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Substitution mechanisms of zirconolite for nuclear waste form: a mini review

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Zirconolite waste forms are advanced ceramic materials for the immobilisation of HLW, particularly actinides. This paper presents a systematic review of seven common substitution mechanisms of nuclear wastes in zirconolite, categorized into two types: charge-compensated substitution and direct substitution. For those substitution modes, three primary phase evolution pathways were identified: *i* zirconolite-2M to zirconolite-4M and/or to pyrochlore; *ii* zirconolite-2M to zirconolite-3O; *iii* zirconolite-2M to zirconolite-3T. The formation of zirconolite-3T or zirconolite-4M as intermediate phases is typically influenced by substitution behaviour, preparation conditions, or fabrication methods. Additionally, the substitution mechanisms of actinides (or REE analogue) in zirconolite were systematically investigated, corresponding to the seven substitution designs. Notably, the preferential occupation of An/REE in Ca sites was verified at simultaneous direct substitution in both Ca and Zr sites. Whilst extensive studies has explored An/REE substitution mechanisms in zirconolite and identified phase evolution pathways to zirconolite-3O, -3T, -4M, and pyrochlore, the substitution behaviour, radiation stability and chemical durability of these defect-fluorite derivatives warrant further systematic investigation.

KEYWORDS

zirconolite, substitution mechanism, nuclear waste form, phase evolution, actinides

1 Introduction

Nuclear power will assume an increasingly significant role in the future global energy structure owing to its high energy density, environmental friendliness and sustainability. It also plays a vital role in mitigating greenhouse gases, particularly in achieving China's carbon peaking and carbon neutrality goals. However, their unique challenge of radioactive waste severely constrains nuclear energy development. The radioactive wastes from spent nuclear fuel contain fission product gases (e.g., Xe, I), metallic fission products (e.g., Mo, Tc, Pb), oxides-forming fission products (e.g., Zr, Ba, Cs, Nb, lanthanides), and transuranic elements (e.g., Pu, Np, Am, Cm) (Ewing, 2015). Radioactive wastes are typically classified as low-level waste (LLW), intermediate-level waste (ILW), and high-level waste (HLW) according to radionuclide inventory, decay heat, and half-life. Robust disposal solutions have been developed and implemented for LLW and short-lived ILW (Hua and Deng, 2006;

Corkhill and Hyatt, 2018; IAEA, 2022). HLW has attracted significant concern due to its long-term radioactivity, high decay heat, chemical and biological toxicity (Slovic et al., 1991; Ewing, 1999; Corkhill and Hyatt, 2018). Should HLW not be safely disposed, resultant environmental and public health impacts could trigger a crisis in public trust, further constraining the development of nuclear energy.

For long-term nuclear waste disposal, numerous options, including outer space, rock melting, sea disposal, ice sheets, were historically considered. Currently, deep geological disposal is accepted by most countries as the definitive solution of radioactive wastes, particularly HLW containing actinides. This preference stems from its multi-barrier disposal systems which could prevent the radionuclides from reaching humans and the environment (Corkhill and Hyatt, 2018; Ewing and Park, 2021). Prior to the deep geological disposal, HLW and long-lived ILW require immobilization in glass, ceramic (or SYNROC) or glass-ceramic matrices (Donald et al., 1997; Ewing, 1999; Weber et al., 2009; Corkhill and Hyatt, 2018). Glass waste forms could accommodate approximately 40 different elements, ~35 wt.% HLW loadings, and have been industrially implemented in several countries (Donald et al., 1997; Weber et al., 2009; Sengupta, 2012; Corkhill and Hyatt, 2018). Vitrification technologies include the induction-heated metallic Melter, cold-crucible Melter, and joule-heated ceramic Melter, but the disadvantages of glass waste forms limited the application in the immobilisation of HLW, for instance, low solubility for special and important radionuclides (e.g., actinides and Tc), thermodynamic metastability, and problematic phase segregation (e.g., yellow phase formation) (Donald et al., 1997; Vance, 2007; Yudintsev et al., 2007; Weber et al., 2009; Ojovan et al., 2021). Ceramic waste forms can immobilise radionuclides within crystalline lattices, offering superior actinides (An) solubility, chemical durability, and radiation tolerance (Donald et al., 1997; Ewing, 1999; Vance, 2007; Yudintsev et al., 2007; Weber et al., 2009; Gregg and Vance, 2017; Orlova and Ojovan, 2019). Ceramic matrices have been regarded as second-generation waste forms, including zirconolite, pyrochlore, zircon, garnet, hollandite, and monazite, but they lack industrial implementation (Donald et al., 1997; Ewing et al., 2004; Vance, 2007; Weber et al., 2009; Wang and Liang, 2012; Stefanovsky and Yudintsev, 2016; Gregg and Vance, 2017; Vance et al., 2017; Fuentes et al., 2018; McMaster et al., 2018; Finkeldei et al., 2021; Lumpkin and Aughterson, 2021; Yudintsev et al., 2023). Glass-ceramic waste forms are designed to merge the advantages of both ceramic and glass to immobilise HLW, incorporate specific waste streams, and facilitate industrial-scale production with existing vitrification infrastructure (Rawlings et al., 2006; Vance, 2007; Yudintsev et al., 2007; Weber et al., 2009; Vance et al., 2012). As a pragmatic compromise, glass-ceramics have been attractive in recent decades. Sphene-based, zirconolite-based and other glass ceramics were devised and studied widely, but the optimization of fabrication process, immobilisation mechanism and performance characteristics require further research (Lee et al., 2006; Rawlings et al., 2006; Weber et al., 2009; Vance et al., 2012; Corkhill and Hyatt, 2018; Ojovan et al., 2021). Consequently, ceramics represent the most promising matrices for the immobilisation of HLW, especially for actinides. Recent advances in high-entropy ceramics offer novel approaches for multi-radionuclide solidification (Wang et al., 2024;

Wang et al., 2025). The oxides ceramic systems, as zirconolite, pyrochlore, garnet, perovskite, have emerged as leading candidates for actinides immobilisation.

Zirconolite, as one of fluorite-derived structures, was first proposed as a main constituent of SYNROC for the immobilisation of HLW towards the end of the 1970s (Ringwood et al., 1979). Since then, the research interest in zirconolite waste forms has grown, and several review articles concerning zirconolite crystal chemistry and stability have emerged. In the 1990s, Matzke et al. reviewed the immobilisation of several actinides and their rare-earth element analogues in zirconolite (Matzke et al., 1990). However, the solid-state behaviour of actinides in zirconolite remained poorly understood. Gieré et al. subsequently summarised 24 possible substitution types and nine typical chemical compositions in natural zirconolite, based on a statistical study of ~300 chemical analyses (Gieré et al., 1998). More recently, Blackburn et al. reviewed the crystal chemistry and aqueous durability of zirconolite, focusing on the incorporation of Ce/Pu/U in the Ca^{2+} site with charge compensators and the Zr^{4+} site, noting typical elemental release rates from zirconolite of less than $10^{-5} \text{ g m}^{-2} \text{ d}^{-1}$ (Blackburn et al., 2021a). The solid solution limits of neutron absorbing additives (Gd, Hf, Sm, In, Cd, B) and Pu in zirconolite were also reviewed (Blackburn et al., 2023). Based on considerations of cost and neutronics, they proposed that Gd and Hf are the most promising oxides for the candidate Pu-zirconolite phase. Yuditsev et al. have provided a detailed analysis of zirconolite-structured phases for the immobilisation of REE-actinide-rich wastes, identifying cold pressing and sintering (CPS), pressure-assisted sintering techniques, or melt crystallization as the most feasible industrial-scale processing routes (Yuditsev et al., 2023). Therefore, this article discusses the substitution mechanism within synthetic zirconolite as a potential matrix for rare-earth elements (REE) and actinides fractions of HLW. It aims to provide a comprehensive literature review on the design and application of REE and actinide solidification from the perspective of substitution mechanism in zirconolite.

2 The substitution mechanism of REE/An in zirconolite

2.1 Theoretical substitution

The ideal chemical formula of zirconolite is $\text{CaZrTi}_2\text{O}_7$ (or ABC_2O_7), space group C2/c (No.15). There are three-type cations, with their coordination environments being CaO_8 , ZrO_7 , TiO_6 and TiO_5 (Rossell, 1980; Whittle et al., 2012). Owing to its three distinct cation sites and the principle of isomorphism, zirconolite exhibits highly flexible element substitution behaviour. Gieré et al. proposed 24 potential substitutions types in zirconolite, but only a few theoretically possible substitutions are significant for the immobilisation of nuclear wastes (Gieré et al., 1998). Over the latest 30 years, numerous studies have been published on the incorporation of REE/An into zirconolite, with the most theoretically important substitutions were listed in Table 1 (Gieré et al., 1998; Weber et al., 2009; Blackburn et al., 2021a; Yuditsev et al., 2023). These substitutions could be classified into two typical

TABLE 1 Theoretically important substitutions of REE/An in zirconolite.

Number	Substitution		Replaced sites
1	2REE/An ³⁺	↔	Ca+Zr
2	REE/An ⁴⁺	↔	Zr
3	REE/An ³⁺ +Me ³⁺	↔	Ca+Ti
4	2REE/An ³⁺ +Me ²⁺	↔	2Ca+Ti
5	REE/An ⁴⁺ +2Me ³⁺	↔	Ca+2Ti
6	REE/An ³⁺ +Me ⁵⁺	↔	Zr+Ti
7	2REE/An ³⁺ +Me ⁶⁺	↔	2Zr+Ti

REE, rare earth elements, An, actinides, Me, metal elements.

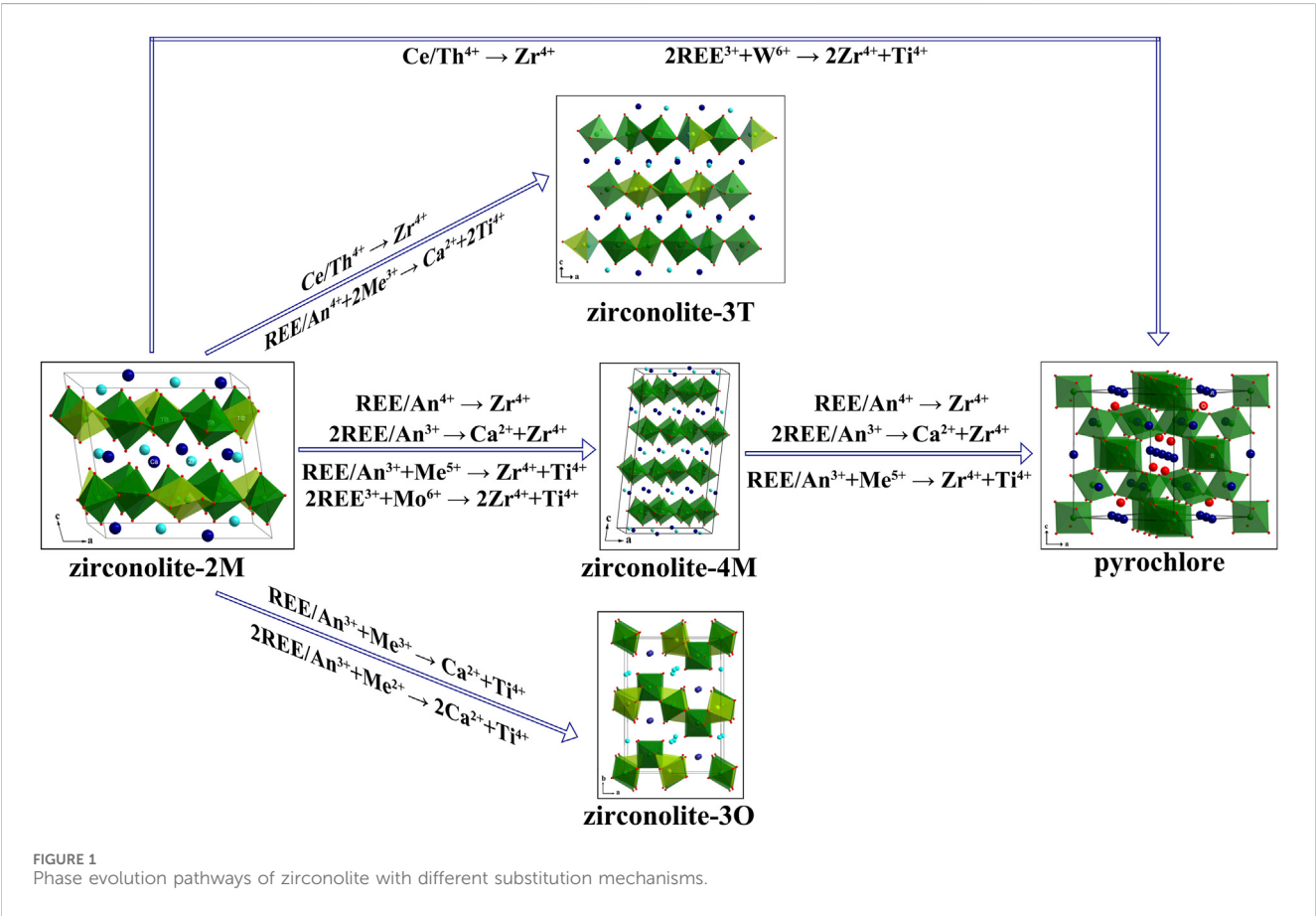
scenarios: direct substitution and co-substitution with charge compensators such as Al³⁺, Fe^{2+/3+}, Mn²⁺, Cr³⁺, Nb⁵⁺ and other metal (Me) elements.

2.2 No charge compensator

Generally, REE/An ions may occupy Ca, Zr, and Ti sites in zirconolite through direct substitution, either individually or via simultaneous multi-cation replacement. The most significant substitutions are types 1# and 2#, which require no charge compensation. Extensive research exists on CaZrTi₂O₇-

REE₂Ti₂O₇ (REE = Ce, Nd, Gd, Sm, Dy, Ho, Y) systems (Jafar et al., 2014a; Jafar et al., 2014b; Jafar et al., 2016; Meng et al., 2016; Zhang et al., 2016b; Zhang et al., 2017c; Yin et al., 2018; Zhang et al., 2018; Zhang et al., 2019; Blackburn et al., 2022; Ji et al., 2024). The dominant crystal phase evolution pathway has been identified from zirconolite-2M to zirconolite-4M, then to pyrochlore. However, high dopant concentrations of Nd and Ce yield primary multiphase assemblages rather than pyrochlore (Jafar et al., 2014b; Zhang et al., 2020). This behaviour can be attributed to the larger ionic radii of Nd³⁺ and Ce³⁺, which exceed the stable R_A/R_B cationic radii ratio required for ordered pyrochlore formation (Ewing et al., 2004). The possible preferential substitution of Ln³⁺ (Ln = Ce, Gd, Nd, Sm, Dy, Ho) in Ca site of zirconolite-2M structure were determined by XANES, Rietveld refinement and other method. However, this preference was not observed in Ca_{1-x}Zr_{1-x}Y_{2x}Ti₂O₇ system due to smaller ionic radii of Y³⁺ (Jafar et al., 2016). For pyrochlore (A₂B₂O₇), all the REE only occupy the A site (Blackburn et al., 2022; Ji et al., 2024). In addition, Analogous zirconolite-to-pyrochlore transformations occur in CaHfTi₂O₇-(Gd/Ce)₂Ti₂O₇ systems (Perera et al., 2002; Zhang et al., 2020).

The direct substitution of An⁴⁺ and Ce⁴⁺ at Zr sites of zirconolite has been extensively studied owing to favourable charge balance and comparable ionic radii. In CaZr_{1-x}M_xTi₂O₇ (M = Ce, U, Pu, Np) systems, phase evolution typically progresses from zirconolite-2M to zirconolite-4M to pyrochlore (Vance et al., 1994b; Begg et al., 1997; Vance et al., 2002; Clark et al., 2017). However, CaZr_{1-x}Th_xTi₂O₇ and select SHS sintered CaZr_{1-x}Ce_xTi₂O₇ systems exhibited direct



transformation of zirconolite-2M to pyrochlore, bypassing the zirconolite-4M phase (Zhang et al., 2015; Blackburn et al., 2021b). Moreover, Wen et al. reported the phase evolution of zirconolite-2M to zirconolite-3T in $\text{CaZr}_{1-x}\text{Ce}_x\text{Ti}_2\text{O}_7$ ($x = 0-0.5$) samples synthesised via solid-state reaction using CaF_2 , Ti, TiO_2 , ZrO_2 , CeO_2 and Fe_2O_3 (Wen et al., 2015). This pathway was observed in $\text{CaZr}_{1-x}\text{Th}_x\text{Ti}_2\text{O}_7$ ($x = 0-0.5$) samples sintered under reducing conditions (Blackburn et al., 2021b). Interestingly, graphite-die hot pressing of $\text{CaZr}_{0.8}\text{Pu}_{0.2}\text{Ti}_2\text{O}_7$ resulted in: *i* complete reduction of Pu^{4+} to Pu^{3+} , *ii* Pu^{3+} substitution at Ca sites of zirconolite, and *iii* ~50% perovskite phase formation (Begg et al., 1997). Multiple studies reported perovskite emergence in $\text{CaZr}_{1-x}\text{Ce}_x\text{Ti}_2\text{O}_7$ systems at elevated Ce concentrations (Zhang et al., 2015; Zhang et al., 2017a; Blackburn et al., 2020a; Kong et al., 2021). The substitution of U^{4+} in Zr sites of zirconolite was determined by Sharivastava et al. using Rietveld refinement (Shrivastava et al., 2005; Shrivastava et al., 2006). Subramani et al. discovered that $\text{U}^{4+/6+}$ incorporated into Zr sites of zirconolite for $\text{CaZr}_{1-x}\text{U}_x\text{Ti}_2\text{O}_{7+x}$ ($0 \leq x \leq 0.4$) (Subramani et al., 2020). Contrastingly, Zhang et al. identified predominant substitution of Ce at Ca sites rather than Zr sites in $\text{CaZr}_{1-x}\text{Ce}_x\text{Ti}_2\text{O}_7$ ($x = 0-0.5$) samples when MoO_3 and Ti served as oxidant and reductant, respectively (Zhang et al., 2016a). It can be attributed to the reduction of Ce^{4+} to Ce^{3+} . In addition, Gatehouse et al. reported zirconolite formation at $x = 0.85-1.3$ in $\text{CaZr}_x\text{Ti}_{3-x}\text{O}_7$ system, indicating mutual Zr/Ti site occupancy (Gatehouse et al., 1981). Nevertheless, there are no articles on the substitution of REE/An at Ti sites. It could be caused by the smaller ionic radii and lower coordination number of Ti relative to Zr, Ca, and REE/An.

2.3 Low-valency ions as charge compensators

Trivalent actinides preferentially occupy the Ca sites in zirconolite. Charge compensation could be achieved through coupled substitutions at Ti sites using low-valency ions. The low-valency ions are defined as those with oxidation states below +4 (i.e., lower than Ti^{4+}), such as Mg^{2+} , $\text{Fe}^{2+/3+}$, Al^{3+} , which are used in industry. In 1994, Vance et al. conducted pioneering work incorporating Nd^{3+} , $\text{Ce}^{3+/4+}$, U^{4+} , Pu^{3+} , Np^{3+} into zirconolite with Al^{3+} as charge compensator (Vance et al., 1994a; Vance et al., 1994b). They found the phase transformation of monoclinic to orthorhombic zirconolite in $\text{Ca}_{1-x}\text{Nd}_x\text{Ce}_x\text{ZrTi}_{2-x}\text{Al}_x\text{O}_7$ samples (Vance et al., 1994b). Begg et al. found that the reducing conditions induced the formation of perovskite (~10%) in $\text{Ca}_{0.8}\text{Pu}_{0.2}\text{HfTi}_{1.8}\text{Al}_{0.2}\text{O}_7$ and $\text{Ca}_{0.8}\text{Pu}_{0.2}\text{HfTi}_{1.6}\text{Al}_{0.4}\text{O}_7$ samples (Begg et al., 1997). Stefanovsky et al. documented the possible phase evolution of zirconolite-2M to higher symmetry structure (zirconolite-3T/6T at $x = 0.4-1.0$, zirconolite-3O at $x \rightarrow 1.0$) in $\text{Ca}_{1-x}\text{Gd}_x\text{ZrTi}_{2-x}\text{Al}_x\text{O}_7$ ($0 \leq x \leq 1$) (Stefanovsky et al., 2002). Zhang et al. synthesised Nd-Al co-doped zirconolite with CuO oxidant, confirming the substitution of Nd at Ca sites and Al at Ti sites (Zhang et al., 2017b). Ji and co-worker had been systematically investigated the phase evolution and substitution mechanism of $\text{Ca}_{1-x}\text{Ln}_x\text{ZrTi}_{2-x}\text{Al}_x\text{O}_7$, $\text{Ca}_{1-x}\text{Ln}_x\text{ZrTi}_{2-x}\text{Fe}_x\text{O}_7$ and $\text{Ca}_{1-x}\text{Ln}_x\text{ZrTi}_{2-x}(\text{Al}, \text{Fe})_x\text{O}_7$ (Ln = La, Nd, Gd, Ho, Yb; $x = 0-1$) solid solutions (Ji et al., 2018; Ma et al., 2018; Ji et al., 2020). They found that: *i* phase

evolution of zirconolite-2M to zirconolite-3O, *ii* Ln completely incorporated into zirconolite (2M/3O) except La, *iii* the substitution of Ln in Ca sites and Al/Fe in Ti sites. A detail research was published on substitution behaviour of Nd co-doped zirconolite with low charge compensator (Mg^{2+}) (Zhou et al., 2022). The results indicated: *i* phase transformation from zirconolite-2M to zirconolite-3O, *ii* the substitution of Nd in CaO_8 sites and Mg in TiO_5 sites for zirconolite-2M, and *iii* the substitution of Nd in CaO_8 site and Mg in $\text{TiO}_4+\text{TiO}_5$ sites for zirconolite-3O. Furthermore, Zhang et al. reported the phase transformation from zirconolite-2M to zirconolite-3T in $\text{Ca}_{1-x}\text{Sm}_x\text{ZrTi}_{2-x}\text{Al}_x\text{O}_7$ compounds when CuO was used as oxidant (Zhang et al., 2019). The phase evolution of zirconolite-2M to zirconolite-3T were observed in Ce^{4+} co-doped zirconolite with two or three trivalent metal cations as charge compensator (Ai et al., 2024a; Ai et al., 2024b). Moreover, the same pathway (zirconolite-2M to zirconolite-3T) was found in $\text{Ca}_{1-x}\text{Pu}_x\text{ZrTi}_{2-x}\text{Fe}_{2x}\text{O}_7$ ($x = 0.1-0.7$) (Gilbert et al., 2010). The zirconolite-2M could accommodate ~35 at% Ce at Ca sites with Cr^{3+} as charge compensator in $\text{Ca}_{1-x}\text{Ce}_x\text{ZrTi}_{2-x}\text{Cr}_{2x}\text{O}_7$ samples (Blackburn et al., 2020b). For $\text{Ca}_{1-x}\text{Ce}_x\text{ZrTi}_{2-x}\text{Cr}_x\text{Fe}_x\text{O}_7$, zirconolite-2M could be formed at $x \leq 0.25$ (Chen et al., 2023). In addition, Hayashizaki et al. found that Ce-Al co-doped could be to prevent perovskite formation than that without charge compensator (Hayashizaki et al., 2025). Wei et al. incorporated Pr (as surrogate of Np) into Ca site with Al^{3+} as charge compensator (Wei et al., 2021). The zirconolite-2M phase could be more than 93 wt.%.

2.4 High-valency ions as charge compensator

High-valency ions are defined as elements with oxidation states exceeding +4 (i.e., higher than Ti^{4+}) in zirconolite waste forms. The high-valency ions (as Nb^{5+} , Mo^{6+}) have been found in spent nuclear fuel and natural zirconolite, so that it is possible to immobilize actinides when employed these high-valency ions as charge compensator (Gieré et al., 1998; Ewing, 2015; Chukanov et al., 2018). Ji et al. systematically investigated the high-valency ion (Nb^{5+}) substitution in $\text{CaZr}_{1-x}\text{Nd}_x\text{Ti}_{2-x}\text{Nb}_x\text{O}_7$ ($x = 0.05-1$) systems, observing the fluorite-derived phase evolution: zirconolite-2M to zirconolite-4M and then to pyrochlore, with zirconolite-3T appearing as a minor intermediate phase between zirconolite-2M and zirconolite-4M. (Ji et al., 2021). Nb^{5+} preferentially occupied Zr sites of zirconolite-2M alongside Nb^{5+} in Ti1 sites. For pyrochlore ($\text{A}_2\text{B}_2\text{O}_7$) structure, the substitution mechanism comprised: *i* $\text{Ca}^{2+}+\text{Nd}^{3+}+\text{Zr}^{4+}$ at A sites, *ii* $\text{Ti}^{4+}+\text{Nb}^{5+}$ at B sites. Song et al. reported identical phase evolution in $\text{CaZr}_{1-x}\text{Sm}_x\text{Ti}_{2-x}\text{Nb}_x\text{O}_7$ ($x = 0.1-1.0$) solid solutions though without zirconolite-3T intermediate phase (Song et al., 2025). Huangfu et al. conducted detailed analysis of $\text{CaZr}_{1-2x}\text{REE}_{2x}\text{Ti}_{2-x}\text{Mo}^{6+}_x\text{O}_7$ (REE = Nd, Sm, Gd, Ho, Yb) samples, noting: *i* the phase evolution of zirconolite-2M to zirconolite-4M, *ii* ~25 at.% solubility limit in zirconolite (2M and 4M), *iii* phase separation into “yellow phase” (CaMoO_4) at $x > 0.25$, coexisting with pyrochlore (Huangfu et al., 2024). In zirconolite-2M structure, Mo^{6+} ion preferentially occupies TiO_5 site. REE (e.g., Sm, Ho) could mainly replace the 8f Zr sites with a considerable amount substitution in 8f Ca sites. In addition, the phase evolution was studied in details for W^{6+} co-doped CaZr_{1-x}

$_{2x}\text{Ln}_{2x}\text{Ti}_{2-x}\text{W}^{6+}_x\text{O}_7$ (Ln = Nd, Sm, Gd, Ho, Yb) (Huang et al., 2024). In this condition, zirconolite-2M directly transformed to pyrochlore with secondary “yellow phase”.

3 Conclusion and outlook

The seven most important substitution mechanisms for actinides (or their REE surrogates) in zirconolite were systematically discussed and summarised. Figure 1 summarises the phase evolution pathways for these substitution mechanisms, which comprise two classes: charge-compensated substitution and direct substitution. For co-substitution with low-valency charge compensators, the phase evolution pathway may progress from zirconolite-2M to zirconolite-3O. Zirconolite-2M would transform to zirconolite-4M and subsequently to pyrochlore for direct substitution and high-valency ion (Nb^{5+}) substitution modes. For higher-valency ions (Me^{6+}), phase evolutions from zirconolite-2M to zirconolite-4M, or directly from zirconolite-2M to pyrochlore, were identified. These are accompanied by the appearance of the hazardous “yellow phase” at high dopant concentrations. Moreover, phase evolution of zirconolite-2M to zirconolite-3T would be observed in substitution modes 5# and partially 2#, under reducing conditions or others special situation. Furthermore, substitution mechanisms, ionic radii, preparation conditions, and processing parameters, amongst other factors, significantly influence phase evolution and the formation of intermediate phases. For model 1#, An/REE preferentially occupy the Ca sites of zirconolite. The preferential or main occupation of An/REE in zirconolite-2M corresponds to the substitution mechanisms detailed in Table 1. Although numerous studies have investigated substitution mechanisms with promising results, other performance indicators of zirconolite under different substitution mechanisms and concentrations, such as irradiation resistance, chemical durability, and mechanical performance, require further investigation. In addition, the substitution mechanisms discussed above are predicated on the zirconolite-2M structure. However, as evidenced by the preceding analysis, substitution induces phase evolution to zirconolite-3O, zirconolite-3T, zirconolite-4M, and pyrochlore. To evaluate the long-term performance of zirconolite waste forms in deep geological disposal, a comprehensive investigation is required into the substitution behaviour, irradiation stability, chemical durability, and other critical properties of these defect-fluorite derived structural ceramics. Finally, this review provides a valuable reference for the design and application of zirconolite in nuclear waste disposal, aerospace materials, and advance functional ceramics. We recommend that future research should prioritize evaluating the long-term performance of multiphase ceramic assemblages resulting from phase evolution, particularly those with defect-fluorite derived structures.

Author contributions

SJ: Project administration, Funding acquisition, Formal Analysis, Investigation, Writing – original draft, Writing – review

and editing, Conceptualization. CZ: Formal Analysis, Writing – original draft, Data curation, Investigation. XJ: Investigation, Formal Analysis, Writing – original draft, Data curation. CG: Formal Analysis, Data curation, Writing – original draft. YZ: Formal Analysis, Writing – review and editing, Supervision. XC: Formal Analysis, Supervision, Writing – review and editing. TD: Resources, Project administration, Writing – review and editing, Investigation, Funding acquisition. TJ: Formal Analysis, Writing – review and editing, Data curation. CL: Funding acquisition, Conceptualization, Resources, Project administration, Supervision, Writing – review and editing.

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