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Insights into the structural, microstructural and physical properties of multiphase powder mixtures

Ruishi Xie [a, *], Yuanli Li [b], Haifeng Liu [a], Xingquan Zhang [a]

[a] Analytical and Testing Center, Southwest University of Science and Technology, Mianyang 621010, China
[b] Department of Materials, Southwest University of Science and Technology, Mianyang 621010, China

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ABSTRACT

In recent years, research interests on structural and microstructural properties of materials have gained enormous impetus owing to their absolute necessity in many fields (*e.g.,* chemistry, physics, synthetic and materials science). Herein, we present an insight into comprehensive ascertainment of the general structural, microstructural and physical properties of multiphase powder mixtures. We systematically explored the properties of the multiphase samples to disclose their structure and chemical composition features using a series of techniques, such as X-ray diffraction, and energy dispersive X-ray spectroscopy. The Rietveld approach based on structure and microstructure refinement analysis was adopted for precise determination of several microstructural parameters as well as phase contents of individual phases in the multiphase materials. The X-ray diffraction results confirms that the samples are multiphase and crystallize in the monoclinic baddeleyite structure with $P\,1\,21/c\,1$ space group, orthorhombic mullite structure with $P\,b\,a\,m$ space group, and cubic molybdenum structure with $I\,m\,{-}3\,m$ space group, respectively. The morphology features of the samples were observed by using scanning electron microscopy. The actual elemental distributions can be easily observed in energy dispersive X-ray spectrometry elemental mappings. To shed more light on the properties of the multiphase powder mixtures, their physical features (*e.g.,* dislocation density, unit cell volume, X-ray density, porosity) were researched in detail. Moreover, the residual stresses of the multiphase samples were investigated by utilizing the $\sin^2\psi$ approach.

1. Introduction

In recent years, with the development of the economy, a large amount of solid wastes has been come into being by industrial production all over the world. These industrial wastes cause serious environmental pollution and wasted resources, which has become an important issues in many countries [1,2]. Waste utilization is a fascinating disposal approach which can cut down or even eliminate disposal costs and potential pollution issues while aggrandizing resource protection [3,4].

Among the solid industrial wastes, Mining industry tailings are produced after beneficiation. Almost all the tailings, which includes copper, gold, iron, bauxite and kaolin tailings [5–9], are created from the mineral mining. With mineral resources dwindling and the demands of environmental conservation growing, tailing recycling has been attracted the attention of countries around the world [10–13]. If mining industry tailings can be reasonably handled, it will not only reduce the exploitation of non-renewable mineral resources, but also safeguard the environment as well as stimulate the development of the ecology. Therefore, for the more rational dispose of mining industry tailings, it is urgent to ascertain the structural and microstructural properties of the multiphase powder mixtures in the tailings. To this end, considerable efforts, experimental as well as theoretical, have been taken into this field; however, systematic investigations remain limited.

On the other hand, the first step toward comprehending a solid material is to know its atomic structure. In the circumstance of materials with long-range order this signifies to ascertain the crystal structure [14,15]. Usually, there are two circumstances to discriminate. In the first case, single crystals of high quality are acquired by pure opportunity or experimental ability, but not by mining industry tailings. In this circumstance the single crystal is tested on a single-crystal diffractometer and structure elucidation is accomplished by one of the many utilizable approaches such as

* Corresponding author.
E-mail address: rxie@foxmail.com (R. Xie).

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direct approach, Patterson search, or more recently, charge flipping [16–18]. In the second circumstance, single crystals of sufficient quality and size are not available. In this circumstance it is essential to optimize the fabrication and/or purification approaches until single-phase powders are acquired. Apparently, this claims numerous skills on the part of the experimentalist and representatively a great amount of time.

Frequently neither single crystals nor single-phase powders are utilizable. As a matter of fact, in many realistic world circumstances mixtures are nearly prevalent. To authenticate the crystal structures from mixtures of unknown components is usually regarded as a troublesome issue [19]. However, this is the situation scientists in widely different realms such as materials science, pharmaceutical fabrication or mineralogy frequently experience. A few approaches to discriminate phases have been recommended for very special situations of mixture analysis where line broadening or preferred orientation take place, but a universal approach that can afford individual structures from mixtures would be a significant breakthrough in these fields.

Physical properties of the materials depend significantly on their structural and microstructural parameters, such as size, shape and crystalline structure [20–22]. Therefore, X-ray analysis technique based on structure and microstructure refinement is usually preferred for accurate measurement of these lattice imperfections in order to acquire materials with desirable properties by choosing the microstructural features accordingly.

In this work, we implement a comprehensive investigation of the general structural, microstructural and physical properties of multiphase powder mixtures. We systematically investigated the properties of the multiphase samples to disclose their structure, chemical composition, and morphology features using a series of techniques, such as X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy dispersive X-ray (EDX) spectroscopy. The Rietveld method based on structure and microstructure refinement analysis was adopted in the present study for precise determination of several microstructural parameters (*e.g.*, lattice parameters, atomic positions, occupancy, site occupancy factor, bond distances and bond angles) as well as phase contents of individual phases in the multiphase materials. Rietveld method was found to be the best indirect approach for microstructure characterization of the multiphase materials containing significant number of overlapping reflections. The X-ray diffraction pattern refined using Rietveld method confirms that the samples are multiphase and crystallize in the monoclinic baddeleyite structure with $P\,1\,21/c\,1$ space group, orthorhombic mullite structure with $P\,b\,a\,m$ space group, and cubic molybdenum structure with $I\,m\,{-}3\,m$ space group, respectively. The morphology of the samples, observed using scanning electron microscopy, reveals the existence of quasi-ellipsoids constituted by a great quantity of tiny particles, smooth aggregates composed by some quasi-spheres, and irregular and angular clumps. The actual elemental distributions can be easily observed in energy dispersive X-ray spectrometry elemental mappings. To shed more light on the properties of the multiphase materials, their physical features (*e.g.,* crystallite size, lattice strain, dislocation density, unit cell volume, X-ray density, porosity) were explored in detail. Moreover, the residual stresses of the multiphase samples were investigated by utilizing the $\sin^2\psi$ approach.

2. Experimental

All the multiphase powder mixtures investigated in this work were obtained from a mining company. All samples were characterized by X-ray diffraction (XRD) with a PANalytical X'Pert PRO X-ray diffractometer. Cu K_α radiation was used (λ = 1.54187 Å), operated at 40 kV and 40 mA, with an X'Celerator ultra-fast detector based on real time multiple strip technology with Bragg-Brentano geometry. The scans were performed in the 2θ range from 10 to 90° with a step scan of 0.016° and 60 s per step in a continuous mode. Structural refinements by Rietveld method and crystalline phase identification were performed with X'Pert HighScore Plus software, and an ICSD PDF-2 plus database (ICSD, The Inorganic Crystal Structure Database, FIZ Karlsruhe). The crystallite size of all samples was estimated by the Williamson-Hall method. The scanning electron microscopy (SEM) images, for the determination of the morphology of the samples, were taken by a Carl Zeiss Ultra 55 field emission scanning electron microscope operating at an accelerating voltage of 15 kV. The elementary composition analysis of the samples was performed using an Oxford IE450X-Max80 energy-dispersive X-ray (EDX) spectrometer, an accessory of field emission scanning electron microscope. All the measurements were performed at room temperature and under ambient conditions.

3. Results and discussion

To shed light upon the structural informations of the multiphase materials, X-ray diffraction (XRD) was implemented. Fig. 1 shows the XRD pattern of the multiphase powder mixtures. The vertical bars indicate peak positions from the powder diffraction standard cards of (a) 98-007-1839, (b) 98-009-7319 and (c) 98-008-0125, respectively. All diffraction peaks can be indexed to the monoclinic phase of baddeleyite (m-ZrO_2) with $P\,1\,21/c\,1$ (No. 14) space group, cubic phase of molybdenum (α-Mo) with $I\,m\,{-}3\,m$ (No. 229) space group, and orthorhombic phase of mullite (o-$Al_{4.68}Si_{1.32}O_{9.66}$) with $P\,b\,a\,m$ (No. 55) space group, and which are in consistent with the ICSD PDF No. 98-007-1839, 98-009-7319 and 98-008-0125, respectively. No traces of additional peaks are observed in the XRD patterns, which confirm the multiphase sample is made of baddeleyite, molybdenum and mullite.

The confirmation of a crystal structure may be deemed complete only when multiple-pattern variants and crystallographic parameters of a model have been totally refined against the observed powder diffraction data. Evidently, the refined model should remain reasonable from both physical and chemical perspectives. The refinement technique, most commonly utilized today, is based on the concept proposed in the middle 1960s by Rietveld. The essence of Rietveld's approach is that experimental powder diffraction data are adopted without segregation of the individual

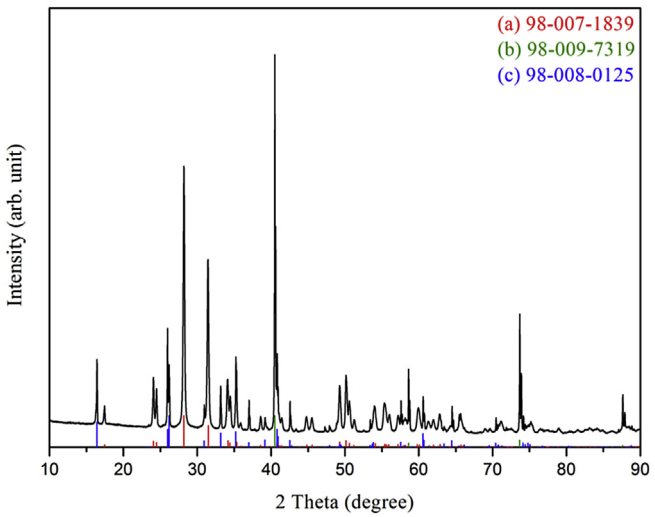

Fig. 1. XRD pattern of the multiphase samples.