## SUPPORTING INFORMATION

## Pressure-Controlled Synthesis of High-Performance

## SrLiAl<sub>3</sub>N<sub>4</sub>:Eu<sup>2+</sup> Narrow-Band Red Phosphors

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## **EXPERIMENTAL METHODS**

**Reagents.** Strontium nitride ( $Sr_3N_2$ , 99.5%) and europium nitride (EuN, 99.9%) were obtained from Materion. Aluminum nitride (AIN, 97%) was acquired from Tokuyama Corporation. Lithium nitride ( $Li_3N$ , 99%) was supplied by Kojundo Chemical Laboratory.

**Synthesis of SrLiAl<sub>3</sub>N<sub>4</sub>:Eu<sup>2+</sup>.** Eu<sup>2+</sup>-doped SrLiAl<sub>3</sub>N<sub>4</sub> red phosphor was synthesized through a solid-state reaction. Stoichiometric amounts of Sr<sub>3</sub>N<sub>2</sub>, Li<sub>3</sub>N, EuN, and AIN were carefully ground in a glove box ( $O_2 < 1$  ppm,  $H_2O < 1$  ppm). The well-mixed precursors were loaded into BN crucibles, which were subjected to hot isostatic pressing (HIP) and gas pressure sintering (GPS). The pressure was increased up to the set point by using a compressor. The furnace was heated to 1,000 °C under different pressure levels for 4 h. The furnace was cooled to room temperature, and the pressure was released to atmospheric pressure. Finally, the as-prepared samples were ground and kept in the glove box for characterization.

Characterization. The phase and purity of the as-prepared powder samples were examined by X-ray diffraction (XRD) analysis using a D2 PHASER diffractometer (Bruker) with CuK $\alpha$  radiation source ( $\lambda$  = 1.5418 Å). Fine structure data including lattice parameters and atom position were obtained by synchrotron X-ray diffraction analysis ( $\lambda$  = 0.774910 Å) of BL01C2 beamline with a Debye–Scherrer camera at the National Synchrotron Radiation Research Center (NSRRC), Taiwan. Rietveld refinement was conducted with Total Pattern Analysis Solutions (TOPAS) 4.2 software. Photoluminescence properties were analyzed by a 150 W Xe lamp and a photomultiplier tube equipped with a FluoroMax-3 Hamamatsu R928 spectrophotometer (HORIBA, Japan). Morphologies of the samples were examined by a scanning electron microscope (SEM, JSM-6700F). The ratio of Eu<sup>2+</sup>/Eu<sup>3+</sup> was examined by Eu L<sub>3</sub>-edge of X-ray absorption near-edge structure spectroscopy in florescence mode at BL17 beamline in NSRRC, Taiwan. Internal quantum efficiency, absorption, and external quantum efficiency were determined using an absolute PL quantum yield spectrometer (QY C11347, Hamamatsu, Japan). The ratio of atomic composition was determined through ICP-AES analysis (Agilent 725-ES ICP-AES, USA).



Figure S1. Photoluminescence excitation spectra of SLA synthesized under different gas pressure levels.



Figure S2. SEM images of SLA synthesized at 0.3, 0.5, 0.7, 0.9, and 50 MPa.

Space group	PĪ						
Cell Parameters	$ \begin{array}{ll} a &= 5.8611(3) \ \text{\AA} & \alpha = 83.602(2)^{\circ} \\ b &= 7.4808(4) \ \text{\AA} & \beta = 76.758(2)^{\circ} \\ c &= 9.9505(3) \ \text{\AA} & \gamma = 79.570(3)^{\circ} \end{array} \\ \end{array} \\ V = 416.07(4) \ \text{\AA}^{3} \\ \end{array} $						
Reliability Factors	$\chi^2 = 1.89$ $R_{wp} = 7.94 \%$ $R_p = 5.77 \%$						
Atoms	X	у	Z	Frac	Beq ( $Å^2$ )		
Sr1	0.0019(21)	0.1334(17)	0.1286(12)	0.990(11)	1.0		
Sr2	0.0186(17)	0.3810(17)	0.3715(12)	0.991(11)	1.0		
Al1	0.5469(32)	0.9527(28)	0.1240(25)	0.953(39)	1.0		
Al2	0.1818(32)	0.9518(29)	0.3991(25)	1.000(41)	1.0		
Al3	0.1670(34)	0.7066(31)	0.1532(27)	1.000(36)	1.0		
Al4	0.8247(30)	0.5563(29)	0.1035(24)	1.000(40)	1.0		
Al5	0.4711(36)	0.5580(38)	0.3741(28)	1.000(31)	1.0		
Al6	0.5645(35)	0.2044(37)	0.3665(29)	1.000(41)	1.0		
N1	0.3657(91)	0.8568(78)	0.0183(61)	1.000(78)	1.0		
N2	0.3729(98)	0.3408(87)	0.5234(57)	1.000(73)	1.0		
N3	0.3703(95)	0.0883(82)	0.2677(64)	1.000(81)	1.0		
N4	0.1556(81)	0.9425(74)	0.5917(59)	1.000(71)	1.0		
N5	0.8437(80)	0.7921(75)	0.1431(63)	1.000(80)	1.0		
N6	0.1906(77)	0.4968(70)	0.0779(57)	1.000(65)	1.0		
N7	0.1773(79)	0.6962(69)	0.3484(61)	1.000(75)	1.0		
N8	0.6538(73)	0.4091(67)	0.2286(56)	1.000(81)	1.0		
Li1	0.444(20)	0.328(16)	0.127(14)	1.00(15)	1.0		
Li2	0.201(19)	0.192(18)	0.647(16)	1.00(15)	1.0		

Table S1. Rietveld refinement results of sample fabricated at 0.1 MPa.

Space group	Pī						
Cell Parameters	$ \begin{array}{ll} a &= 5.8620(4) \ \text{\AA} & \alpha = 83.652(3)^{\circ} \\ b &= 7.4770(4) \ \text{\AA} & \beta = 76.652(4)^{\circ} \\ c &= 9.9379(5) \ \text{\AA} & \gamma = 79.442(4)^{\circ} \end{array} \\ \end{array} \\ V = 415.55 \ (4) \ \text{\AA}^{3} \\ \end{array} $						
Reliability	$\chi^2 = 2.15$						
Factors	$R_{wp} = 8.35 \%$ $R_p = 5.62 \%$						
Atoms	Х	у	Z	Frac	Beq(Å <sup>2</sup> )		
Sr1	0.0079(27)	0.1317(22)	0.1280(16)	1.000(13)	1.0		
Sr2	0.0148(24)	0.3773(24)	0.3715(16)	1.000(13)	1.0		
Al1	0.5426(70)	0.9609(59)	0.1296(41)	1.000(52)	1.0		
Al2	0.1779(61)	0.9407(63)	0.4007(49)	1.000(63)	1.0		
Al3	0.1694(81)	0.7025(57)	0.1484(56)	0.992(45)	1.0		
Al4	0.8148(68)	0.5552(64)	0.1108(47)	1.000(62)	1.0		
Al5	0.4778(64)	0.5575(58)	0.3756(49)	1.000(42)	1.0		
Al6	0.5746(54)	0.2018(57)	0.3693(44)	1.000(55)	1.0		
N1	0.368(13)	0.859(13)	0.026(10)	1.00(12)	1.0		
N2	0.348(13)	0.355(13)	0.520(10)	1.00(11)	1.0		
N3	0.370(14)	0.091(12)	0.264(10)	1.00(12)	1.0		
N4	0.150(12)	0.948(14)	0.5795(95)	1.000(97)	1.0		
N5	0.864(11)	0.794(13)	0.154(10)	1.00(10)	1.0		
N6	0.211(12)	0.458(12)	0.081(10)	1.00(10)	1.0		
N7	0.199(11)	0.708(14)	0.3530(96)	1.000(96)	1.0		
N8	0.634(15)	0.403(13)	0.236(10)	1.00(14)	1.0		
Li1	0.457(25)	0.324(23)	0.121(21)	1.00(21)	1.0		
Li2	0.182(23)	0.193(27)	0.643(23)	1.00(17)	1.0		

Table S2. Rietveld refinement results of sample fabricated at 100 MPa.