

## Preparation and characterization of cerium-gadolinium monazites as ceramics for the conditioning of minor actinides



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### ABSTRACT

Monazites are of considerable interest for immobilization of minor actinides. In this work, the  $Gd_{1-x}Ce_xPO_4$  ( $0 \leq x \leq 1$ ) series is synthesized via conventional solid-state reaction using Ce as the surrogate for minor actinide americium (Am), with microstructure/phase structure characterized by XRD, HRTEM,  $\mu$ -Raman, and SEM as well. The optimized temperature for preparation of monazite-type  $Gd_{0.4}Ce_{0.6}PO_4$  solid solution is more than 1300 °C. At 1400 °C, the  $GdPO_4$  ceramic is essentially monazite with little metastable xenotime phase. The metastable xenotime ( $GdPO_4$ ) to monazite transformation occurs during the immobilizing phase of Ce element. The formation of monazite-type  $Gd_{1-x}Ce_xPO_4$  solid solutions in the range  $0.1 \leq x \leq 1$  is confirmed by the Rietveld refinement and Raman spectra. The change of Ce content has no significant influence on the morphology of the monazite-type  $Gd_{1-x}Ce_xPO_4$  compounds.

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### 1. Introduction

The immobilization of minor actinides, such as Am, Np, Cm, has been a major concern in nuclear industry due to their long-term radiotoxic contribution to the high-level waste (HLW). In this regard, progressed efforts have been made to immobilize these minor actinides in a specific host matrix (Crum et al., 2012; Donald et al., 1997; Lee et al., 2006; Sengupta, 2012). Concerning the long-term safety of geological disposal, the waste form should possess phase stability and high leach resistance. Phosphate-based ceramics such as monazite, which has excellent physical and chemical properties, are considered as suitable host materials for immobilization of minor actinides (Arinicheva et al., 2014; Boatner, 2002; Bregiroux et al., 2007b; Kowalski et al., 2015; Oelkers and Montel, 2008; Pratheep Kumar and Gopal, 2015; Schlenz et al., 2013; Terra et al., 2006; Zhang and Vance, 2008).

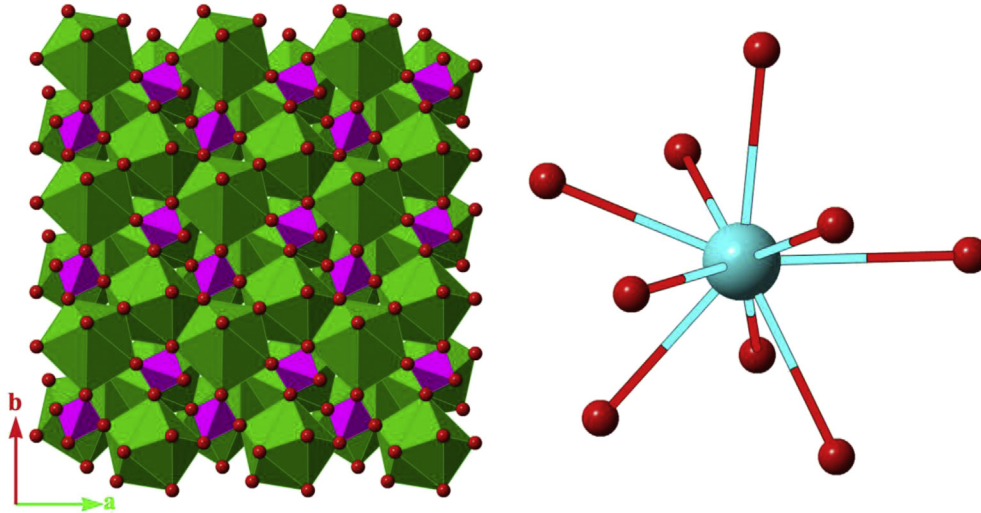
Monazite with a general formula  $REPO_4$  ( $RE = La$  to  $Gd$ ) is a naturally orthophosphate considered as one of the most water-resistant minerals. The crystal structures of monazite (space

group P21/n with  $Z = 4$ ) consist of chains of alternating  $PO_4$  tetrahedra and  $REO_9$  polyhedra (Beall et al., 1981; Mullica et al., 1985; Ni et al., 1995). Fig. 1 shows the crystal structure of synthetic  $GdPO_4$  as an example (Ni et al., 1995). Due to irregular coordination of oxygen atoms around RE and the existence of the distorted  $PO_4$  tetrahedra, the monazite structure exhibits considerable flexibility to accommodate ions differing in the size and charge. As a result, a considerable body of experimental work has focused on the monazite structure-types for immobilizing the actinides (Pu, Th, U) (Bregiroux et al., 2007b; Kowalski et al., 2015; Pratheep Kumar and Gopal, 2015; Schlenz et al., 2013; Zhang and Vance, 2008). On this basis, the  $Pu^{3+}$  and  $Pu^{4+}$  incorporation and the partial reduction of Pu(IV) in Pu(III) in monazite-type compounds are mentioned (Bregiroux et al., 2007b).

Cerium (Ce) is frequently used to simulate minor actinide americium (Am) owing to its similar charge, size and chemical property (Krauskopf, 1986; Schlenz et al., 2013; Shannon, 1976). However, there are few experimental studies concerning the immobilization of  $Am^{3+}$  simulated by  $Ce^{3+}$  in the monazite structures. Moreover,  $GdPO_4$  has dimorphism and can form both monazite and metastable xenotime structures (Clavier et al., 2011; Hay et al., 2014, 2013bib\_Hay\_et\_al\_2013; Ushakov et al., 2001). The  $Am^{3+}$  incorporation in the  $GdPO_4$  matrix has not been illustrated systematically. This may limit their further application for

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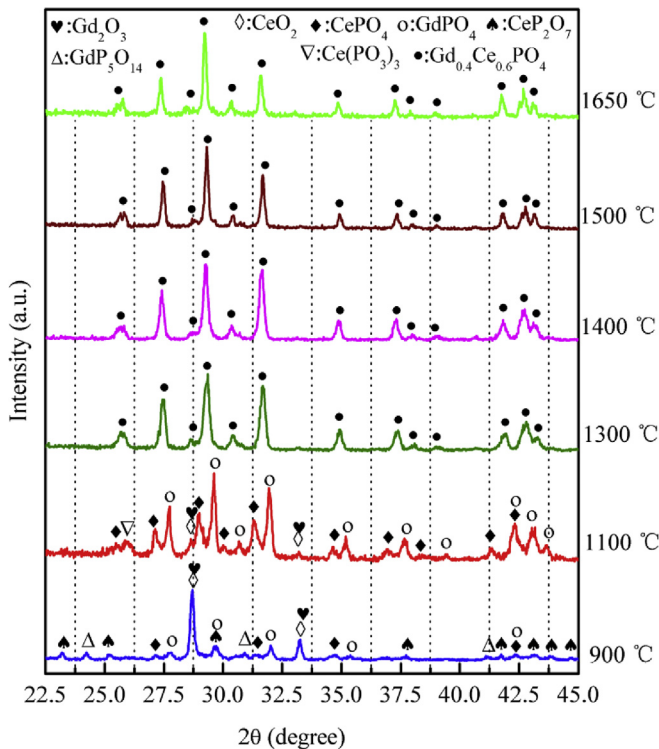
**Fig. 1.** ( $3 \times 3 \times 3$ ) cell representation of  $\text{GdPO}_4$  (left) and nine-fold coordination of gadolinium in  $\text{GdPO}_4$  (right). Viewing direction parallel [001] ([ $\text{GdO}_9$ ] – medium green, [ $\text{PO}_4$ ] – magenta, O – red, Gd – cyan). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

geological disposal of nuclear waste. To close the gap, in this paper, the  $\text{Gd}_{1-x}\text{Ce}_x\text{PO}_4$  ( $0 \leq x \leq 1$ ) ceramics are prepared by conventional solid-state reaction using Ce as the surrogate for Am. The effect of the Ce content and sintering temperature on the structure and microstructure of the  $\text{Gd}_{1-x}\text{Ce}_x\text{PO}_4$  system are investigated. The data of structural properties (linear fit of unit cell parameters and unit volumes) provided in this study can offer important information desired for their practical application.

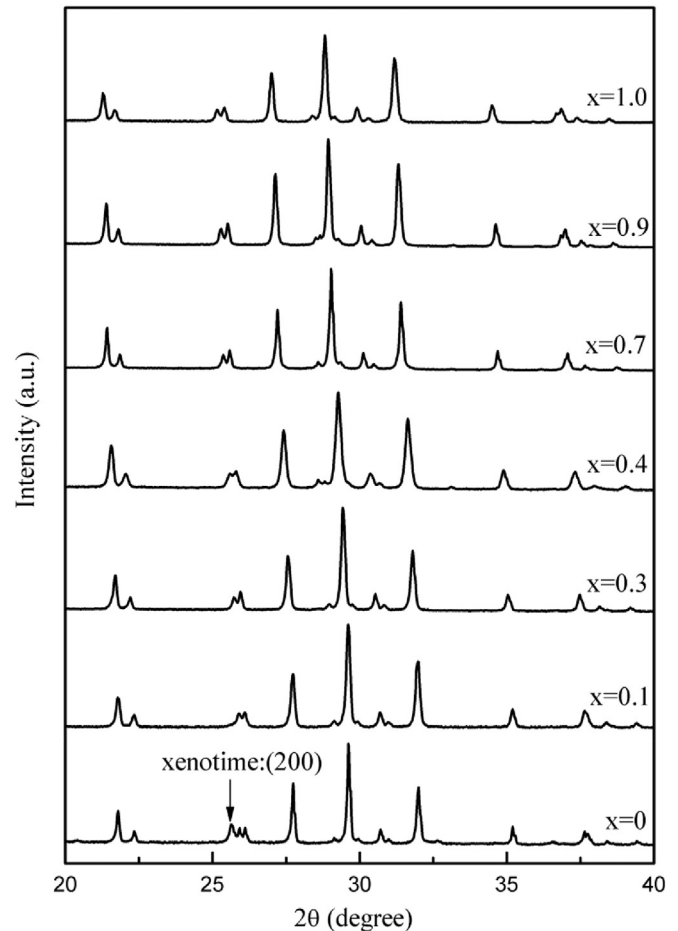
## 2. Experimental section

### 2.1. Preparation of samples

$\text{Gd}_{1-x}\text{Ce}_x\text{PO}_4$  ( $0 \leq x \leq 1$ ) compounds are prepared by a



**Fig. 2.** XRD analysis of  $\text{Gd}_2\text{O}_3\text{-CeO}_2\text{-NH}_4\text{H}_2\text{PO}_4$  mixture calcined at several temperatures.



**Fig. 3.** Representative X-ray diffraction patterns of  $\text{Gd}_{1-x}\text{Ce}_x\text{PO}_4$  compositions.

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