



A detailed geochemical investigation of post-nuclear detonation trinitite glass at high spatial resolution: Delineating anthropogenic vs. natural components

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ABSTRACT

This study documents, for the first time, the combined abundances of major and trace elements at high spatial resolution for trinitite glass, which was produced during the first atomic weapon test (Trinity site; New Mexico, USA). The results indicate that the chemical composition of trinitite is largely dependent on the precursor mineral phases found in the arkosic sand at the Trinity site. The chemical compositions of trinitite were evaluated using principal component analysis, which indicates that trends may be attributed to mixing between an anthropogenic component and phases (both major and minor) within the arkosic sand. The resolvable anthropogenic component in trinitite is made up of metals including Al, Co, Cr, Cu, Fe, Ga, Mg, Mn, Nb, Pb, Ta, and Ti. Uranium in trinitite appears to have two sources: natural U-bearing phases and the tamper used in the device. The concentrations of volatile anthropogenic metals (Co, Cr, Cu, and Pb) are enriched in samples that originate from >74 m away from ground zero. This increase may be related to a temperature-controlled fractionation, and implies that the (lower temperature) peripheral zone of a blast site is the optimal area to sample volatile metals for nuclear forensic analysis.

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1. Introduction

Nuclear proliferation is, arguably, the current greatest threat to modern civilization. An increasing number of countries are not participating in negotiations to cease the development of nuclear technologies or reduce the total number of available nuclear weapons. While it is unlikely that a traditional state would issue a nuclear strike, a significant nuclear threat exists from non-state actors or terrorist groups. The increase of undocumented/unaccounted for nuclear weapons and material enhances the likelihood of a rogue group acquiring and detonating a non-traditional nuclear device in an act of terrorism. Distinguishing the chemical composition (or signature) of a nuclear device, in terms of fuel type, fission products, and device components, is critical in order to determine the region of origin of that device. Identifying the provenance of nuclear materials is necessary for accurate source attribution. As such, chemical and isotopic characterizations of post-detonation materials are foremost goals in nuclear forensics.

Historic test sites offer a place to study post-detonation products and establish protocols that will yield rapid, accurate, and precise results. Test sites are ideal for establishing nuclear forensics protocols because the compositions of weapons employed are relatively well documented; therefore, these provide a means to verify any results gained from these

investigations. “Trinitite,” post-detonation materials from the first atomic weapon test conducted at the Trinity site, are available for public research and have been the focus of several previous studies (e.g., Schlauf et al., 1997; Parekh et al., 2006; Semkow et al., 2006; Eby et al., 2010; Fahey et al., 2010; Belloni et al., 2011; Bellucci and Simonetti, 2012; Bellucci et al., 2013a,b,c; Wallace et al., 2013). Therefore, trinitite samples provide ideal materials to establish post-detonation nuclear forensics techniques.

The Trinity test took place at 5:29:45 a.m. on July 16, 1945 at the White Sands Proving Grounds just south of Alamogordo, NM. The Trinity device, Gadget, was an implosion-type ^{239}Pu device. Prior to detonation, Gadget was elevated to a height of 30.5 m upon a steel tower. The 21 kt explosion yielded a mushroom cloud with a height of 15.2 to 21.3 km and a temperature of ~8430 K (Eby et al., 2010). This fireball engulfed the test site, steel bomb tower, bomb components, the surrounding desert sand, and upon cooling formed a glassy layer named “trinitite”. The trinitite layer extended radially to 370 m away from ground zero (Storms, 1965). A two-step model has been proposed for the formation of trinitite: 1) production of molten glass both on the ground and in the mushroom cloud, and 2) subsequent to incorporation of solid material (non-molten mineral phases, metal, and droplets) raining down from the cloud on the upper surface of this solidifying glass (Belloni et al., 2011; Bellucci and Simonetti, 2012; Wallace et al., 2013). Trinitite formed extremely rapidly and did not have time for any physical or chemical equilibrium processes. As such, there are many different morphological types of trinitite (e.g., fallout beads,

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dumbbell trinitite, red inclusions, black inclusions, white inclusions, and coke-bottle colored inclusions; Table 1, Eby et al., 2010). While each variety of trinitite contains unique forensic evidence, this study is focused on characterizing “green” trinitite, which is most common.

By examining “green” trinitite, the main purpose of this study is to help establish a geochemical point of reference for future studies on the remaining morphological types of trinitite. This goal will be accomplished by acquiring spatially resolved major and trace element analyses of “green” trinitite; this information will then be used to resolve the components derived from the Gadget and site materials (e.g., the blast tower) versus those from the surrounding geology (i.e., natural background). Of particular note, the major and trace element compositions of trinitite will yield useful information on the distribution and incorporation of device components within trinitite; however this information may not yield direct clues as to the nature of the fuel and radionuclides involved with the Trinity test. Hence, this study was performed in tandem with an investigation into the isotopic composition of radionuclides (e.g., Cs, Sr, Eu, Pu) within trinitite glass (Wallace et al., 2013).

1.1. Components from the blast tower and device

The core of Gadget was constructed of concentric shells, with a 2.5 cm diameter Be neutron initiator in the center, followed by a 9.2 cm diameter “super grade” Pu–Ga alloy core, a 22 cm diameter tamper constructed from natural U, and finally a 22.9 cm diameter boron-plastic shell (Rhodes, 1986). The core consisted of ‘super-grade’ Pu with a $^{240}\text{Pu}/^{239}\text{Pu}$ of .0128–0.016 (Parekh et al., 2006; Fahey et al., 2010). Surrounding the core of the device was the implosion assembly, which consisted of three concentric circles of explosives and aluminum shells (Rhodes, 1986). The explosives used in the device were of RDX, TNT, and Baratol, which is a mixture of TNT and $\text{Ba}(\text{NO}_3)_2$ (Rhodes, 1986). Several studies have documented components in trinitite likely originating from Gadget, including Cu from the wiring used in the device or monitoring equipment (Eby et al., 2010; Bellucci and Simonetti, 2012), Pb with ambiguous origins (Eby et al., 2010; Fahey et al., 2010; Bellucci and Simonetti, 2012; Bellucci et al., 2013c), and W–Ta–Ga alloy, most probably a piece of the tamper or electronics (Bellucci and Simonetti, 2012). Iron and Fe–Ti inclusions have also been observed in

trinitite and have been interpreted to derive from the blast tower (Eby et al., 2010; Fahey et al., 2010; Bellucci and Simonetti, 2012).

1.2. Components from sand

The dominant surface geology at the White Sands Missile Range is arkosic sand with alluvial, aeolian, evaporitic, and volcanic components (Ross, 1948; Eby et al., 2010). Arkosic sand consists predominantly of quartz (SiO_2), K-feldspar (KAlSi_3O_8), and may contain minor amounts of carbonates (CaCO_3), sulfates (BaSO_4 , $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), chlorides (NaCl), and detrital zircon (ZrSiO_4); other U-bearing trace mineral phases include monazite (REE, Th, PO_4) and apatite ($\text{Ca}_5(\text{PO}_4)_3$ (F, Cl, OH)). Also present are clay and mafic minerals, specifically hornblende ($\text{Ca}_2(\text{Mg}, \text{Fe}, \text{Al})_5(\text{Al}, \text{Si})_8\text{O}_{22}(\text{OH})_2$), olivine ($(\text{Fe}, \text{Mg})_2\text{SiO}_4$), magnetite (Fe_3O_4), ilmenite (FeTiO_3), and augite ($(\text{Ca}, \text{Na})(\text{Mg}, \text{Fe}, \text{Al}, \text{Ti})(\text{Si}, \text{Al})_2\text{O}_6$); (Ross, 1948; Staritzky, 1950; Pettijohn, 1963; Love et al., 2008; Eby et al., 2010; Fahey et al., 2010). Knowledge of the pre-detonation geology is of critical importance in order to resolve the addition of chemical components originating from the bomb and those from the surrounding blast site; detection and identification of these components is a foremost goal of a nuclear forensics study. Metals abundant in both the device and blast tower, such as Fe and U are typically present at low abundances in arkosic sandstone (e.g., <1 wt.% $\text{FeO}_{\text{total}}$; Pettijohn, 1963). Additionally, Pu and related activation/fission products would have been non-existent in a natural environment at the time of the test. Hypothetically, these elements will provide the best avenue for discerning components from the test site, Gadget, and surrounding geology (e.g., Wallace et al., 2013).

Of utmost importance, as shown here in Figs. 1 and 2 and in previous investigations (Eby et al., 2010; Fahey et al., 2010; Wallace et al., 2013), trinitite is extremely heterogeneous (petrographically and chemically) at the micron scale. Therefore, a variety of micro-analytical techniques are necessary to effectively evaluate the compositional and isotopic distributions in trinitite and potentially resolve any device components/mixing end-members (e.g., Hainley et al., 2012; Koeman et al., 2012). For example, the recent study by Wallace et al. (2013) reports combined alpha and beta radiography results (maps) in conjunction with scanning electron microscopy (SEM)–back scatter electron (BSE) imaging; the former indicate areas within trinitite that are abundant in alpha (U, Pu) and beta emitter (Cs) radionuclides. Hence, this combined, multiple imaging and LA–(MC)–ICP–MS analysis methodology results in an efficient manner for documenting isotopic variations of radionuclides at high spatial resolution (10s of micron scale; Bellucci et al., 2013b,c; Wallace et al., 2013).

The major element chemical maps indicate that trinitite is composed of relict quartz and feldspar grains and a heterogeneous melt glass (Figs. 1, 2), and supports the earlier observations by Eby et al. (2010). This study is explicitly focused on identifying and characterizing the composition of chemical components, other than quartz, that were incorporated into the melt glass. Subsequently, mixing end-members are determined using traditional geochemical techniques and the statistical multivariate analytical technique, principal component analysis (PCA). Electron microprobe analysis (EMPA) and laser ablation–ICP–MS techniques provide the means to quantitatively determine the abundances of both major (concentrations of >1 wt.%) and trace elements, respectively, in situ with spatial resolution on the scale of 10s of microns. Utilizing these techniques, the first detailed spatially resolved geochemical investigation of trinitite melt glass is reported here.

2. Samples

All of the samples of trinitite analyzed here ($n = 13$) were purchased from Mineralogical Research Corporation (www.minresco.com). The samples have wide range of morphologies but generally consist of trinitite melt glass on the top surface (i.e., exposed to blast) and transition into un-melted desert sand towards the bottom side

Table 1
Samples, inclusion information, and morphological descriptions.

Sample #	Notes
1	Samples with glass-like fused top surface. This is the most common type of surface feature seen on the trinitite specimens.
2	Specimens with large gas pockets in the surface or around perimeter.
3	The bottom of each sample in this group exhibits the rough texture of the sandy desert surface, which remained untouched by the blast. The tops of the specimens have lumpy, convoluted, sand included, or otherwise irregular surfaces.
4A	These samples contain red inclusions. The red inclusions are due to the presence of copper. Copper is not native to the mineralogy of the blast area. Therefore, the copper is thought to have derived from the copper wiring used in the instrumentation on the project.
4B	These samples contain black inclusions. Black inclusions are thought to be remnants of blast tower.
4C	These samples contain light colored glass of light colored flow marks.
4D	These samples contain blue inclusions. Blue inclusions are small, glassy, white to very light robin's egg blue color. These inclusions are extremely rare. No explanation in literature.
4E	These samples contain white inclusions. White inclusions consist of small masses of the partially to totally fused white feldspar or quartz.
4F	These samples contain “Coke bottle green” color inclusions.
5A	Layered specimens have lighter colored trinitite glass bases, usually with darker colored surfaces.
5B	Unusual protuberances or casts on the bases of these specimens.
5D	Elongated, finger-like extrusions. Always found in small pieces. Very delicate and rare because they are fragile and easily broken.
5E	Lace-like specimens, always small, very delicate.
5F	Specimens with possible unfused, microscopic iron beads.

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