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ARTICLE

Combustion Reaction Synthesis of $Y_{2-x}Gd_xTi_2O_7$ Pyrochlore and Its Aqueous Durability as Nuclear Waste Material

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Abstract: Pyrochlore-based $Y_{2-x}Gd_xTi_2O_7/Cu$ composite waste forms were rapidly prepared within 5 min by combustion reaction plus quick pressing (CR/QP) synthesis route, where CuO was used as the oxidant and Ti as the reductant. As the surrogate of trivalent actinide nuclides, Gd was introduced to substitute the Y site with nominal formulation of $Y_{2-x}Gd_xTi_2O_7$. The results demonstrate that Gd can totally replace the Y site and $Gd_2Ti_2O_7$ pyrochlore can be readily synthesized. There is no impurity phase generated in the products, and phase separation is not detected in the pyrochlore matrix. The selected $Gd_{1.0}$ waste form exhibits promising aqueous durability as the 42 d normalized leaching rates of Cu, Y and Gd elements are evaluated as low as 2.4×10^{-2} , 1.1×10^{-5} and $5.3 \times 10^{-6} \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$.

Key words: combustion reaction synthesis; nuclear waste; pyrochlore; chemical durability

The safety disposal of high-level radioactive nuclear wastes (HLWs) becomes a major concern for the sustainable development of nuclear science and technology all over the world^[1,2]. The long-time isolation of HLWs from biosphere requires the immobilization of radionuclides in highly durable matrices, which should possess excellent chemical, mechanical, thermodynamic and radiation stabilities. At present, the immobilization of nuclear wastes in glass matrix (namely vitrification) was adopted as a large-scale solidification process in the nuclear industry^[3,4]. From the viewpoints of thermodynamic stability and loading capacity of actinide nuclides, glass matrix is not the most promising waste form. Contrary to glass, ceramic materials exhibit higher stability and can incorporate a significantly larger amount of actinide nuclides without obvious degradation of the waste form performance. As a result, highly stable and durable matrices of monophasic or as-

semblage of ceramics were proposed as an candidate material for nuclear waste management in recent decades^[5-7].

The naturally existed pyrochlore and zirconolite minerals are important host minerals of actinides in ceramic waste matrices^[8-10]. Pyrochlore and zirconolite exhibit closely related crystal structure as they are derived from the ordered anion-deficient fluorite structure. Moreover, pyrochlore exhibits much higher radiation tolerance over zirconolite. In 1990s, multi-phase pyrochlore-rich titanate ceramic waste forms were employed for the immobilization of excess weapon grade plutonium in the United States^[11,12]. From then on, titanate and zirconate pyrochlores are the mostly concerned ceramic matrix materials in relevant research field. Pyrochlore-based waste forms usually show high loading capacity, high chemical flexibility and radiation tolerance for the immobilization of long-life actinide-bearing radioactive wastes^[13-21].

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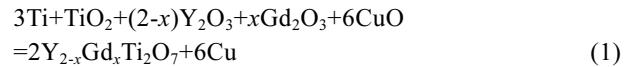
In the previous studies, pyrochlore-based waste forms were mainly synthesized from solid state reaction and liquid phase synthesis routes (such as co-precipitation, liquid mix and sol-gel)^[15-21]. These approaches are usually time consuming and costly. Meanwhile, high temperature sintering (1500 °C or even higher) is an indispensable procedure to obtain crystalline immobilization of actinides, which inevitably induces the risk of nuclides evaporation during the sintering process (such as U, Pu, Tc, Cs and Sr). Muthuraman et al^[22] proposed an alternative synthesis approach, combustion reaction (CR) synthesis, for the disposal of nuclear wastes. It is worth noting that the reaction temperature and speed of CR can be tailored by the design of CR system and adjusting the raw reactants. For nuclear waste immobilization, CR route is advantageous in some special applications when the implement condition is confined.

In our previous studies, we explored the CR preparation of Ce-bearing zirconolite ($\text{CaZrTi}_2\text{O}_7$) waste forms using CuO as the oxidant and Ti as the reductant^[23]. Meanwhile, hydraulic quick pressing (QP) was introduced to get highly densified samples. Recently, we have synthesized $\text{Y}_2\text{Ti}_2\text{O}_7$ and Nd-bearing $\text{Y}_2\text{Ti}_2\text{O}_7$ pyrochlores from similar CR/QP process using Fe_2O_3 and CuO as the oxidants^[24-28]. The as-prepared sample exhibits an analogous self-encapsulated morphology with pyrochlore core and metallic cladding (Fe and Cu). For the CR prepared Nd-bearing $\text{Y}_2\text{Ti}_2\text{O}_7$ pyrochlores using Fe_2O_3 as the oxidant, Nd could not totally substitute the Y site of $\text{Y}_2\text{Ti}_2\text{O}_7$ pyrochlore. The maximum x value in $\text{Y}_{2-x}\text{Nd}_x\text{Ti}_2\text{O}_7$ is 1.0 with the maintaining of pyrochlore structure. The phase separation of pyrochlore matrix inevitably occurs to form Y-rich and Nd-rich pyrochlore phases.

In this study, we have conducted the rapid synthesis of $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7/\text{Cu}$ composite waste forms by identical CR/QP technique with CuO as the oxidant. Gd_2O_3 was introduced as the surrogate of trivalent actinides according to the crystal chemistry^[28]. Gd is considered as a neutron poison for fission reactions because it exhibits much higher thermal neutron capture cross-section (4.9×10^4 barns for Gd and 0.184 barns for Zr)^[29]. Thus, Gd-bearing ceramic and glass-ceramic waste forms could prevent critical event for the nuclear wastes heavily loaded with fissile actinide isotopes (such as ^{239}Pu and ^{235}U)^[30]. The phase composition, microstructure, Gd_2O_3 immobilization and aqueous durability of CR/QP synthesized Gd-bearing samples were investigated.

1 Experiment

Y_2O_3 , Gd_2O_3 , Ti , TiO_2 and CuO powders with purity higher than 99% and granularity lower than 200 meshes were purchased as the raw reactants. The combustion reactions of $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7/\text{Cu}$ composites were designed as follows:



From our previous studies^[26, 27], Nd could not completely replace the Y site in Nd-bearing $\text{Y}_2\text{Ti}_2\text{O}_7$ pyrochlores ($\text{Y}_{2-x}\text{Nd}_x\text{Ti}_2\text{O}_7$). Several impurity phases were generated when the x value is higher than 1.0. In this study, the x value is tailored from 0.25 to 2.0 ($x=0.25, 0.5, 0.75, 1.0, 1.5, 2.0$) to explore the maximum Gd_2O_3 loading amount in the Gd-bearing pyrochlores. The introduced Gd_2O_3 content also represents the waste loading of trivalent actinides. The CR/QP experiments were conducted similarly as our previous reports (Fig.1)^[26, 27]. On the basis of temperature measurement, the red-hot samples were compacted at about 45 MPa by a hydraulic press for holding 60 s. A suitable time delay was explored for the compression after CR completion, which is beneficial to highly densified specimens. In this study, the delay time was fixed to be 20~30 s after the tungsten wire ignition. The whole combustion and densification takes no more than 5 min. After cooling, the solidified specimens were cut and polished to 0.3 μm for subsequent characterizations.

The as-prepared specimens were pulverized and characterized by X-ray diffraction (XRD, X'Pert PRO, PANalytical B.V.) by $\text{Cu K}\alpha$ radiation. The XRD results were further analyzed by Rietveld refinement using Fullprof-2k software package. Microstructure and elemental distribution analyses were conducted by field-emission scanning electron microscopy (FESEM; Zeiss Ultra-55, Oberkochen, Germany) equipped with energy-dispersive X-ray (EDX) spectrometer attachment. Chemical stability of the solidified specimen was evaluated according to the static standard of materials characterization center (MCC-1)^[31, 32]. The representative $\text{Gd}_{1.0}$ sample was cut and ground to external dimension of 17.6 mm \times 5.8 mm \times 5.7 mm, which was suspended by a nylon wire and immersed in 80 mL deionized water within PTFE container. The container was finally moved into a 90 °C baking oven with testing duration of 1~42 d. The immersed sample and leaching solutions (leachates) were retrieved with certain intervals according to the MCC-1 standard. The leachate concentrations (C_i) of Y and

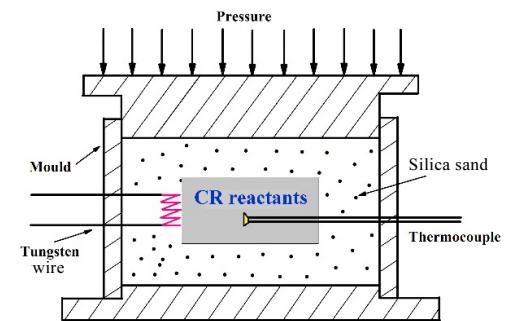


Fig.1 Schematic diagram of the proposed CR/QP process

Gd were measured by inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7700 \times spectrometer, CA, USA), while the Cu concentration was obtained by inductively coupled plasma (ICP, iCAP 6500, MA, USA).

2 Results and Discussion

2.1 Phase composition of $Y_{2-x}Gd_xTi_2O_7$ pyrochlores

The CR experiments were conducted as mentioned above, where Gd-bearing $Y_2Ti_2O_7$ pyrochlore and metallic Cu were designed as the target products. The tungsten wire ignition of all the $Y_{2-x}Gd_xTi_2O_7$ ($x=0.5\sim2.0$) samples were sustainably conducted with high heating release. The phase compositions of obtained samples were analyzed with the XRD patterns demonstrated in Fig.2. Different from the Nd-doped $Y_2Ti_2O_7$ pyrochlores, the phase compositions of Gd-bearing samples conform to the original design, where Gd-bearing $Y_2Ti_2O_7$ (Fd-3m, PDF No. 42-0413) and metallic Cu are indexed as the constituent phases. Gd can totally substitute the Y site as pyrochlore structure can be kept as shown in the $Gd_{2.0}$ sample. However, there is a small amount of SiO_2 impurity phase existing in all these samples. As 70~100 meshes silica sand was used as the heat insulator and pressure transmission medium, it might be pressed into the sample under high temperature and pressure. Thus, we can hardly remove the pollution of SiO_2 during the XRD analysis. On the other hand, the byproduct of metallic Cu is a valuable constituent phase because of its relatively high stability. Some metallic constituents in nuclear waste can be stabilized by Cu under the molten state.

According to the phase evolution principle of $A_2B_2O_7$ materials^[13-15], the formation of crystal structure is mainly dependent on the r_A/r_B value (r_A and r_B represent the cation radius of elements A and B, respectively). Cubic pyrochlore structure is formed with $r_A/r_B=1.46\sim1.78$ while defect fluorite structure is formed when r_A/r_B value exceeds this scope. As the cation radius of Y, Gd and Ti are 0.090, 0.0938 and 0.0605 nm, respectively, the r_A/r_B values are calculated to be among 1.49~1.55 for the $Y_{2-x}Gd_xTi_2O_7$ ($x=0.5\sim2.0$) samples. In other words, the formation of pyrochlore structure can be expected for all the $Y_{2-x}Gd_xTi_2O_7$ samples. This ordered pyrochlore structure is testified by the typical diffraction peaks of (311) and (511) located at about 38° and 47°. The most intensive peaks of pyrochlore (among 30°~31°) for the Gd_0 , $Gd_{1.0}$ and $Gd_{2.0}$ samples are extracted and magnified as depicted in Fig.2b. The diffraction peaks gradually shift toward the lower angle with the increase of x value (30.65° for Gd_0 and 30.40° for $Gd_{2.0}$). This phenomenon is associated with the occupation of Y site by larger Gd cation when substitutional solid solution was formed during this reaction. As the cation radius of Gd^{3+} (0.0938 nm) is higher than that of Y^{3+} (0.090 nm), the incorporation of Gd leads to elevated lattice constant and unit cell volume.

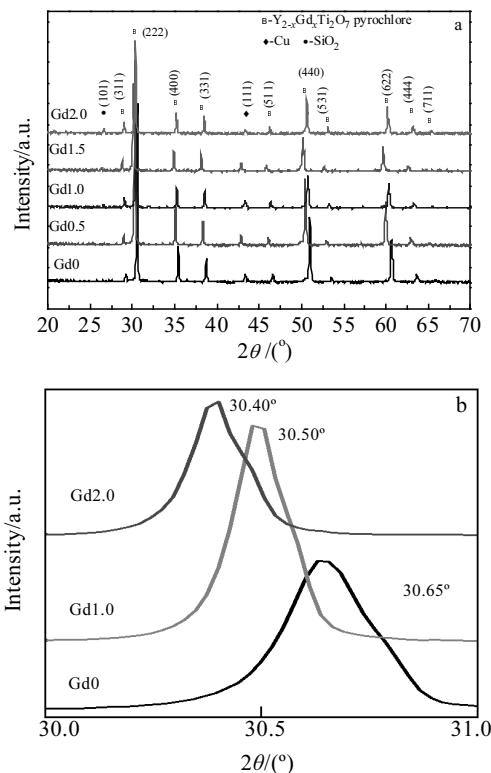


Fig.2 XRD patterns of the CR synthesized Gd-bearing $Y_{2-x}Gd_xTi_2O_7$ ($x=0\sim2.0$) samples

The XRD results of all the Gd-bearing samples were analyzed in detail by Rietveld refinement. The reported structural details of cubic $Y_2Ti_2O_7$ pyrochlore were employed as the initial model. The refined XRD pattern of $Gd_{0.5}$ sample is typically shown in Fig.3a. The structural parameters and diffraction peaks of $Gd_{0.5}$ sample fit greatly well with $Y_2Ti_2O_7$ pyrochlore and Cu phases. The unit cell parameters and unit cell volumes are computed and depicted in Fig.3b, which were measured from XRD refinement of the Gd-bearing samples with x values of 0~2.0. The unit cell parameter and unit cell volume increase gradually with the increment of Gd content, which is attributed to the higher radius of Gd^{3+} (0.0938 nm) over that of Y^{3+} (0.090 nm). And the unit cell parameters and x value exhibits a linear relationship, which can be reflected by two mathematical equations as inserted in Fig.3b. This is reasonable as the substitutional solid solution is formed in the $Y_{2-x}Gd_xTi_2O_7$ pyrochlores.

2.2 Microstructure and elemental distribution analysis

The reaction products of $Y_{2-x}Gd_xTi_2O_7$ pyrochlore/Cu composites were compressed at 45 MPa after a suitable time delay of combustion (20~30 s). The densified $Gd_{0.5}$, $Gd_{1.0}$, $Gd_{1.5}$ and $Gd_{2.0}$ samples were observed with the

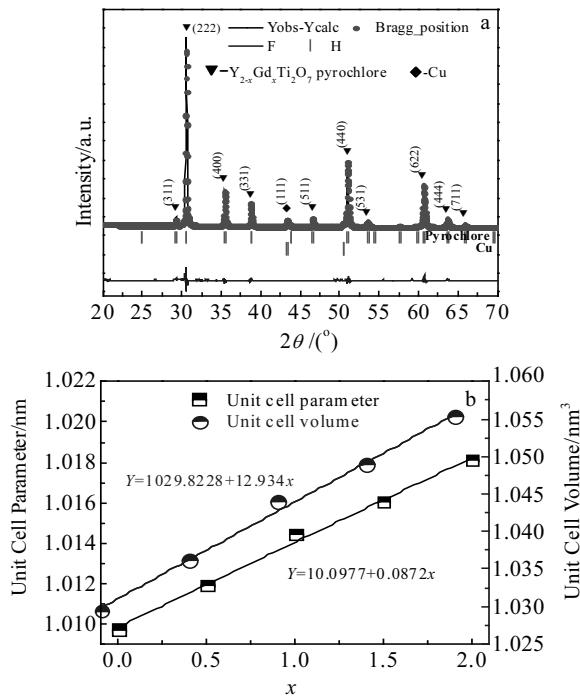


Fig.3 Rietveld refined XRD pattern of the $\text{Gd}_{0.5}$ sample (a); unit cell parameter and unit cell volume of $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ ($x = 0\sim 2.0$) phase (b)

back-scattered electron (BSE) images illustrated in Fig.4. According to our previous report^[27], the CR prepared Nd-bearing pyrochlore ($\text{Y}_{2-x}\text{Nd}_x\text{Ti}_2\text{O}_7$) matrix separates into two substitutional solid solutions when using Fe_2O_3 as the oxidant. During the SEM observation of this study, the generated Cu phase was excluded and only the ceramic matrix phase was characterized. All the Gd-bearing samples exhibit uniform morphology without contrast discrepancy, which means phase separation does not exist in the ceramic matrix. In other words, there is only one solid solution phase in the ceramic matrix because Y and Gd can be mutually substituted under arbitrary proportion. The morphology of $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ ceramic matrix is similar to typical cast-dendrite structure in metallic materials. No grain boundaries can be observed in the prepared samples, which is highly different from the high temperature sintered ceramic counterparts. This kind of morphology may be beneficial to improve the chemical durability of target waste forms.

The EDX mapping and spotting analysis of the $\text{Gd}_{1.0}$ sample were conducted to further analyze the phase composition and Gd immobilization. Fig.5a presents the mixed EDX mapping image of Cu, Y and Gd elements. The elemental distribution is further elucidated from their individual mapping images as shown in Fig.5b~5d. It can be distinctly observed that there is Cu phase in this area.

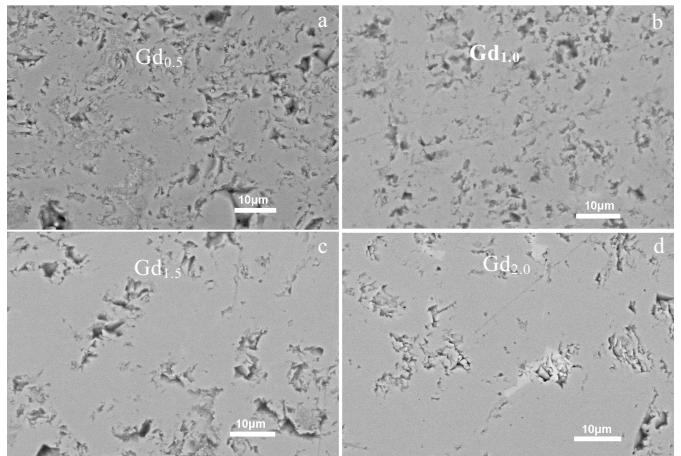


Fig.4 BSE images of the $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ ($x = 0.5\sim 2.0$) samples: (a) $\text{Gd}_{0.5}$, (b) $\text{Gd}_{1.0}$, (c) $\text{Gd}_{1.5}$, and (d) $\text{Gd}_{2.0}$

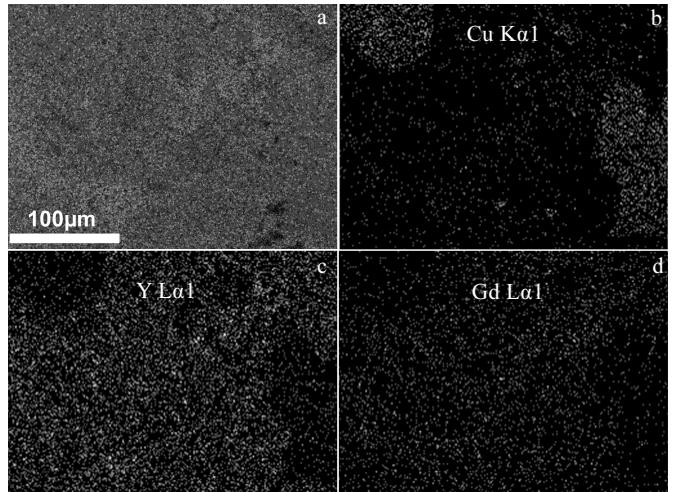


Fig.5 EDX mapping images of the representative $\text{Gd}_{1.0}$ sample: (a) mixed elemental, (b) Cu, (c) Y, and (d) Gd

Except for Cu, the Y and Gd elements distribute uniformly in the ceramic phase. This result testifies that there is no phase separation in the $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ ceramic phase. EDX spotting results further confirm this elemental distribution with semi-quantitative results, where the chemical formulation of pyrochlore phase is computed as $\text{Y}_{1.05}\text{Gd}_{0.96}\text{Ti}_{1.96}\text{O}_7$. This elemental composition is close to the originally designed $\text{Y}_{1.0}\text{Gd}_{1.0}\text{Ti}_2\text{O}_7$. Meanwhile, the SEM-EDX results are in agreement with the XRD results, which further verify that $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ waste forms are rapidly synthesized by this CR/QP method.

2.3 Aqueous durability of Gd-bearing waste form

Standard MCC-1 measurements were conducted at 90 °C to estimate the leaching behavior of typical $\text{Gd}_{1.0}$ sample. The normalized leaching rate (LR_i) directly reveals the chemical stability of relevant radioactive nuclides stabilization.

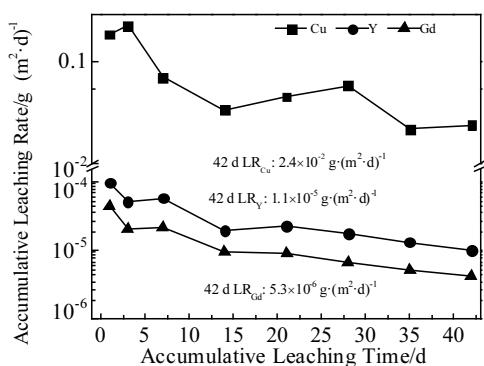


Fig.6 Effect of accumulative leaching time on leaching rate of the $\text{Gd}_{1.0}$ sample

The calculated 1~42 d LR_i values of Cu, Y and Gd elements are depicted in Fig.6. The LR_{Cu} value exhibits a slight increment at 3 d and among 14~28 d. However, the LR_{Y} and LR_{Gd} values decrease congruently as the soaking time is prolonged from 1 to 42 d except for a slight fluctuation at 7 d. Similarly, the 42 d LR_{Cu} , LR_{Y} and LR_{Gd} are the lowest values for Cu, Y and Gd elements, which are measured to be 2.4×10^{-2} , 1.1×10^{-5} and $5.3 \times 10^{-6} \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$, respectively. As reflected from the LR_i values, this leaching tendency is reasonable as the elemental leaching becomes almost unchanged when the soaking time is prolonged^[33]. This steady stage is associated with the pseudo equilibrium state, which is generated between the formation and dissolution of protective layer at the sample surface. It should be noted that the LR values of Cu, Y and Gd are comparable of titanate Synroc (about $10^{-3} \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$)^[34] and significantly lower than that of typical borosilicate glass (about $1 \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$)^[35]. The leaching results demonstrate that the prepared $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7/\text{Cu}$ composite is a durable material for nuclear waste immobilization. And the simple and rapid CR/QP technique is a candidate approach for nuclear waste management.

3 Conclusions

1) Densified $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7/\text{Cu}$ composite waste forms are readily synthesized within 5 min from CR/QP technique. $\text{Y}_{2-x}\text{Gd}_x\text{Ti}_2\text{O}_7$ ($x=0\sim 2.0$) pyrochlore is fabricated as the main ceramic phase. The lattice constant and crystal cell volume of pyrochlore phase increase linearly with the increment of x value.

2) The ceramic matrix is composed of single phase pyrochlore rather than separated solid solutions. The selected waste form ($\text{Gd}_{1.0}$) demonstrates promising aqueous durability with 42 d normalized leaching rates of Cu, Y and Gd elements as low as 2.4×10^{-2} , 1.1×10^{-5} and $5.3 \times 10^{-6} \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$.

3) From the viewpoints of aqueous stability and waste loading, the prepared pyrochlore-based composite is a promising waste form and the CR/QP technique is a candidate approach for nuclear waste disposal.

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燃烧反应制备 $Y_{2-x}Gd_xTi_2O_7$ 烧绿石及其作为核废物固化材料的稳定性研究

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摘要: 烧绿石基陶瓷材料是理想的核废物固化基材, 本研究采用燃烧反应结合快速加压工艺, 以 CuO 为氧化剂、Ti 为还原剂, 在 5 min 内快速制备出烧绿石基 $Y_{2-x}Gd_xTi_2O_7/Cu$ 复合材料固化体。以 Gd 作为三价锕系核素的模拟物, 设计 Gd 取代 $Y_{2-x}Gd_xTi_2O_7$ 烧绿石中的 Y 位。结果表明, Gd 能完全取代 Y 位形成 $Gd_2Ti_2O_7$ 烧绿石, 所得产物没有杂质相出现, 且在烧绿石基体中没有出现相分离现象。选择 $Gd_{1.0}$ 样品进行浸出试验, 结果表明该样品具有优异的水热稳定性, Cu、Y 和 Gd 元素的 42 d 归一化浸出率分别低至 2.4×10^{-2} 、 1.1×10^{-5} 和 $5.3 \times 10^{-6} \text{ g} \cdot (\text{m}^2 \cdot \text{d})^{-1}$ 。

关键词: 燃烧反应制备; 核废物; 烧绿石; 化学稳定性

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