

Supporting information attached to

Tracing Nitridation Reaction Toward Efficient Production of Oxynitride Glasses as Hosts for Bright Luminescence Centers

Xun Liu^{1,2}, Takeo Ohsawa¹, Noriko Saito¹, Kohsei Takahashi¹, Takashi Takeda¹, Kenzo Deguchi¹, Shinobu Ohki¹, Tetsuo Kishi³, Tetsuji Yano³, Hiroyo Segawa^{1,3}, Naoki Ohashi^{1,2,4}

¹ National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

² Interdisciplinary Graduate School of Engineering Sciences, Kyusyu University, 6-1, Kasugakoen, Kasuga, Fukuoka 816-8580, Japan

³ Department of Chemistry and Materials Science, Graduate School of Science and Engineering, Institute of Science Tokyo, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8552, Japan

⁴ Materials DX Research Center for Element Strategy, Institute of Science Tokyo, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan

I. Phase identification

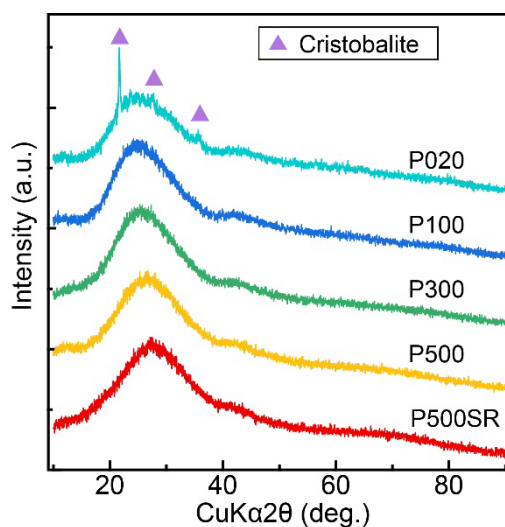


Figure S1 X-ray diffraction pattern for the powder after ammonolysis with various ammonia flow rate. Sample IDs are show in main text. Crystalline phase, cristobalite, was formed in the P020 powder prepared by ammonolysis under small ammonia flow rate (20cm³/min).

II. Atmosphere in the furnace during ammonolysis

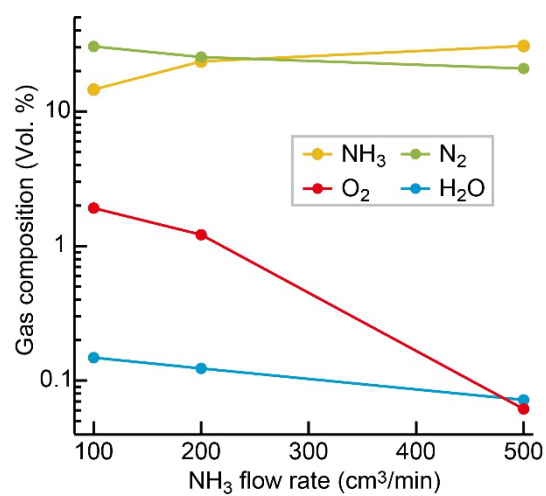


Figure S2 Composition of the exhaust gas from the furnace during ammonolysis. With smaller ammonia flow rate, actual ammonia concentration in the furnace was relatively low due to decomposition and the residual oxygen gas concentration due to out-gas from the furnace was relatively high. With very high flow rate, relatively high ammonia concentration was achieved.

III. Chemical composition analysis

A. Typical results of XRF analyses

Table S1 Result of Xray fluorescence analysis for PwO powder (prepared by firing in the air). Charge compensation in the powder was considered by assuming formal charge (Q) of each element. Particularly, formal charge of europium was assumed to be divalent. The summation of charge indicates slight excess of anion, likely presence of protons for compensation.

Element	Mol% (M)	Formal charge (Q)	Charge (Q×M)
N	0.0	-3	0.0
O	62.9	-2	-125.8
Al	4.2	3	12.6
Si	24.5	4	98.2
Sr	2.8	2	5.6
Eu	0.8	2	1.7
Sum. charge			-7.8

Table S2 Result of Xray fluorescence analysis for P500SR powder (prepared by ammonolysis in ammonia flow rate at 500 ml/min). Charge compensation in the powder was considered by assuming formal charge (Q) of each element. Particularly, formal charge of europium was assumed to be divalent. The obvious excess of negative charge is an indication for the presence of proton in the powder.

Element	Mol% (M)	Formal charge (Q)	Charge (Q×M)
N	26.4	-3	-79.3
O	42.8	-2	-85.6
Al	4.1	3	12.4
Si	23.1	4	92.3
Sr	2.8	2	5.5
Eu	0.8	2	1.7
Sum. charge			-53.1

B. N_{eff} determined by XPS measurements

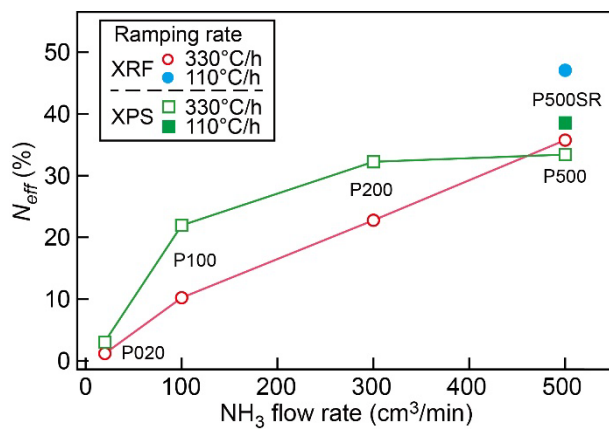


Figure S3 Effective nitrogen concentration (N_{eff}) in the obtained powder as a function of ammonia flow rate on ammonolysis. In this figure, the results of X-ray photoemission (XPS) analyses are superimposed on the results of X-ray fluorescence (XRF) analyses show in the main text. The definition of N_{eff} and sample IDs are also shown in main text.

IV. Microstructures

A. Microstructure of the gel before ammonolysis

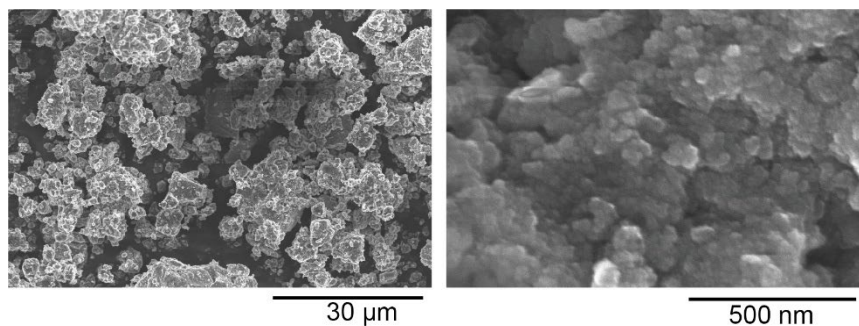


Figure S4 Secondary electron microscope image of the gel powder before ammonolysis.

B. Microstructures of the powder after ammonolysis

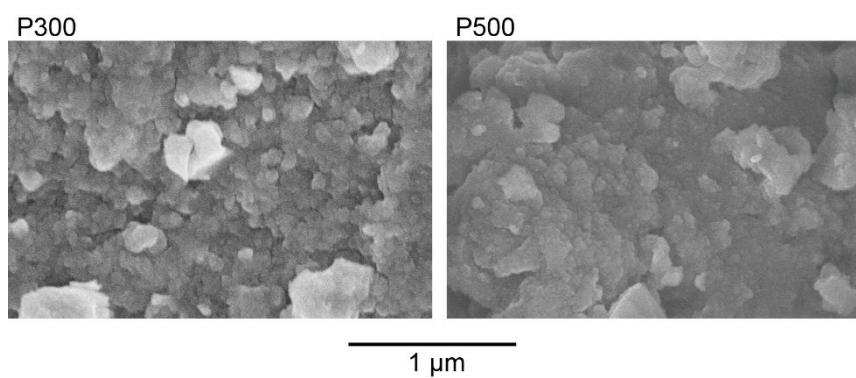


Figure S5 Secondary electron microscope image of the gel powder after ammonolysis at the ammonia flow rate of 300 cm³/min (P300) and 500 cm³/min (P500).