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**Table I.** Molar compositions of (Am-La) and (Nd-La) glass- ceramics and Base glass.

	Composition in mol.%								
	SiO <sub>2</sub>	B <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	CaO	La <sub>2</sub> O <sub>3</sub>	Am <sub>2</sub> O <sub>3</sub>	Nd <sub>2</sub> O <sub>3</sub>	R
Am-La sample theoretical	56.60	10.19	16.12	4.99	8.21	1.95	1.95	—	0.50
Am-La sample corrected <sup>†</sup>	56.77	10.23	16.16	5.01	8.23	1.95	1.65	—	0.54
Nd-La sample	56.60	10.20	16.12	4.99	8.20	1.95	—	1.95	0.50
Base glass	58.89	10.61	16.77	5.19	8.51	—	—	—	

<sup>†</sup>Corrected to the Am content measured by Calorimetry  
R = La/(La+Am) or La/(La+Nd)

**Table II.** Cell parameters computed from the XRD Patterns of (Am-La) and (Nd-La) glass-ceramics and  $\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$  ceramic.

Cell Parameters	(Am-La)	(Nd-La)	$\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$
a (nm)	0,960(2)	0,961(3)	0,959(1)
c (nm)	0,707(1)	0,707(1)	0,708(1)
V (nm <sup>3</sup> )	0,564(5)	0,565(6)	0,564(3)

**Table III.** Electron Probe Microanalysis of (Am-La) and (Nd-La) residual glass matrix (mol. %).

	Residual glass composition from EPMA (mol. %)								$R_{\text{residual matrix}}^{\dagger}$
	SiO <sub>2</sub>	B <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	CaO	Am <sub>2</sub> O <sub>3</sub>	Nd <sub>2</sub> O <sub>3</sub>	La <sub>2</sub> O <sub>3</sub>	Matrix
(Am-La) sample	57.21	9.67	18.24	4.90	7.30	1.07	—	1.61	0.60
(Nd-La) sample	57.04	9.27	17.15	5.14	7.62	—	1.86	1.92	0.51

$^{\dagger}R = \text{La}/(\text{La}+\text{Am})$  or  $\text{La}/(\text{La}+\text{Nd})$

**Table IV.** Electron Probe Microanalysis of (Am-La) and (Nd-La) crystalline phases (atomic %). SiO<sub>4</sub> tetrahedra's stoichiometry was fixed at 6 in order to present results according to the stoichiometry Ca<sub>w</sub>Ln<sub>x</sub>Ac<sub>y</sub>(SiO<sub>4</sub>)<sub>6</sub>O<sub>z</sub>.

	Crystals composition from EPMA (atomic %)					R <sub>crystal</sub> <sup>†</sup>
	Ca	La	Nd	Am	O	Crystal
Am-La sample	2.02	3.96	—	4.07	1.99	0.49
Nd-La sample	2.81	3.62	3.57	—	1.59	0.50
Theoretical	2	4	4		2	0.50

<sup>†</sup>R=La/(La+Am) or La/(La+Nd)

**Table V.** SiO<sub>4</sub> Q<sup>n</sup> fitted areas of (Am-La) and (Nd-La) residual glass and Base glass Raman spectra.

	Raman shift (cm <sup>-1</sup> )				
	933 Q <sup>2</sup> <sub>Ln/Ac</sub>	970 Q <sup>2</sup>	998 Q <sup>3</sup> <sub>Ln/Ac†</sub>	1070 Q <sup>3</sup>	1148 Q <sup>4</sup>
Base glass	—	17	—	72	11
(Am-La) residual glass	11	—	28	56	5
(Nd-La) residual glass	8	—	36	53	3

†Ln: lanthanides, Ac: actinides

**Table VI.** Characteristics of the  $\nu_1$  Raman motion of crystals formed in (Am-La) and (Nd-La) glass-ceramics and  $\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$  ceramic reference.

	(Am-La) crystals	(Nd-La) crystals	$\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$ ceramic reference
Raman $\nu_1$ position ( $\text{cm}^{-1}$ )	852(1)	854(1)	857(1)
Raman $\nu_1$ FWHM ( $\text{cm}^{-1}$ )	29(2)	23(1)	15(2)

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White and dark phases correspond to crystals and residual glass, respectively.

**Fig. 4.** a) (Am-La) GC and b) (Nd-La) GC and  $\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$  ceramic reference X-ray diffraction patterns.

\*Additional reflections from Pt residues from the crucible contribute to the pattern of the (Nd-La) sample.

**Fig. 5.** (a) Raman spectra of base glass (lanthanide-free reference), (Am-La) and (Nd-La) residual glass matrices.

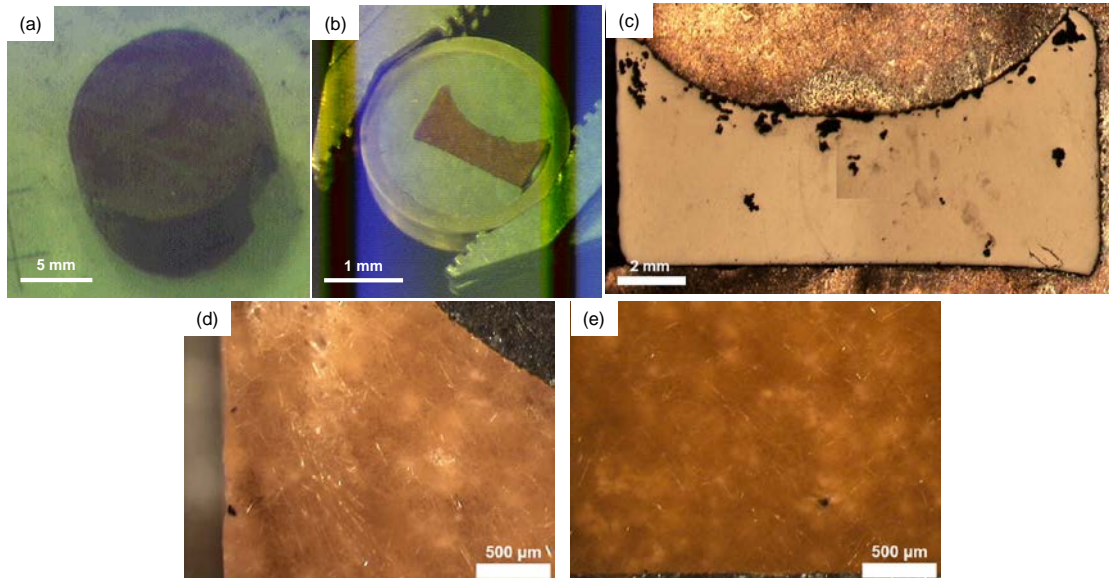
Base Glass: a)  $\text{SiO}_4\text{Q}^2$  ( $970\text{ cm}^{-1}$ ), b)  $\text{SiO}_4\text{Q}^3$  ( $1070\text{ cm}^{-1}$ ), c)  $\text{SiO}_4\text{Q}^4$  ( $1148\text{ cm}^{-1}$ ),

(Am-La) and (Nd-La) residual glass: d)  $\text{Q}^1/\text{Q}^2_{\text{Ln/Ac}}$  ( $933\text{ cm}^{-1}$ ), e)  $\text{Q}^2/\text{Q}^3_{\text{Ln/Ac}}$  ( $998\text{ cm}^{-1}$ ), b)  $\text{SiO}_4\text{Q}^3$  ( $1070\text{ cm}^{-1}$ ), and c)  $\text{SiO}_4\text{Q}^4$  ( $1148\text{ cm}^{-1}$ ).

(b) Raman spectra of (Am-La) and (Nd-La) crystals.

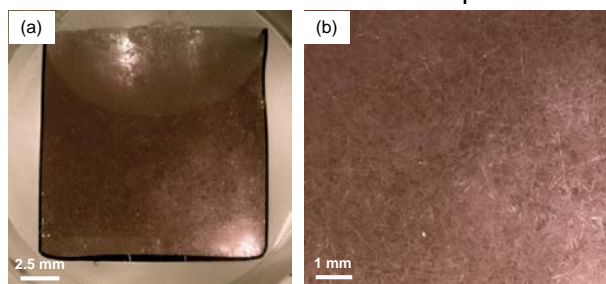


Radioactive Am-La sample

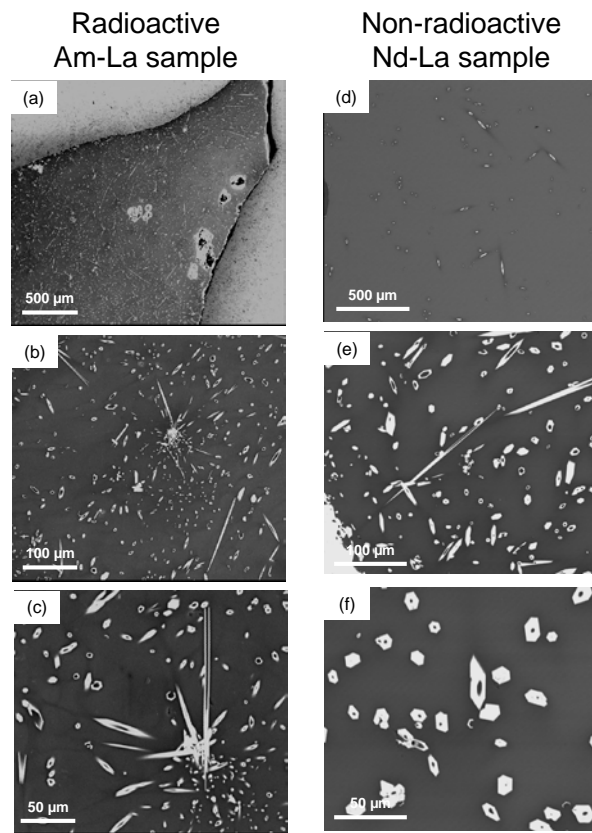


**Fig. 1.** (Am-La) GC optical microscopy images.

Non-radioactive Nd-La sample

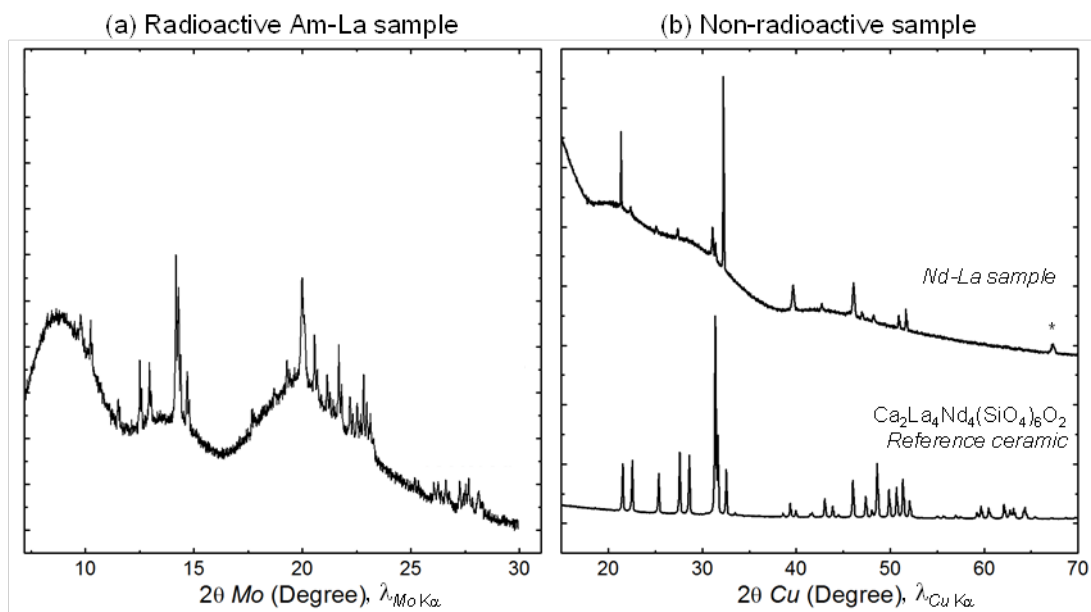


**Fig. 2.** (Nd-La) GC optical microscopy images.



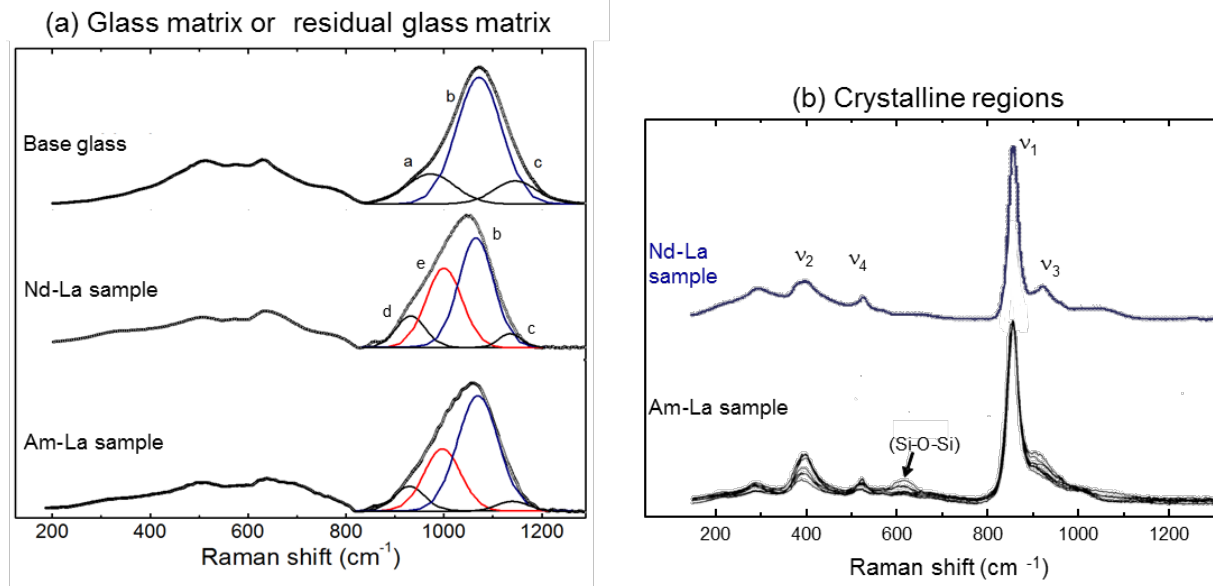
**Fig. 3.** (a-c) (Am-La) GC and (d-f) (Nd-La) GC SEM micrographs (backscattered electrons).

White and dark phases correspond to crystals and residual glass, respectively.



**Fig. 4.** a) (Am-La) GC and b) (Nd-La) GC and  $\text{Ca}_2\text{La}_4\text{Nd}_4(\text{SiO}_4)_6\text{O}_2$  ceramic reference X-ray diffraction patterns.

\*Additional reflections from Pt residues from the crucible contribute to the pattern of the (Nd-La) sample.



**Fig. 5.** (a) Raman spectra of base glass (lanthanide-free reference), (Am-La) and (Nd-La) residual glass matrices.

Base Glass: a)  $\text{SiO}_4\text{Q}^2$  ( $970\text{ cm}^{-1}$ ), b)  $\text{SiO}_4\text{Q}^3$  ( $1070\text{ cm}^{-1}$ ), c)  $\text{SiO}_4\text{Q}^4$  ( $1148\text{ cm}^{-1}$ ),

(Am-La) and (Nd-La) residual glass: d)  $\text{Q}^1/\text{Q}^2_{\text{Ln/Ac}}$  ( $933\text{ cm}^{-1}$ ), e)  $\text{Q}^2/\text{Q}^3_{\text{Ln/Ac}}$  ( $998\text{ cm}^{-1}$ ), b)  $\text{SiO}_4\text{Q}^3$  ( $1070\text{ cm}^{-1}$ ), and c)  $\text{SiO}_4\text{Q}^4$  ( $1148\text{ cm}^{-1}$ ).

(b) Raman spectra of (Am-La) and (Nd-La) crystals.