## **List of Figures**

Figure 1.1 The Classification of crystalline ceramic compounds as suitable waste forms for
radioactive waste3
Figure 1.2 Pyrochlore structure, A <sub>2</sub> B <sub>2</sub> O <sub>7</sub> , exhibits the 8-fold coordination and 6- fold
coordination polyhedron [12]6
Figure 1.3 The structural transition from pyrochlore structure A <sub>2</sub> B <sub>2</sub> O <sub>6</sub> O' or A <sub>2</sub> B <sub>2</sub> X <sub>6</sub> Y to ideal
fluorite structure [12]6
Figure 1.4 The periodic table highlights elements with oxidation states 3+ (red) and 4+(blue)
which may help to form the pyrochlore phase [30]7
Figure 1.5 Representation of the crystal structure of La <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> pyrochlore. The magenta and
light green are ZrO <sub>6</sub> trigonal antiprisms and LaO <sub>8</sub> cubes or scalenohedra13
Figure 2.1 Schematic diagram of the synthesis process of La <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> and Gd <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> pyrochlore
oxides27
Figure 2.2 Schematic diagram of irradiation process of zirconate pyrochlore oxides (La <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub>
and Gd <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> )29
Figure 2.3 The real photograph of low energy ion beam controlling unit (a), irradiation unit
(b), samples loaded target ladder (c), and (d) scanning ion beam during ion irradiation29
Figure 2.4 Represents the schematic diagram of different components of an XRD. The dotted
circles represent the goniometer where the sample is placed in the center32
Figure 2.5 Schematic representation of the configuration of GIXRD33
Figure 2.6 Schematic representation of electron-sample interaction in an FE-SEM35
Figure 2.7 Illustration of Rayleigh, Stokes, and anti-Stokes scattering36
Figure 2.8 Schematic diagram of a Raman spectrometer
Figure 2.9 Schematic representation of electron-sample interaction in a TEM38
Figure 3.1 Representation of the XRD patterns of LZO (a) and (b) illustrates the magnified
view of (111) diffraction plane as a function of annealing temperature and time44
Figure 3.2 Depiction of (a) crystallite size and (b) lattice strain at various annealing
temperatures for 96 h45
Figure 3.3 Rietveld refinement of the LZO samples annealed at three different temperatures,
i.e., 1200°C, 1300°C, and 1500°C for 96 h
Figure 3.4 Dependence of lattice parameters (a) and (b) X-ray density of LZO samples on
annealing temp, and time

Figure 3.5 Dependence of degree of x <sub>48f</sub> and cation ordering on annealing temperature and
time
Figure 3.6 FE-SEM micrographs of LZO samples annealed at three different temperatures and
times
Figure 3.7 Dependence of grain size on annealing temperature and time (a); representation of
(b) $ln (G_t^2-G_0^2)$ vs $ln (t)$ and (c) Arrhenius plot
Figure 3.8 Evolution of grain growth of LZO samples as a function of annealing temp. and
time
Figure 4.1 FE-SEM micrograph of virgin LZO sample (a) and (b) size histogram58
Figure 4.2 Illustration of $S_e$ and $S_n$ plot (a) and $S_n/S_e$ ratio plot in respect of depth of ion
irradiated LZO
Figure 4.3 GI-XRD patterns of before and after irradiated LZO samples (a, d); enlarged view
of (222) reflection (b, e) and (c, f) superstructure reflections, i.e., (331) and (511)60
Figure 4.4 Illustration of intensity (a), FWHM (b), and (c) strain with the enhancement of ion
fluence
Figure 4.5 Represents the GIXRD pattern of virgin LZO (a), (b) calculated pattern, and (c)
disparity of observed and calculated pattern; (d) crystal structure of LZO63
Figure 4.6 Rietveld refinement of the post irradiated LZO samples at $\sim\!88$ K (a-c) and (d-f) 300 kg.
K with the fluence of $1.0 \times 10^{13}$ , $5.0 \times 10^{13}$ , and $1.0 \times 10^{14} \text{ ions/cm}^2$ 63
Figure 4.7 Raman spectra of irradiated LZO sample (a) ~88 K and (b) 300 K; (c-d) illustrates
the deconvoluted Raman spectra of the LZO sample (1.0E13 ions/cm²)
Figure 4.8 Represents the FWHM (a) and (b) intensity of vibrational modes with the function
of fluence at two different temperatures
Figure 4.9 HR-TEM image of (a) un-irradiated, (b) irradiated@300K, and (c)
irradiated@~88K LZO sample
Figure 5.1 FE-SEM micrograph (a) and (b) size distribution histogram of un-irradiated LZO
sample76
Figure 5.2 GIXRD patterns of LZO (a, b); enlarged view of (331), and (511) reflections and
(c-d) magnified of (222) reflection
Figure 5.3 Representation of (a) intensity, (b) FWHM, (c) lattice swelling, and (d) strain of
LZO samples in respect of ion fluence
Figure 5.4 Rietveld refinement of pristine LZO (a) and representation of crystal structure of
LZO, produced from the CIF file
Figure 5.5 Refined GIXRD patterns of LZO (a-c) low temperature (88 K) and (d-f) 300 K. 80

Figure 5.6 (a-c) Dependence of cell volume expansion, cation disordering, and (c) X <sub>48f</sub> on
fluence at 88 K and 300 K
Figure 5.7 Raman spectra of LZO (a) 300 K, (b) 88 K, and (c-d) deconvoluted Raman spectra
of LZO84
Figure 5.8 Illustration of bending and stretching vibrations in LZO samples
Figure 5.9 (a-d) Dependence of intensity and FWHM on fluence at 300 K and 88 K
Figure 6.1 XRD patterns of GZO samples sintered at (a) 1400°C and (b) 1500°C96
Figure 6.2 Variation in the intensity (a) and (b) FWHM of GZO14 and GZO15 samples upon
sintering at 1400°C and 1500°C; and (c) Williamson-Hall plot of GZO15 sample98
Figure 6.3 Rietveld refined XRD pattern of (a) GZO14 and (b) GZO15 samples. In Fig. (a-b)
the top and bottom magenta color lines represent the peak positions for the pyrochlore and
fluorite phase, respectively
Figure 6.4 FE-SEM image of (a) GZO14 and (b) GZO15 samples sintered at 1400°C and
1500°C
Figure 6.5 Deconvoluted Raman modes of (a) GZO14 and (b) GZO15 samples; (c-d) variation
in the intensity and FWHM of Raman modes of GZO14 and GZO15 samples102
Figure 6.6 SRIM simulated collision events and ion ranges (a-b) and (c) variation of $S_e$ and $S_n$
as a function of depth of GZO15 sample
Figure 6.7 XRD patterns of (a) GZO14 and (b) GZO15 samples upon irradiation of 100 MeV
$I^{7+}$ at the fluence of $1.0 \times 10^{14}$ ions/cm <sup>2</sup>
Figure 6.8 Deconvoluted Raman spectra of (a) GZO14 and (b) GZO15 upon irradiation of 100
MeV iodine ions
Figure 7.1 XRD patterns (a), (b) W-H plot, and (c) Rietveld refinement of the pristine GZO
sample. The superstructure reflections are marked with an asterisk (*)118
Figure 7.2 FE-SEM micrograph of pristine GZO sample
Figure 7.3 XRD patterns of GZO sample before and after irradiation with $100~\text{MeV}~\text{I}^{7+}$ ions as
a function of ion fluence.
Figure 7.4 Variation of fluorite phase fraction as a function of ion fluence irradiated with 100
MeV I <sup>7+</sup> ions
Figure 7.5 Raman spectra of pristine and post irradiated GZO samples
Figure 7.6 Deconvoluted Raman spectra of pristine and post irradiated GZO samples 124
Figure 8.1 Illustrates the impact of irradiation temperature and ion fluence on the degradation
of crystallinity (damage) of La <sub>2</sub> Zr <sub>2</sub> O <sub>7</sub> samples134

of 48f oxygen with function on ion fluence
List of tables
$\textbf{Table 2.1} \ \text{Represents the irradiation parameters of irradiated } La_2Zr_2O_7 \ pyrochlore30$
$\textbf{Table 2.2} \ \text{The simulated value of } S_e,  S_n,  \text{and}   R_p \ \text{for zirconated pyrochlore oxides for } Xe^{4^+}  \text{and} $
I <sup>7+</sup> ions31
Table 3.1 Depicts the Rietveld refined "figure of merit" parameters (R <sub>B</sub> , R <sub>wp</sub> , R <sub>exp</sub> , and $\chi^2$ ). 46
Table 4.1 Summarized the damage quantified from the GIXRD and Raman spectroscopy of
LZO samples.
Table 4.2 Structural parameters of the pre and post-irradiated LZO samples. 64
Table 4.3 Depicts the vibrational mode frequencies and assigned vibrational mode of pre and
post irradiated LZO samples. 67
Table 5.1 Structural parameters of pre and post-irradiated LZO samples. 81
Table 5.2 Raman frequencies and assigned modes of LZO samples before and after irradiation.
85
Table 6.1 Displays the variation in the intensity of pyrochlore superstructure peaks in respect
of (222) diffraction plane for GZO14 and GZO15 samples
$\textbf{Table 6.2}. \ \textbf{Structural parameters (space group, lattice parameters,} \ \chi^2, \ \textbf{crystallite size, and lattice}$
strain) were quantified from the Rietveld refinement and W-H plots of pyrochlore structure
GZO samples

Figure 8.2 Variation of superlattice reflections, cell volume, cation disorder, and 'x' parameter