presented, here.

The experimentally determined $^{235}U/^{30}Si$ ratio profiles at the three chosen interfaces were fit in IGOR Pro software using non-linear least squares regression analysis with a modified Gaussian function:

$$G = A + Bx + Cx^2 + \frac{E}{\sigma\sqrt{2\pi}} e^{\frac{-(x-\mu)^2}{2\sigma^2}} \quad (2)$$

where $\sigma$ is the standard deviation of the distribution, $\mu$ is the mean of the distribution, $E$ is a fit scaling parameter, and $A + Bx + Cx^2$ is a quadratic expression (with fitting coefficients $A$, $B$, and $C$) added to allow for minor compositional variation on either side of the interface and still acquire a Gaussian fit of the peak. The compositional data