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3D chemical segmentation of fly ash particles with X-ray computed tomography and electron probe microanalysis

Qinang Hu [a], M. Tyler Ley [a,*], Jeffrey Davis [b], Jay C. Hanan [c], Robert Frazier [d], Yanli Zhang [c]

[a] *Oklahoma State University, Department of Civil and Environmental Engineering, Stillwater, OK, USA*
[b] *National Institute of Standards and Technology, Microanalysis Research Group, Gaithersburg, MD, USA*
[c] *Oklahoma State University, Department of Mechanical and Aerospace Engineering, Stillwater, OK, USA*
[d] *B&T Engineering, Tulsa, OK, USA*

HIGHLIGHTS

- The technique showed consistent performance on three different fly ash particles.
- The technique found two constituents that made up over 50% of the particles.
- The segmentation values for these constituents were similar between particles.
- Three dimensional constituent models were created for the fly ash particles.
- The particles were not uniform in their chemistry with depth.

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ABSTRACT

A novel data fusion technique is presented that combines X-ray computed tomography (CT) and electron probe microanalysis to investigate fly ash particles. The technique is called Tomography Assisted Chemical Correlation (TACCo). This technique fuses 2D compositional data with 3D X-ray CT data. The method produces 3D constituent and microstructure maps. Results and observations are presented that show the power of the technique for data visualization as well as quantitative analysis.

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1. Introduction

Fly ash is a waste product gathered from the emissions of coal fired power plants. It is estimated that the worldwide production is over 600 million metric tons [1]. Currently only 40% of this material is used in other process and the other material is placed in special landfills.

Fly ash is made up of crystalline and amorphous phases containing predominantly Al, Si, Ca and Fe [2–5]. The particles are generally spherical with diameters ranging from less than 1 μm to more than 1 mm [1,2,5]. Trace deposits of metals such as arsenic, cesium, lead, selenium, cadmium, and zirconium have been found within these materials [6–8]. Fly ash is commonly used as a low cost construction binder for stabilization of soil, a partial replacement of portland cement in a concrete mixture, and the predominate binder in a geopolymer concrete [1,9].

Despite the widespread use of this material, there are still a number of important unanswered questions. Some of these include the concern that heavy metals can be leached from the fly ash in solutions. Past research has also shown that fly ash in concrete mixtures has the ability to improve the strength, reduce the permeability, improve the rheology of fresh concrete mixtures, and provide resistance to a number of common durability problems. Fly ash can either act as a supplementary binder or through a secondary pozzolanic reaction with the alkaline pore solution and calcium hydroxide depending on the properties of the fly ash. Substitution levels for portland cement in a concrete mixture are commonly limited to 20% by mass. This limit is imposed because fly ash is not a manufactured material and therefore not all sources have the same performance in concrete. If this material was better understood then the usage levels could be increased.

* Corresponding author. Tel.: +1 405 744 5257.
E-mail address: tyler.ley@okstate.edu (M.T. Ley).

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This paper presents a new analytical technique that uses a combination of electron probe microanalysis (EPMA) and micro X-ray computed tomography (μCT) to produce 3D maps of the microstructure and distribution of chemical constituents within complex particles. This methodology has been named **T**omography **A**ssisted **C**hemical **Co**rrelation or **TACCo** by the authors. This technique has the ability to identify different constituent phases and map their 3D location. While the work focuses on applying this technique to fly ash, TACCo applies to any material that can be investigated with both X-ray tomography and a scanning electron beam.

High flux electron beams are commonly used for elemental analysis of materials. An incident electron beam with sufficient energy will cause an atom to emit a characteristic photon through the photoelectric effect [10]. This information can be used to create detailed compositional maps of the surface of materials through the use of energy dispersive X-ray spectrometers [10–13]. To obtain reliable data with the technique a sample must be polished, conductive (or coated in conductive carbon), and analyzed in a high vacuum. While this technique, known as EPMA, is widely used, it has several challenges as it can damage the sample at high probe currents (i.e. >100 nA), only allows observations of the composition in the first few micrometers to be made, and must be examined in a high vacuum environment [10,11].

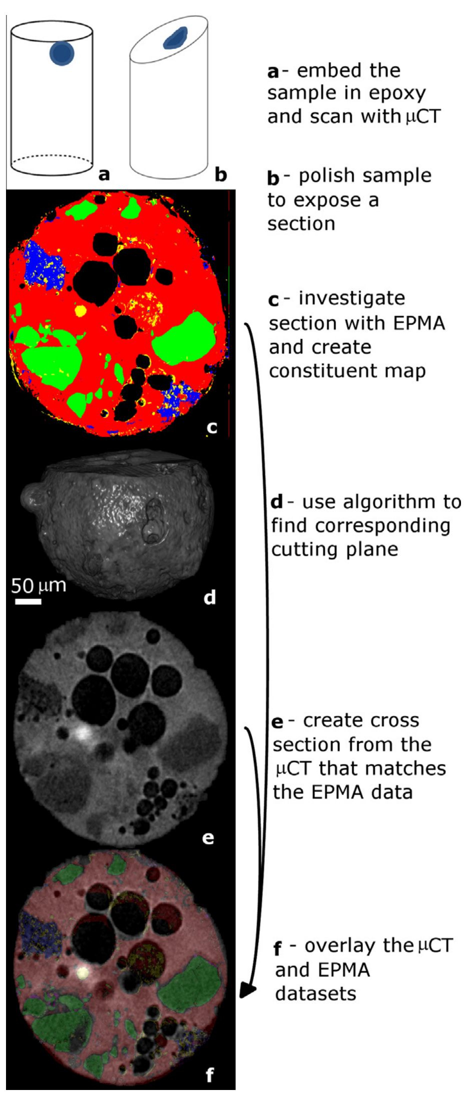

Fig. 1. An overview of the processes used for fusing the EPMA and μCT data sets.

It is common in the medical sciences to use X-ray computed tomography (CT) to non-destructively image the internal structure of organisms. This technique uses a series of X-ray radiographs at small angles of rotation that are coupled to produce a 3D model [14–16]. While this technique does not provide direct compositional information about the irradiated materials it does provide clues about compositional consistency. In the CT scan, solid materials with different mass absorption coefficients and densities will appear to have different gray values. However this data does not provide *a priori* compositional information.

A new generation of tomography techniques is being used that allows the user to enter any domain and find the spatial resolution of unique chemical constituents by combining μCT with information from X-ray Fluorescence. These investigations have been found to require days to gain the data that is needed [17–19]. Furthermore, this technique is limited to 3D models at roughly 30 μm. The required investigation time for the TACCo analysis can be on the order of hours and produces 3D maps at the resolution size of the parent μCT and EPMA with the current best resolution at 10 s of nm [20,21]. Furthermore, once the data has been fused between the μCT and EPMA, then one can continue to only use the μCT for 3D constituent information.

The ability to characterize fly ash at an intimate scale provides new tools to understand, extend, and improve its usage. Specific needs to characterize the elemental makeup, the reactivity, and the location of these constituents within a particle will improve the use. This can have significant impacts on the economy, durability, strength, rheology, and sustainability of modern concretes. The majority of previous work has focused on the measurement of bulk properties of fly ash [22–25]. A few others have tried to characterize fly ash in more depth [5,23,26,27].

These needs to characterize complex powders are not just common to the investigation of fly ash but are common to the study of catalysts, pharmaceuticals, ceramics, contaminated soils, coal, zeolites, asteroids, geologic materials, biomaterials, and others. While these materials have been investigated by either μCT or EPMA no previous publication has successfully fused this data. The presented methods could be useful for any material that can be analyzed by both μCT and EPMA.

2. Methods

A graphical summary of the methods used is shown in Fig. 1.

2.1. Sample description and preparation

This paper investigates three fly ash particles from a single source. The fly ash is classified as ASTM C 618 class C. The results from the bulk XRF analysis is given in Table 1. Particles with diameters larger than 200 μm were used, as they were easy to handle and provided a sufficient area to provide a large compositional and μCT data set. The methods described can be used for any size particle as long as the analytical technique provides adequate resolution. Work on smaller particles will be included in future publications.

A plastic cylinder mold that is open on the top and bottom is filled with a low viscosity epoxy. The epoxy was chosen as it is easy to polish, has a low X-ray attenuation coefficient, and shows minimal outgassing in a vacuum. Before the epoxy hardened, a fly ash particle was placed on the surface of the freshly mixed epoxy and allowed to sink to the bottom. The specimen was then left for a day to harden. A drawing of the specimen is shown in Fig. 1a.