Radiation-Induced Amorphization of Ce-Doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ Silicate Apatite

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Abstract:

Ce-doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite (Ce=0.05 and 0.5) were irradiated with 1 MeV Kr^{2+} ion beam irradiation at different temperatures and their radiation response and the cation composition dependence of the radiation-induced amorphization were studied by *in situ* TEM. The two Ce-doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatites are sensitive to ion beam induced amorphization with a low critical dose (0.096 dpa) at room temperature, and exhibits significantly different radiation tolerance at elevated temperatures. Ce concentration at the apatite A^{I} site plays a critical role in determining the radiation response of this silicate apatite, in which the Ce^{3+} rich $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ displays lower amorphization susceptibility than $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$ with a lower Ce^{3+} occupancy at the A^{I} sites. The critical temperature (T_c) and activation energy (E_a) change from 667.5 ± 33 K and 0.162 eV of $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ to 963.6 ± 64 K and 0.206 eV of $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$. We demonstrate that the radiation tolerance can be controlled by varying the chemical composition, and enhanced radiation tolerance is achieved by increasing the Ce concentration at the A^{I} site.

Keywords: silicate apatite; radiation effects; ion beam irradiation; chemical composition

1 Introduction

The apatite structure has the form of $A^I_4A^{II}_6(BO_4)_6X_2$, in which A^I , A^{II} =Ca, Na, rare earth, fission products and/or actinides; B= Si, P, V or Cr and X= OH, F, Cl, I or O. A^I and A^{II} are the two different cationic sites with difference in their local symmetries and atomic scale environments because each A^I cation has nine O atoms as nearest neighbors and each A^{II} cation is surrounded by one anion and six O atoms of three different types. In addition, the A^{II} cations and anions are located in the flexible tunnels.[1] Compounds with the apatite-type structure have been intensively studied because of their potential applications as luminescent materials.[2, 3] Recently, rare earth (RE)-activated apatite-type silicate compounds have been drawn great attentions because of their excellent luminescent characteristics.[3-5] Among the many synthetic oxyapatites, the ternary rare-earth metal silicate $Mg_2Y_8(SiO_4)_6O_2$ is the efficient host lattice for the luminescence of various RE^{3+} (Ce^{3+} , Tb^{3+} , Dy^{3+} , Sm^{3+} , and Eu^{3+}) ions and mercury-like ions.[6] This oxyapatite host lattice consists of two cationic sites, that is, the 9-fold coordinated 4f sites with C_3 point symmetry and 7-fold coordinated 6h sites with C_s point symmetry. Both sites are suitable and easily accommodate a great variety of rare earth- and transitional-metal ions. In addition to the application as luminescent material, this rare-earth metal silicate is also good for nuclear waste disposal.

Silicate apatite $Mg_2Y_8(SiO_4)_6O_2$ is an extremely durable material that has important applications in ionic conductors[7, 8] and host phases for actinide immobilization and fission products[9]. As a host phase for actinides, silicate apatite must endure high radiation doses associated with alpha decay of the actinides and beta decay of the fission products. Natural apatite minerals are greatly resistant to alpha-decay and neutron irradiation[10]. Studying the radiation effects on silicate apatite is of great importance since radiation induced amorphization of silicate apatite results in volumetric swelling and increases the aqueous dissolution rates[11]. The ion beam irradiation induced amorphization in many different complex oxide structures[12-14] and compositions[15-19] has been investigated, and numerous models have been developed to explain various aspects of the amorphization process. Radiation stability of materials is very important for their application in nuclear waste forms because the host material's stability and long term durability can be negatively affected by the radioactive decay of the incorporated radionuclides in nuclear waste which generates displacive and ionizing radiations. Radiation stability maybe related with factors like chemical composition[20], different energy loss

mechanism[1, 21], structural disorder[22], and grain size[23, 24]. Therefore, the radiation effects in different apatite compositions are very interesting and efforts have been made to study the radiation stability of different apatite composition as nuclear waste forms, like fully phosphate fluorapatite[25], mono-silicate fluorapatite[26], synthetic britholite[21] and fully silicate apatite[17, 27]. Rare-earth silicates with the apatite structure have been observed as actinide host phases in a devitrified borosilicate HLW glass[28], a multiphase ceramic waste form[29], a glass-ceramic waste form[30], and a cement waste form[19]. Radiation effects in $\text{Ca}_2\text{RE}_8(\text{SiO}_4)_6\text{O}_2$ have been studied in detail by Cm-doping[11, 31] and ion beam-irradiation experiments[32-34].

By changing the chemical composition, mainly the A-site cations and B-site element, the apatite structure can be adjusted. The high ionic conductivity of apatite can be caused by the trivalent cations at the A-sites which resulted in nonstoichiometric composition and disordered anions in the channels at interstitial sites.[35-37] When smaller atoms were replaced by large atoms at the A-site or B-site, the apatite unit cell usually expanded due to the large unit cell parameters of the large atoms. Controlling the chemical composition of materials is a good way to improve the material's radiation stability since this way can affect the energies of defect production and annealing. There are two main factors that affect the radiation tolerance of the material due to the chemical composition: one is the anti-site defect formation energies caused by changes in cation radius ratio, and the other one is the ratio of electronic to nuclear stopping power (ENSP). To understand the chemical composition effects on the silicate apatite radiation stability, we performed the ion beam irradiation experiments on two silicate apatite compositions, $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ and $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$, with significant difference at the A^{I} and A^{II} sites in order to investigate the effects of A^{I} and A^{II} site composition on the radiation damage and defect annealing processes in the silicate apatite structure.

2 Experimental details

The Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite powders were synthesized by solid state reaction with different compositions (Ce=0.05 and 0.5). For the investigation of the silicate apatite radiation tolerance by the composition dependence, the ion beam irradiation and *in situ* TEM observations were conducted using the IVEM-Tandem Facility at the Argonne National Laboratory (ANL) with 1 MeV Kr^{2+} ions. The IVEM-Tandem Facility consists of a Hitachi H-

9000NAR TEM interfaced with an NEC Tandem ion accelerator, which enables *in situ* TEM observation of the radiation damage processes under intensive ion bombardment. Irradiation experiments in a wide temperature range from room temperature to 848K were achieved by using a heating stage equipped with the TEM. The heavy ion Krypton was chosen because of its ability to achieve high dpa in the irradiated samples within a relatively short time, as well as its inert nature to prevent reaction with the irradiated materials. By observing the selected area electron diffraction (SAED) patterns, the crystalline to amorphous transformation was monitored *in situ*. An objective aperture was inserted to cover the low index diffraction rings of the SAED in order to obtain the dark field imaging. The critical amorphization fluence, the fluence at which complete amorphization occurs, was determined when all of the diffraction maximum in the SAED patterns disappeared. The bright field images of the two silicate apatite were shown in Fig. 1. The observed critical amorphization fluence is converted to displacements per atom (dpa) using SRIM-2008 simulations using displacement threshold energies of 50 eV for Mg, Y, Ce, Si and O in $\text{Mg}_2\text{Y}_x\text{Ce}_{8-x}(\text{SiO}_4)_6\text{O}_2$, which was based on previous theoretical computations.[1, 38] In this paper, we focus on the Ce content difference and we have two different chemical compositions: $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ denominated as Ce0.5 and $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$ denominated as Ce0.05.

3 Results and discussion

3.1 Room temperature ion beam irradiation of Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite

Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite with different Ce content were irradiated by 1 MeV Kr^{2+} at room temperature. As observed with *in situ* TEM, they all underwent a crystalline/amorphous (c/a) phase transformation under 1 MeV Kr^{2+} irradiation as a result of the ballistic interaction and the subsequent atomic displacements. However, with the varying of the chemical composition, that is the different content of Ce, the critical dose of amorphization changed, which indicates that the radiation ability depends on the composition of the A^{I} and A^{II} sites. Fig. 2 showed a series of TEM dark field images and SAED patterns taken at different ion doses for Ce0.5 silicate apatite. Fig. 2(a) clearly showed the spotted crystalline diffraction pattern and high-contrast TEM dark field image, which indicated the crystalline structure of the pristine Ce0.5. Small amorphous domains began to appear at a low dose of 0.032 dpa (Fig. 2b), indicating that the direct ion beam irradiation process may be responsible for amorphization. At a dose of 0.064 dpa, extended radiation damage was observed from TEM dark field image and

SAED pattern, in which a significant diffuse halo ring appeared in SAED pattern and accompanied by the weakening of the diffraction pattern (Fig. 2c). Finally, the TEM dark field image showed a fully amorphous matrix with a SAED diffusion halo ring completely replacing the diffraction spot pattern when the dose reaches at 0.096 dpa, which is the critical amorphization dose (D_c) for Ce0.5 silicate apatite at room temperature. The room temperature ion irradiation of Ce0.05 showed a similar amorphization process to that of the Ce0.5 at room temperature, with a similar critical dose of 0.096 dpa. The irradiation-induced amorphization process for Ce0.05 was shown in Fig. 3.

3.2 Temperature dependence of radiation tolerance of Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite

Temperature plays a critical role on the radiation tolerance since it affects the defect dynamics and the radiation damage process. In general, a higher Frenkel pair recombination rate and defect interaction with the materials structure result from the increased mobility of radiation induced defects at elevated temperature. This process leads to the suppression of radiation damage due to a dynamic annealing effect. When the temperature exceeds a critical value, which the radiation defect production is smaller than the annealing rate, materials that are subject to radiation induced amorphization at room temperature will become highly radiation resistant, *i.e.* the materials can't be amorphized even at extremely high doses. Therefore, the critical temperature (T_c) is an indicator of materials' radiation tolerance, and the lower the T_c is, the higher the radiation tolerance. In order to understand the temperature dependence of the Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite, the two Ce-doped silicate apatite samples, Ce0.5 and Ce0.05, were studied using 1 MeV Kr^{2+} ion beam irradiation. The enhanced radiation tolerance at elevated temperature was confirmed by the *In situ* TEM observation for these two silicate apatites. For example, at 798 K, Ce0.05 remained highly crystalline until the dose reached 0.16 dpa and 0.192 dpa, which is almost 2 times as the critical dose of 0.096 dpa at room temperature, as shown in Fig. 4.

In addition, *In situ* TEM also showed significantly different temperature dependence of the irradiation-induced amorphization processes for the two silicate apatites with different Ce content at the A^{I} and A^{II} sites. The Ce0.5 becomes highly resistant to amorphization above ~ 667K, whereas the Ce0.05 can still be amorphized at almost 848K. The D_c values of these two silicate apatites at different temperature can be seen from Fig. 5(a). The temperature dependence curve were fit by an empirical exponential function[27, 39, 40] in the form of

$$D_c = \frac{D_0}{1 - \exp[(E_a/K)(1/T_c - 1/T)]}$$

where D_0 is the critical amorphization dose extrapolated at $T = 0$ K, E_a is the activation energy for defect annealing, and T_c is the critical amorphization temperature. From the fitting curve, the T_c for Ce0.5 is 667.5 ± 33 K while T_c for Ce0.05 is 963.6 ± 64 K. The T_c decreases with the increasing of Ce content, which indicates enhanced radiation tolerance with a higher Ce content.

The critical dose varies with the increase of temperature, which is associated with increased defect mobility and epitaxial recrystallization at the c/a interface of amorphous cascade structures.[17] The process is the result of simultaneous radiation-induced defect production and dynamic recovery process with activation energy, E_a , which are related with defect migration/recombination in the crystal structure or epitaxial recrystallization at the c/a interface. After calculation, E_a for Ce0.5 is 0.162 eV while for Ce0.05 is 0.206 eV. The activation energy of Ce0.5 is much lower than Ce0.05, which means that it is easier for defect migration/recombination at the c/a interface in Ce0.5. Therefore, decreased T_c and enhanced radiation tolerance are found with increasing Ce content in the silicate apatite. It is also noted that the critical dose of Ce0.5 and Ce0.05 are the same at room temperature but become significantly different at elevated temperature. The critical dose of Ce0.05 increases to 0.144 dpa while the critical dose of Ce0.5 largely increases to 0.26 dpa at 573K, indicating different dynamics of the defect annealing.

3.3 Composition dependence on radiation tolerance for Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite

The greater radiation susceptibility to radiation induced amorphization for $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$ as compared with $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ may be attributed to the variation of the Ce^{3+} concentration, which results in the change of unit cell volume. The Ce^{3+} ions are completely dissolved in the $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ host lattice without inducing significant changes in the hexagonal crystal structure. However, a higher Ce^{3+} doping concentration leads to an increase in the apatite lattice parameters, because of the larger ion radius of Ce^{3+} with respect to Y^{3+} . According to Li *et al.* [2], the most prominent crystal structure of $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ is the two Y^{3+} ions in 4f (C_3) sites and six Y^{3+} ions in 6h (C_s) sites, while Mg^{2+} ions only occupy the 4f (C_3) site. In addition, a previous study[41] about the crystal structure of $\text{Sr}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ indicates

that Y^{3+} occupies two different types of cationic sites A^I and A^{II} . According to Blasse's theory[42], the cationic site with free-oxygen nearby prefers to accommodate cations with high charge or small radius in order to compensate the underbonded valence[43]. Because the ionic radii of Ce^{3+} and Y^{3+} at AI sites and AII sites are 1.196 and 1.075, 1.07 and 0.96, respectively, the larger Ce^{3+} cations are more likely to occupy A^I sites rather than A^{II} sites. Li *et al.* [2] suggested that the unit cell parameters for $Mg_2Y_8(SiO_4)_6O_2$ is $a=b=9.280 \text{ \AA}$, $c= 6.626 \text{ \AA}$, and $V= 494.13 \text{ \AA}^3$. The a , c and volume values gradually increase with the increase of Ce^{3+} doping content because of the larger ion radius of Ce^{3+} than Y^{3+} . As a result of the unit cell expansion due to the increase in Ce substitution at A^I site, the radiation tolerance of silicate apatite is enhanced, such that Ce0.5, having higher Ce^{3+} occupancy at A^I site, are less sensitive to radiation-induced amorphization with relatively lower critical amorphization temperature, as compared to Ce0.05 with a lower Ce^{3+} occupancy at A^I site. In general, Ce^{3+} content at the A^I site of silicate apatite leads to change of unit cell volume, which may contribute to the enhancement of their radiation stability.

4 Conclusions

The radiation response of Ce-doped $Mg_2Y_8(SiO_4)_6O_2$ silicate apatite (Ce=0.05 and 0.5) was investigated by using 1 MeV Kr^{2+} ion beam irradiations at different temperatures, and the cation composition dependence of the radiation tolerance of silicate apatite was studied. The radiation tolerance of silicate apatite $A^I_4A^{II}_6(SiO_4)_6O_2$ can be tailored by controlling the cationic species at the apatite A-site. Increasing Ce^{3+} content at the A^I site of silicate apatite results in the change of unit cell volume and lattice parameters, which enhances the radiation tolerance of the material. The critical temperature changed from $667.5 \pm 33 \text{ K}$ of $Mg_2Y_{7.5}Ce_{0.5}(SiO_4)_6O_2$ to $963.6 \pm 64 \text{ K}$ of $Mg_2Y_{7.95}Ce_{0.05}(SiO_4)_6O_2$, and the activation energy E_a , is 0.162 eV in $Mg_2Y_{7.5}Ce_{0.5}(SiO_4)_6O_2$, and 0.206 eV in $Mg_2Y_{7.95}Ce_{0.05}(SiO_4)_6O_2$ is 0.206. This result highlights that the chemical composition control is an important method for designing radiation resistant materials as nuclear waste forms.

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References

1. F. Y. Lu, Z. L. Dong, J. M. Zhang, T. White, R. C. Ewing, and J. Lian, RSC Advances, 2013. **3**(35): p. 15178-15184.
2. G. G. Li, D. L. Geng, M. M. Shang, Y. Zhang, C. Peng, Z. Y. Cheng, and J. Lin, The Journal of Physical Chemistry C, 2011. **115**(44): p. 21882-21892.
3. S. Ferdov, R. A. S. Ferreira, and Z. Lin, Chemistry of Materials, 2006. **18**(25): p. 5958-5964.
4. Y. Q. Shen, R. Chen, G. G. Gurzadyan, J. L. Xu, H. D. Sun, K. A. Khor, and Z. L. Dong, Optical Materials, 2012. **34**(7): p. 1155-1160.
5. D. Kim, D. Park, N. Oh, J. Kim, E. D. Jeong, S. J. Kim, S. Kim, and J. C. Park, Inorganic Chemistry, 2015. **54**(4): p. 1325-1336.
6. J. Lin, and Q. Su, Journal of Materials Chemistry, 1995. **5**(8): p. 1151-1154.
7. S. Nakayama, M. Sakamoto, M. Higuchi, K. Kodaira, M. Sato, S. Kakita, T. Suzuki, and K. Itoh, Journal of the European Ceramic Society, 1999. **19**(4): p. 507-510.
8. Y. Masubuchi, M. Higuchi, S. Kikkawa, K. Kodaira, and S. Nakayama, Solid State Ionics, 2004. **175**(1): p. 357-360.
9. W. J. Weber, A. Navrotsky, S. Stefanovsky, E. R. Vance, and E. Vernaz, MRS bulletin, 2009. **34**(01): p. 46-53.
10. R. Tisserand, M. Rebetez, M. Grivet, S. Bouffard, A. Benyagoub, F. Levesque, and J. Carpena, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2004. **215**(1): p. 129-136.
11. R. C. Ewing, W. J. Weber, and F. Clinard, Progress in nuclear energy, 1995. **29**(2): p. 63-127.
12. Y. Zhang, J. Lian, C. M. Wang, W. Jiang, R. C. Ewing, and W. J. Weber, Physical Review B, 2005. **72**(9): p. 094112.
13. J. M. Zhang, F. X. Zhang, M. Lang, F. Y. Lu, J. Lian, and R. C. Ewing, Acta Materialia, 2013. **61**(11): p. 4191-4199.
14. A. Meldrum, L. A. Boatner, W. J. Weber, and R. C. Ewing, Journal of Nuclear Materials, 2002. **300**(2): p. 242-254.
15. J. Lian, J. Chen, L. M. Wang, R. C. Ewing, J. M. Farmer, L. A. Boatner, and K. B. Helean, Physical Review B, 2003. **68**(13): p. 134107.
16. J. Lian, R. C. Ewing, L. M. Wang, and K. B. Helean, Journal of materials research, 2004. **19**(05): p. 1575-1580.
17. L. M. Wang, and W. J. Weber, Philosophical Magazine A, 1999. **79**(1): p. 237-253.
18. L. M. Wang, S. X. Wang, W. L. Gong, R. C. Ewing, and W. J. Weber, Materials Science and Engineering: A, 1998. **253**(1): p. 106-113.
19. W. J. Weber, Y. W. Zhang, H. Y. Xiao, and L. M. Wang, RSC Advances, 2012. **2**(2): p. 595-604.
20. R. C. Ewing, A. Meldrum, L. M. Wang, and S. X. Wang, Reviews in Mineralogy and Geochemistry, 2000. **39**(1): p. 319-361.
21. S. Utsunomiya, S. Yudinsev, L. M. Wang, and R. C. Ewing, Journal of Nuclear Materials, 2003. **322**(2): p. 180-188.

22. J. M. Zhang, J. Lian, F. X. Zhang, J. W. Wang, A. F. Fuentes, and R. C. Ewing, *The Journal of Physical Chemistry C*, 2010. **114**(27): p. 11810-11815.
23. J. Lian, J. M. Zhang, V. Pointeau, F. X. Zhang, M. Lang, F. Y. Lu, C. Poinssot, and R. C. Ewing, *Journal of Nuclear Materials*, 2009. **393**(3): p. 481-486.
24. F. Y. Lu, Y. Q. Shen, X. Sun, Z. L. Dong, R. C. Ewing, and J. Lian, *Acta Materialia*, 2013. **61**(8): p. 2984-2992.
25. S. Soulet, J. Carpena, J. Chaumont, O. Kaitasov, M. O. Ruault, and J. C. Krupa, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 2001. **184**(3): p. 383-390.
26. S. Soulet, J. Carpena, J. Chaumont, J. C. Krupa, and M. O. Ruault, *Journal of nuclear materials*, 2001. **299**(3): p. 227-234.
27. F. Y. Lu, T. K. Yao, Y. Danon, J. R. Zhou, R. C. Ewing, and J. Lian, *Journal of the American Ceramic Society*, 2015. **98**(10): p. 3361-3366.
28. T. Chilalla, and J. Mendel. *Ceramics in nuclear waste management*. in *Proceedings of an Intern. Symp. held in Cincinnati, Ohio, 30 Apr.-2 May 1979, sponsored in part by the Nucl. Div. of the Am. Ceram. Soc.* 1979.
29. W. J. Weber, J. Wald, and H. Matzke, *Journal of Nuclear Materials*, 1986. **138**(2): p. 196-209.
30. L. L. Hench, D. E. Clark, and E. L. Yen-Bower, *Nuclear and Chemical Waste Management*, 1980. **1**(1): p. 59-75.
31. W. J. Weber, and L. M. Wang, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 1994. **91**(1): p. 63-66.
32. W. J. Weber, L. M. Wang, and N. Yu, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 1996. **116**(1): p. 322-326.
33. W. J. Weber, R. C. Ewing, and A. Meldrum, *Journal of Nuclear Materials*, 1997. **250**(2): p. 147-155.
34. R. Devanathan, K. E. Sickafus, W. J. Weber, and M. Nastasi, *Journal of nuclear materials*, 1998. **253**(1): p. 113-119.
35. A. Najib, J. E. H. Sansom, J. R. Tolchard, P. R. Slater, and M. S. Islam, *Dalton Transactions*, 2004(19): p. 3106-3109.
36. A. Orera, E. Kendrick, D. C. Apperley, V. M. Orera, and P. R. Slater, *Dalton Transactions*, 2008(39): p. 5296-5301.
37. J. E. H. Sansom, J. R. Tolchard, M. S. Islam, D. C. Apperley, and P. R. Slater, *Journal of Materials Chemistry*, 2006. **16**(15): p. 1410-1413.
38. C. Meis, *Journal of nuclear materials*, 2001. **289**(1): p. 167-176.
39. S. X. Wang, L. M. Wang, and R. C. Ewing, *Physical Review B*, 2000. **63**(2): p. 024105.
40. W. J. Weber, R.C. Ewing, and L. M. Wang, *Journal of Materials Research*, 1994. **9**(03): p. 688-698.
41. Y. Q. Shen, A. Tok, and Z. L. Dong, *Journal of the American Ceramic Society*, 2010. **93**(4): p. 1176-1182.
42. I. Brown, *The chemical bond in inorganic chemistry, the bond valence model. IUCr monographs on crystallography 12. International Union of Crystallography*. 2002, Oxford Science Publications, Oxford.
43. G. Blasse, *Journal of Solid State Chemistry*, 1975. **14**(2): p. 181-184.

Caption of Figures:

Fig. 1 Transmission electron microscopy images of (a) $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ and (b) $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$

Fig. 2 Room temperature TEM dark field images and SAED patterns show the 1 MeV Kr^{2+} ion beam irradiation induced amorphization process of the $\text{Mg}_2\text{Y}_{7.5}\text{Ce}_{0.5}(\text{SiO}_4)_6\text{O}_2$ silicate apatite. (a) unirradiated, (b) 0.032 dpa (1×10^{14} ions/cm²), (c) 0.064 dpa (2×10^{14} ions/cm²), (d) 0.096 dpa (3×10^{14} ions/cm²);

Fig. 3 Room temperature TEM dark field images and SAED patterns show the 1 MeV Kr^{2+} ion beam irradiation induced amorphization process of the $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$ silicate apatite. (a) unirradiated, (b) 0.032 dpa (1×10^{14} ions/cm²), (c) 0.064 dpa (2×10^{14} ions/cm²), (d) 0.096 dpa (3×10^{14} ions/cm²);

Fig. 4 SAED patterns of $\text{Mg}_2\text{Y}_{7.95}\text{Ce}_{0.05}(\text{SiO}_4)_6\text{O}_2$ silicate apatite showing (a) crystallinity at 0.16 dpa (5×10^{14} ions/cm²) at an elevated temperature of 798 K and (b) amorphization at 0.096 dpa (3×10^{14} ions/cm²) at room temperature.

Fig. 5 (a) Temperature dependence of D_c for Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite; (b) T_c dependence on Ce content in Ce doped $\text{Mg}_2\text{Y}_8(\text{SiO}_4)_6\text{O}_2$ silicate apatite

Fig. 1

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