

## Supplementary Information

### Removal of $^{90}\text{Sr}$ from highly $\text{Na}^+$ -rich liquid nuclear waste with a layered vanadosilicate

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## Experimental Section

**Materials:** Sodium silicate solution I (17-19% Na<sub>2</sub>O, and 35-38% SiO<sub>2</sub>, Kanto), sodium silicate solution II (10.6% Na<sub>2</sub>O, and ~26.5% SiO<sub>2</sub>, Sigma-Aldrich), sodium silicate solution III (27% SiO<sub>2</sub>, 8% Na<sub>2</sub>O, Merck), tetraethyl orthosilicate (Si(OC<sub>2</sub>H<sub>5</sub>)<sub>4</sub>, 98%, Acros), vanadium (V) oxide (V<sub>2</sub>O<sub>5</sub>, 99%, Aldrich), titanium isopropoxide [Ti(OC<sub>3</sub>H<sub>7</sub>)<sub>4</sub>, 98%, Junsei], TiCl<sub>3</sub> solution (20% in 3% HCl, Alfa Aesar), TiCl<sub>3</sub> solution (15% m/m solution of TiCl<sub>3</sub> in 10% m/m HCl, Merck), copper sulfate pentahydrate (CuSO<sub>4</sub>·5H<sub>2</sub>O, 99%, Alfa-Aesar), oxalic acid dihydrate (H<sub>2</sub>O<sub>4</sub>C<sub>2</sub>·2H<sub>2</sub>O 99%, Sigma-Aldrich), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>, 95%, Duksan), hydrochloric acid (HCl, 36.5-38.0%, Sigma-Aldrich), sodium hydroxide (NaOH, 93%, Samchun), sodium chloride (NaCl, 99%, Samchun), potassium chloride (KCl, 99%, Oriental), potassium fluoride (KF, 95%, Samchun), strontium chloride hexahydrate (SrCl<sub>2</sub>·6H<sub>2</sub>O, 98.5%, Samchun), potassium carbonate (K<sub>2</sub>CO<sub>3</sub>, 99%, Aldrich) manganese powder, (Mn, 325 mesh, 99.3%, metals basis, Alfa Aesar), tin powder (Sn, < 150 μm, 99.5% trace metals basis, Sigma-Aldrich), sulfur powder (S, 98%, Junsei), sodium tetraphenylborate (NaTPB, 99.5%, Acros), ethylenediaminetetracetic acid disodium salt dihydrate (EDTA, 99-101%, Sigma-Aldrich), ethanol (EtOH, 95%, SK), phlogopite mica (K-PMica, near Perth, Ontario, Canada), were purchased and used without further purification.

**Synthesis of Na-SGU-7:** A gel consisting of Na<sub>2</sub>SiO<sub>3</sub>, V<sub>2</sub>O<sub>5</sub>, oxalic acid, NaOH, NaCl and H<sub>2</sub>O (distilled and deionized water) was prepared, where the mole ratios of the synthesis gels in terms of SiO<sub>2</sub>:V<sub>2</sub>O<sub>5</sub>:H<sub>2</sub>C<sub>2</sub>O<sub>4</sub>:Na<sub>2</sub>O:NaCl:H<sub>2</sub>O were 5.49:1:3.67:5.30:9.99:365. The gel was prepared as follows. *Preparation of silicon source solution:* A NaOH solution (3 g NaOH and 20 g of H<sub>2</sub>O) was added into sodium silicate solution composed of 12.2 g of Na<sub>2</sub>SiO<sub>3</sub> (17-19% Na<sub>2</sub>O, and 35-38%, SiO<sub>2</sub>, Kanto) and 12 g of water. A NaCl solution (8 g of NaCl and 33 g of water) was added into the above mixture and the mixture was stirred vigorously. *Preparation of vanadium source solution:* Oxalic acid (6.4 g) was added into a 100-mL round bottomed flask containing water (25 g) and heated until oxalic acid is dissolved. Subsequently, V<sub>2</sub>O<sub>5</sub> (2.5 g) was added into the flask. The heterogeneous mixture was heated for 20 min, during which the yellow V<sub>2</sub>O<sub>5</sub> powder dissolved completely into the solution, and the solution turned greenish blue. The greenish blue V source solution was then cooled to room temperature and subsequently added into sodium silicate solution in a dropwise manner. The mixture was extremely sticky; therefore the mixture was homogenized using a spatula and continued for stirring. After aging for 15 h at room temperature, the gel was transferred into a 50 ml Teflon-lined autoclave, and placed in preheated oven at 220 °C for 40 h under a static condition. The purple crystals were collected by centrifugation and washed with copious amounts of water and finally washed with ethanol and dried at 80 °C for 3 h.

**Synthesis of Na-CST (Sodium crystalline silicotitanate,  $\text{Na}_2\text{Ti}_2\text{SiO}_7 \cdot 2\text{H}_2\text{O}$ ):** Sodium form of crystalline silicotitanate (Na-CST) was synthesized according to reported procedure<sup>1</sup>. The gel was prepared by mixing of titanium isopropoxide [ $\text{Ti}(\text{OC}_3\text{H}_7)_4$ ] and tetraethyl orthosilicate [TEOS,  $\text{Si}(\text{OC}_2\text{H}_5)_4$ ] in a plastic beaker. Titanium isopropoxide (4.56 g) was mixed with 3.33 g of TEOS, where the mole ratio of Ti:Si was 1:1. A NaOH solution (6.32 M, 26 mL) was added into above mixture with vigorous stirring, upon which a white powder precipitated. The heterogeneous mixture was stirred for 4 h at room temperature and then transferred into a 65 mL Teflon-lined autoclave using 15 mL of distilled-deionized water. The autoclave was then placed in a preheated oven at 170 °C for 10 days. The solid was collected by centrifugation, washed with copious amounts of water, and dried at 100 °C for 1 h.

**Preparation of H-CST (Hydrogen crystalline silicotitanate,  $\text{H}_2\text{Ti}_2\text{SiO}_7 \cdot x\text{H}_2\text{O}$ ):** The sodium form of crystalline silicotitanate (Na-CST,  $\text{Na}_2\text{Ti}_2\text{SiO}_7 \cdot 2\text{H}_2\text{O}$ ) was converted to hydrogen crystalline silicotitanate (H-CST,  $\text{H}_2\text{Ti}_2\text{SiO}_7 \cdot x\text{H}_2\text{O}$ ) according to the reported procedure<sup>2</sup>. First, 500 mg of Na-CST powder was dispersed in 70 ml of 0.1 M aqueous HCl solution. The solution was stirred (300 rpm) at RT for 1 h. The solid was collected by centrifugation, washed with copious amounts of water, and dried at 100 °C for 1 h.

**Synthesis of KMS-1 [ $\text{K}_{2x}\text{Mn}_x\text{Sn}_{3-x}\text{S}_6$  ( $x = 0.5-0.95$ ):** KMS-1 was synthesized according to reported procedure<sup>3</sup>. Briefly, a mixture was prepared by the mixing elemental Sn (7.14 g), Mn (1.656 g), S (5.78 g),  $\text{K}_2\text{CO}_3$  (4.16 g), and water (40 ml). The mixture was stirred for 1 h at room temperature and transferred in a Teflon-lined stainless-steel autoclave. The autoclave was placed in a preheated oven at 200 °C for 96 h under a static condition. After hydrothermal reaction, the autoclave was cooled to room temperature naturally. The brown polycrystalline product was collected by centrifugation and wash with water and dried at 60 °C for 6 h.

**Preparation of K-depleted phlogopite mica by topotactic leaching:** K-depleted phlogopite mica was prepared according to reported procedure<sup>4</sup>. Briefly, large crystals of phlogopite mica [ $\text{KMg}_3\text{Si}_3\text{AlO}_{10}(\text{OH})_2$ , ideal formula] were obtained from near Perth, Ontario, Canada. The crystals were broken into small pieces by hand followed by grinding them using a grinder in the presence of water. After grinding, the mica slurry was dispersed in water and allowed to sediment for 1 h. The supernatant solution containing the mica particles with the sizes smaller than 1  $\mu\text{m}$  was collected for the  $\text{Sr}^{2+}$  removal study. The phlogopite mica (size <1  $\mu\text{m}$ , 3 g) was stirred with 100 ml of a solution containing 1.0 M sodium chloride (NaCl), 0.2 N sodium tetraphenylborate (NaTPB), 0.01 M disodium ethylenediaminetetracetic acid (EDTA) for 24 h at 70 °C. This procedure was repeated four times to ensure the maximum removal of  $\text{K}^+$  from the galleries of the mica. The sample was collected by centrifugation and washed repeatedly

with a solution containing 40% 0.5 M NaCl and 60% acetone (v/v) to remove the precipitated potassium tetrphenylborate (KTBP) from the sample. After initial washings, the K-depleted phlogopite  $[\text{Na}_{0.90}\text{K}_{0.1}\text{Mg}_{1.75}\text{Si}_{2.75}\text{AlO}_{8.25}(\text{OH})_2]$  was washed with a copious amount of deionized water to remove all entrained NaCl and subsequently allowed them to dry in the air.

**Synthesis of ETS-10:** A silicon source solution was first prepared by dissolving  $\text{Na}_2\text{SiO}_3$  (18.4 g, 17-19%  $\text{Na}_2\text{O}$ , and 35-38%,  $\text{SiO}_2$ , Kanto) in water (60 g). Into this, a NaOH solution (2.4 g of NaOH and 20 g of water) was added with vigorous stirring, and the mixture was stirred for 2 h. For the preparation of titanium source solution, titanium isopropoxide (5.7 g),  $\text{H}_2\text{SO}_4$  (4.5 g), and water (35 g) were mixed together and heated at 100 °C for 90 min, 10 ml water was finally added into the mixture and allowed to cool at room temperature. The titanium source solution was added drop wise into the silicon source solution, and the mixture was stirred for 1 h. A dilute KF solution (1.2 g of KF and 15 g of water) was added into the above mixture. The mixture was aged for 16 h at room temperature and transferred into a Teflon-lined autoclave, and heated at 200 °C for 22 h under a static condition. After cooling the autoclave to room temperature, the crystals were collected by centrifugation, and washed with copious amounts of water. The scanning electron microscopy (SEM) shows the typical crystal size 200-300 nm, and those crystals could also be used as the seed for AM-6 and SGU-29 synthesis.

**Synthesis of AM-6:** *Preparation of silicon source solution:* A NaOH solution (3 g NaOH and 20 g of water) was added into the sodium silicate solution composed of 12.2 g of  $\text{Na}_2\text{SiO}_3$  (17-19%  $\text{Na}_2\text{O}$ , and 35-38%,  $\text{SiO}_2$ , Kanto) and 40 g of water. A dilute KCl solution (3 g of KCl and 10 g of water) was added into the above mixture and the mixture was vigorously stirred. *Preparation of vanadium source solution:*  $\text{H}_2\text{SO}_4$  (4.9 g) was added into a 100-mL round bottom flask containing water (10 g). Subsequently,  $\text{V}_2\text{O}_5$  (1.7 g) and EtOH (4 g) were sequentially added into the flask. The mixture was refluxed for 40 min, during the heterogeneous mixture was turned into blue solution. The blue vanadium source solution was added into the silicon source solution in a dropwise manner. The mixture was aged for 15 h at room temperature; seed ETS-10 (50 mg) was added into the gel and the gel was transferred into a 50 ml Teflon-lined autoclave, and placed in a preheated oven at 230 °C for 48 h, under a static condition. The precipitated pale yellow crystals were collected, washed, dried at 100 °C for 1 h.

**Synthesis of SGU-29:** SGU-29 was synthesized according to reported procedure<sup>5</sup>. The silicon source solution was prepared by mixing of sodium silicate (40 g, 10.6 %  $\text{Na}_2\text{O}$ , and ~26.5%  $\text{SiO}_2$ , Sigma-Aldrich), 1.3 g of NaOH, 12 g of KCl, 18.5 g of NaCl, and 60 g of water. The mixture was vigorously stirred (800 rpm) at room temperature for 3 h. The copper source solution was prepared by dissolving  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  (9 g) in 30 g of water containing 1.2 g of  $\text{H}_2\text{SO}_4$ . The copper source solution was then added

into the sodium silicate solution in a dropwise manner. The mixture was aged for 15 h at room temperature, and the pH was adjusted to 10.66 (if required) by adding diluted H<sub>2</sub>SO<sub>4</sub> in water. The seed ETS-10 (100 mg) was added into the gel and the gel was transferred into the 50 ml Teflon-lined autoclaves, and placed in a preheated oven at 215 °C for 24 h under a static condition. The precipitated light purple crystals were collected by centrifugation at 8000 rpm, and washed with copious amounts of water. The sample was dried at 100 °C for 2 h.

**Synthesis of ETS-4:** The gel consisting of Na<sub>2</sub>SiO<sub>3</sub>, Ti[OCH(CH<sub>3</sub>)<sub>2</sub>]<sub>4</sub>, H<sub>2</sub>SO<sub>4</sub>, NaOH, KF, and H<sub>2</sub>O were prepared, where the mole ratio of the synthesis gel in terms of SiO<sub>2</sub>:TiO<sub>2</sub>:H<sub>2</sub>SO<sub>4</sub>:Na<sub>2</sub>O:KF:H<sub>2</sub>O were 5.3:1:2.11:7.26:0.95:303, respectively. A silicon source solution was first prepared by dissolving 18.3 g of Na<sub>2</sub>SiO<sub>3</sub> solution (17-19% Na<sub>2</sub>O, and 35-38%, SiO<sub>2</sub>, Kanto) in 58 g of water. Into this, a NaOH solution (4 g of NaOH and 20 g of water) was added with vigorous stirring, and the mixture was stirred for 2 h. For the preparation of titanium source solution, titanium isopropoxide (6 g), H<sub>2</sub>SO<sub>4</sub> (4.5 g), and water (20 g) were mixed together and heated at 100 °C for 90 min, 10 ml H<sub>2</sub>O was finally added into the mixture and allowed to cool at room temperature. The titanium source solution was added dropwise into the silicon source solution, and the mixture was stirred for 1 h. A dilute KF solution (1.2 g of KF and 15 g of water) was added into the above mixture. The mixture was aged for 16 h at room temperature and transferred into a Teflon-lined autoclave, and heated at 200 °C for 22 h under a static condition. After cooling the autoclave to room temperature, the crystals were collected by centrifugation, and washed with copious amounts of water and dried at 80 °C for 2 h.

**Synthesis of AM-1:** AM-1 was synthesized according to reported procedure<sup>6</sup>. Briefly, a NaOH solution (2.85 g NaOH and 13 g of water) was added into 20.1 g of sodium silicate solution (27% SiO<sub>2</sub>, 8% Na<sub>2</sub>O, Merck) and the mixture was stirred for 10 min. Subsequently, 16.5 g of TiCl<sub>3</sub> solution (20% in 3% HCl, Alfa Aesar) was added in a dropwise manner. The mixture was vigorously stirred for 5 h at room temperature, transferred into a Teflon-lined autoclave, and heated at 230 °C for 96 h under a static condition. After cooling the autoclave to room temperature, the crystals were collected by centrifugation, and washed with copious amounts of water and dried at 80 °C for 2 h. In order to obtain 0.5~1 μm size crystal; 100 mg of these AM-1 crystals was grounded using mortar and pestle, and added as seeds to the same starting gel and submitted to a new hydrothermal reaction at 230 °C for 24 h.

**Synthesis of AM-4:** AM-4 was synthesized according to reported procedure<sup>7</sup>. A NaOH solution (14.76 g NaOH and 39 g of water) was added into 27.1 g of sodium silicate solution (27% SiO<sub>2</sub>, 8% Na<sub>2</sub>O, Merck) and the mixture was stirred for 30 min. Subsequently, 27.1 g of TiCl<sub>3</sub> solution (15% m/m solution of TiCl<sub>3</sub> in 10% m/m HCl, Merck) was added in a dropwise manner. The mixture was stirred for 4 h at room

temperature and transferred into a Teflon-lined autoclave. The hydrothermal reaction was performed at 230 °C for 96 h under a static condition. The reaction mixture was cooled to room temperature, and the product was collected by centrifugation, and washed with copious amounts of water and dried at 80 °C for 2 h. In order to obtain 1~2  $\mu\text{m}$  size crystal; 100 mg of these AM-4 crystals was grounded using mortar and pestle, and added as seeds to the same starting gel and submitted to a new hydrothermal reaction at 230 °C for 24 h.

**Preparation of NaX:** Purchased zeolite X (Lot no 943196110142 from UOP) was ion exchanged with  $\text{Na}^+$  by placing 1 g of zeolite X powder in a glass vial containing 90 mL of 1 M NaCl solution and subsequently stirred at 70 °C for 120 min. This procedure was repeated twice. After ion-exchange the sample was washed with copious amount of water until the chloride test with  $\text{AgNO}_3$  solution was negative.

**Preparation of Ca(81)-SGU-7:** Na-SGU-7 was ion exchanged with  $\text{Ca}^{2+}$  by placing 1 g of SGU-7 powder in a glass vial containing 90 mL of 0.50 M aqueous  $\text{CaCl}_2$  solution and subsequently stirred at 70 °C for 180 min. This procedure was repeated three times to achieve maximum degree of ion-exchange.

**Preparation of Sr-SGU-7:** Na-SGU-7 was ion exchanged with  $\text{Sr}^{2+}$  by placing 1 g of SGU-7 powder in a glass vial containing 90 mL of 0.25 M  $\text{SrCl}_2$  solution and subsequently stirred at 70 °C for 180 min. This procedure was repeated three times. After ion-exchange the sample was washed with copious amount of water until the chloride test with  $\text{AgNO}_3$  solution was negative.

**Methods:  $\text{Sr}^{2+}$  Capture.** Capture of  $\text{Sr}^{2+}$  from  $\text{Sr}^{2+}$ -containing solutions by  $\text{Sr}^{2+}$ -removers was conducted as follows. Into a 50 mL glass container 20 mL of a  $\text{Sr}^{2+}$ -containing solution and 20 mg of a sorbent were added. The mixture was initially sonicated for 5 min followed by stirring for 12 or 24 h at 25 °C or at the desired temperature by using a temperature-controlled water bath. In most experiments, the volume to mass ratio ( $V/m$ ) was 1 L/g or 1 L of solution per gram of  $\text{Sr}^{2+}$ -remover. Only the liquid part of the heterogeneous solution was isolated by centrifugation. The concentrations of  $\text{Sr}^{2+}$  in the supernatant solutions were determined by inductively coupled plasma–mass spectroscopy (ICP-MS). The amount of radioactive  $^{226}\text{Ra}$  in a solution was estimated by measuring the alpha radiation emitted from the daughter nuclide  $^{222}\text{Rn}$ . The concentration of radioactive  $^{137}\text{Cs}$  was estimated using a gamma counter. Each data point represents the average value of three measured values.

**Capture of radioactive  $^{90}\text{Sr}^{2+}$ :** Radioactive waste solutions with the initial activities of 28 and 32 Bq/mL (5.4 and 6.2 ppt of  $^{90}\text{Sr}$ , respectively) were obtained from Eckert & Ziegler (Georgia, USA). Required amounts of  $\text{NaNO}_3$  were added to the solutions to obtain  $^{90}\text{Sr}^{2+}$ -containing 2 M and 5 M  $\text{Na}^+$  solutions.

Capture of trace amounts of radioactive  $^{90}\text{Sr}^{2+}$  was conducted by adding 20 mg of a  $\text{Sr}^{2+}$ -remover into 20 ml of  $^{90}\text{Sr}^{2+}$ -containing 2 M or 5 M  $\text{Na}^+$  solution. The mixture was sonicated and magnetically stirred for 24 h at 25 °C. The solution was centrifuged at 8,000 rpm and filtered. The amount of  $^{90}\text{Sr}^{2+}$  in the supernatant solution was estimated using the Cerenkov radiation technique.

*Capture of radioactive  $^{226}\text{Ra}^{2+}$ :* A radioactive  $^{226}\text{Ra}^{2+}$  solution with the initial activity of 43 Bq/mL (1.16 ppb) was prepared by dissolving  $^{226}\text{RaCl}_2$  in water at the concentration of about  $5 \times 10^{-9}$  M. Into this solution a required amount of NaCl was added to prepare 2 M  $\text{Na}^+$  solution. The concentration of radioactive  $^{226}\text{Ra}^{2+}$  in the supernatant solution after  $^{226}\text{Ra}^{2+}$  capture was determined by the  $^{222}\text{Rn}$  emanation method using a liquid scintillation counter.

*Preparation of pH-dependent  $\text{Sr}^{2+}$ -containing solutions:*  $\text{Sr}^{2+}$ -containing solutions with different pH were prepared by adding required amounts of HCl or NaOH into the  $\text{Sr}^{2+}$  solution whose initial  $\text{Sr}^{2+}$  concentration was adjusted to 1.02 ppm.

*Preparation of the reaction mixture for 1-equivalent condition.* In a typical experiment, 5 mg of SGU-7 powder and 1.027 L of 1.03 ppm of  $\text{Sr}^{2+}$  containing solution were introduced into a 2 L glass flask. The mixture was sonicated for 5 min and kept at room temperature for 12 h while stirring the solution. The volume to mass ratio of the solution was 205 L/g.

**Instrumentation.** A computer-controlled Agilent (7700x Series) inductively coupled plasma mass spectrometer (ICP-MS) with a octopole reaction system (ORS) with improved performance of helium (He) collision mode (detection limit 0.2 ppt for Sr) was used to determine the concentrations of  $\text{Sr}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$  and  $\text{Na}^+$  ions in solution. Eight standard solutions in the range of 0-1000 ppb were prepared by diluting standard solutions provided from Agilent. Standard samples were prepared in a 5% nitric acid solution with 1 ppb  $^{153}\text{Eu}$  internal standard in order to correct for instrumental drift and matrix effects during analysis. The highly  $\text{Na}^+$ -rich solutions (2-5 M  $\text{Na}^+$  ions) were diluted by a factor of 10 or 20 to lower the  $\text{Na}^+$  concentrations.

**Characterizations:** Scanning electron microscopy (SEM) images were obtained using a field-emission scanning electron microscope JEOL JSM-7600 and Hitachi S-4300 operating at an acceleration voltage of 15 kV and 20 kV, respectively. Powder X-ray diffraction (XRD) patterns were obtained using a Rigaku D/MAX-2500/pc diffractometer. HRTEM images and selected area electron diffraction (SAED) patterns were collected on the JEM-2100F as well as on a JEOL JEM-4010 microscope operated at 400 kV ( $C_s = 0.8$  mm, point resolution = 1.5 Å) with LaB6 filament. Energy dispersive X-ray spectroscopy (EDX) analysis was done on a JEOL JEM-2100F. 3D EDT datasets were collected using a high-tilt specimen

holder on a JEOL JEM 2100F microscope operated at 200 kV ( $C_s = 0.8$  mm, point resolution = 1.9 Å) with Schottky-type field emission gun, equipped with Gatan US1000 CCD camera (2K × 2K). HRTEM images were simulated using a software *Emap*<sup>8</sup>. The structures were solved by direct methods using Sir 2014<sup>9</sup>. High-resolution spherical aberration corrected ( $C_s$ -corrected) scanning transmission electron microscopy (STEM) was performed using a JEOL Grand ARM operated at 300 kV. This microscope is equipped with a cold field emission gun and a double corrector for the STEM and TEM modes assuring a point resolution in either case of 0.7 Å. Prior to observation the samples were deeply crushed using mortar and pestle and dispersed in absolute ethanol. Few drops of the suspension were placed onto a holey carbon copper microgrid.

**Multi-slice STEM simulations:** To simulate the STEM-ADF data of the SGU-7 along the [010] orientation we used the QSTEM program<sup>10</sup>. The crystal supercell dimensions used were  $48.28 \times 103.22 \times 23.8$  Å<sup>3</sup>, where 20 Å was used as ideal thickness because of the good match between the experimental and simulated data and because of the information that was obtained from the SEM observations. The parameters used were:  $C_s = 0$  mm,  $U_{acc} = 300$  kV, an inner collection angle of 30 mrad and half-convergence angle of 15 mrad.

**Zero-field cooled (ZFC) susceptibility** measurements of SGU-7 sample was conducted from 4 to 300 K in a magnetic field of 1000 G on a SQUID magnetometer (MPMS5) at the Korea Basic Science Institute located at Korea University. **Electron spin resonance (ESR)** spectra were measured at room temperature on a Bruker A200 electron spin resonance spectrometer. **Raman spectra** of the samples were recorded on a homemade setup equipped with an Ar<sup>+</sup> ion laser (Spectra-Physics Stabilite 2017) as an excitation beam source, a spectrometer (Horiba Jobin Yvon TRIAX 550), and a CCD detector (Horiba Jobin Yvon Symphony) cooled at -196 °C. The wavelength of the excitation beam was 514.5 nm. **XPS** data were measured using ESCALAB250 XPS system, from Korea Basic Science Institute located at Busan. Sr<sup>2+</sup> uptake and chemical compositions were determined using ICP-MS (detection limit 1 ppb), 7700 series, Agilent Technologies.

**High-resolution powder diffraction** data have been recorded at the ID22 beamline at the European Synchrotron Radiation Facility (ESRF, France). Samples has been packed in 0.7- and 0.5-diameter borosilicate capillaries. A first series of diffraction patterns has been collected at an energy of 31 keV (0.3999 Å) and used for Pawley fits. In an attempt to remove part of the water molecules without inducing critical changes, a second series of diffraction patterns has been collected after *in situ* pumping and moderate heating (50 °C - 100 °C) at an energy of 35 keV (0.3543 Å). These patterns have been used for

the Rietveld refinement of Na-SGU7 and Sr-SGU7. Pawley and Rietveld refinements have been carried out using the TOPAS software package<sup>11</sup>.

**Calculation Details.** Molecular dynamics (MD) simulations were performed at 298 K. All calculations were done by the BIOVIA Materials Studio 2016 package. We used canonic (NVT) ensemble and Nose thermostat with Q ratio of 0.01. A very short time step 1 fs was used in order to effectively integrate the equation of motion. After the initial 300 ps ( $3 \times 10^5$  steps) equilibration, 10 ns ( $1 \times 10^7$  steps) run was proceeded with the frame output every 2,500 steps. Thus, total of 4,000 configurations are used to get the averaged data. And also, we performed at least three simulations at each model and got the averaged values. Na-SGU-7 ( $\text{Na}_{38}\text{H}_2\text{Si}_{80}\text{V}_{20}\text{O}_{220} \cdot 55\text{H}_2\text{O}$ ,  $P12_1/a1$ ,  $a = 23.58 \text{ \AA}$ ,  $b = 30.04 \text{ \AA}$ ,  $c = 12.31 \text{ \AA}$ ,  $\beta = 100.2^\circ$ ) and Na-CST ( $\text{Na}_8\text{Ti}_8\text{Si}_4\text{O}_{28} \cdot 8\text{H}_2\text{O}$ ,  $P4_2/mcm$ ,  $a = b = 7.8082 \text{ \AA}$ ,  $c = 11.9735 \text{ \AA}$ ,  $\alpha = \beta = \gamma = 90^\circ$ )<sup>12</sup>, were used as initial structure models. To get  $\text{Sr}^{2+}$  ion-exchange models, we built only one  $\text{Sr}^{2+}$  ion exchange models to mimic the experimental conditions of low  $\text{Sr}^{2+}$  concentration (ppt) compared with the extremely high  $\text{Na}^+$  ion concentration (ppm). Universal force field (UFF) was used. The framework charges were assigned using the charge equilibration method. Charges of Na, Sr, and O and H of  $\text{H}_2\text{O}$  were assigned as +1, +2, -0.672 and +0.336, respectively. The electrostatic summation was carried out by the Ewald method with the accuracy of  $1.0 \text{ e}^{-5}$  kcal/mol and the van der Waals interaction was truncated using the cubic spline method with the cutoff distance of  $18.5 \text{ \AA}$  and applied the long-range correction.

### References for Experimental Section

1. D. M. Poojary, R. A. Cahill and A. Clearfield, *Chem. Mater.*, 1994, **6**, 2364-2368.
2. A. J. Celestian, J. D. Kubicki, J. Hanson, A. Clearfield and J. B. Parise, *J. Am. Chem. Soc.*, 2008, **130**, 11689-11694.
3. M. J. Manos, N. Ding and M. G. Kanatzidis, *Proc. Natl. Acad. Sci. U.S.A.*, 2008, **105**, 3696-3699.
4. S. Komarneni and R. Roy, *Science*, 1988, **239**, 1286-1288.
5. S. J. Datta, C. Khumnoon, Z. H. Lee, W. K. Moon, S. Docao, T. H. Nguyen, I. C. Hwang, D. Moon, P. Oleynikov, O. Terasaki and K. B. Yoon, *Science*, 2015, **350**, 302-306.
6. C. Rubio, C. Casado, S. Uriel, C. T  lez and J. Coronas, *Materials Letters*, 2009, **63**, 113-115.
7. Z. Lin, J. Rocha, P. Brandao, A. Ferreira, A. P. Esculcas, J. D. P. de Jesus, A. Philippou and M. W. Anderson, *J. Phys. Chem. B*, 1997, **101**, 7114-7120.
8. P. Oleynikov, *Cryst. Res. Technol.*, 2011, **46**, 569-579.
9. M. C. Burla, R. Caliendo, B. Carrozzini, G. L. Cascarano, C. Cuocci, C. Giacovazzo, M. Mallamo, A. Mazzone and G. Polidori, *J. Appl. Cryst.*, 2015, **48**, 306-309.
10. C. T. Koch, *Determination of core structure periodicity and point defect density along dislocations*, Arizona State University, 2002.
11. A. A. Coelho, J. Evans, I. Evans, A. Kerna and S. Parsons, *Powder Diffraction*, 2011, **26**, S22-S25.

## Tables S1-S6

**Table S1.** Comparison of structural parameters of Na(88)-SGU-7 and Sr(95)-SGU-7 obtained from Synchrotron powder XRD.

Name	Na(95)-SGU-7	Sr(95)-SGU-7
Analysis technique	Synchrotron powder XRD	Synchrotron powder XRD
Wavelength	0.3543 Å	0.3543 Å
Source	ID22 beamline at the European Synchrotron Radiation Facility (ESRF, France)	ID22 beamline at the European Synchrotron Radiation Facility (ESRF, France)
Chemical composition in unit cell	Na <sub>59</sub> Si <sub>96</sub> V <sub>20</sub> O <sub>332</sub>	Sr <sub>25</sub> Na <sub>8</sub> Si <sub>96</sub> V <sub>20</sub> O <sub>337</sub>
Crystal system	Monoclinic	Monoclinic
Space group	<i>P</i> 1 2 <sub>1</sub> /a 1 (no. 14)	<i>C</i> 1 2/m 1 (no. 12)
Unit cell	a = 23.5144(2) Å	a = 23.4286(7) Å
	b = 29.924(-8) Å	b = 29.971(2) Å
	c = 12.2871(1) Å	c = 12.3063(8) Å
	β = 100.772(9)°	β = 101.441(5)°
Volume[Å <sup>3</sup> ]	8493.623	8469.647
R <sub>p</sub>	0.0601	0.0567
R <sub>wp</sub>	0.0854	0.0768

**Table S2.** Comparison of structural parameters of Na(88)-SGU-7 and Sr(95)-SGU-7 obtained from 3D EDT.

Name	Na(95)-SGU-7	Sr(95)-SGU-7
Analysis technique	3D EDT	3D EDT
Wavelength		
Source		
Chemical composition in unit cell	Na <sub>56</sub> O <sub>260</sub> Si <sub>96</sub> V <sub>20</sub>	Sr <sub>14</sub> V <sub>20</sub> Si <sub>96</sub> O <sub>536</sub>
Crystal system	Monoclinic	Monoclinic
Space group	<i>P</i> 1 2 <sub>1</sub> /a 1 (no. 14)	<i>C</i> 1 2/ <i>m</i> 1 (no. 12)
Unit cell	a = 23.578 Å	a = 23.700 Å
	b = 30.039 Å	b = 29.600 Å
	c = 12.311 Å	c = 12.000 Å
	β = 100.15°	β = 101.00°
Volume[Å <sup>3</sup> ]	8582.9	8263.6
R <sub>p</sub>	--	--
R <sub>wp</sub>	--	--

**Table S3.** Sr<sup>2+</sup> capture from simulated waste solution using indicated adsorbents.

Sample	Initial concentration (ppb)	Final concentration (ppb)	Sr <sup>2+</sup> removal (%)	$K_d$ (mL/g)	Log $K_d$
<b>Na-SGU-7</b>	35.0	0.31	99.11	110982	5.05
<b>Na-CST</b>	35.0	0.69	98.03	49798	4.70
<b>KMS-1</b>	35.0	1.92	94.53	17267	4.24
<b>ETS-10</b>	35.0	3.69	89.46	8488	3.93
<b>AM-6</b>	35.0	5.18	85.19	5751	3.76
<b>ETS-4</b>	35.0	10.96	68.68	2193	3.34
<b>AM-1</b>	35.0	11.73	66.49	1984	3.30
<b>AM-4</b>	35.0	12.24	65.03	1860	3.27
<b>NaMica</b>	35.0	13.31	61.97	1630	3.21
<b>LTA</b>	35.0	13.72	60.81	1552	3.19
<b>FAU(NaX)</b>	35.0	14.51	58.53	1411	3.15
<b>SGU-29</b>	35.0	18.80	46.29	862	2.94
<b>H-CST</b>	35.0	23.56	32.69	486	2.69

Simulated waste solution composition: Sr<sup>2+</sup> (35 ppb), Na<sup>+</sup> (115,000 ppm, or 5 M), pH = 14. Batch condition 1 L/g, equilibrium time 12 h, at 25 °C.

**Table S4.** Selective Sr<sup>2+</sup> capture from groundwater using various sorbents.

Sample	Chemical Composition	Initial concentration (ppb)	Final concentration (ppb)	Sr <sup>2+</sup> removal (%)	$K_d$ (mL/g)	Log $K_d$
<b>Na-SGU-7</b>	Na <sub>1.9</sub> VSi <sub>4</sub> O <sub>10.95</sub> ·2H <sub>2</sub> O	42.2	0.29	99.31	144517	5.16
<b>Na-CST</b>	Na <sub>2</sub> Ti <sub>2</sub> SiO <sub>7</sub> ·2H <sub>2</sub> O	42.2	0.30	99.29	140611	5.15
<b>KMS-1</b>	K <sub>2x</sub> Mn <sub>x</sub> Sn <sub>3-x</sub> S <sub>6</sub> (x = 0.5-0.95)	42.2	2.23	94.72	17924	4.25
<b>FAU</b>	Na <sub>88</sub> (Al <sub>88</sub> Si <sub>104</sub> O <sub>384</sub> )·xH <sub>2</sub> O	42.2	4.29	89.84	8846	3.95
<b>AM-1</b>	Na <sub>2</sub> TiSi <sub>4</sub> O <sub>11</sub> ·xH <sub>2</sub> O	42.2	5.69	86.52	6418	3.81
<b>LTA (NaA)</b>	NaAlSiO <sub>4</sub> ·xH <sub>2</sub> O	42.2	14.97	64.53	1819	3.26
<b>Na-Pmica</b>	Na <sub>0.9</sub> K <sub>0.1</sub> Mg <sub>1.75</sub> Si <sub>2.75</sub> AlO <sub>8.25</sub> (OH) <sub>2</sub>	42.2	20.38	51.71	1071	3.03
<b>SGU-29</b>	Na <sub>1.15</sub> K <sub>0.84</sub> CuSi <sub>5</sub> O <sub>12</sub> ·xH <sub>2</sub> O	42.2	25.90	38.63	629	2.80
<b>AM-6</b>	Na <sub>1.12</sub> K <sub>0.85</sub> VSi <sub>5</sub> O <sub>13</sub> ·xH <sub>2</sub> O	42.2	26.29	37.70	605	2.78
<b>ETS-10</b>	Na <sub>1.11</sub> K <sub>0.83</sub> TiSi <sub>5</sub> O <sub>13</sub> ·xH <sub>2</sub> O	42.2	26.45	37.32	595	2.77
<b>ETS-4</b>	Na <sub>1.41</sub> K <sub>0.22</sub> TiSi <sub>2.7</sub> O <sub>8.2</sub> ·xH <sub>2</sub> O	42.2	28.32	32.89	490	2.69
<b>AM-4</b>	Na <sub>1.8</sub> TiSi <sub>1.8</sub> O <sub>6.5</sub> ·xH <sub>2</sub> O	42.2	35.78	15.21	179	2.25
<b>H-CST</b>	H <sub>2</sub> Ti <sub>2</sub> SiO <sub>7</sub> ·xH <sub>2</sub> O	42.2	40.64	3.70	38	1.58

Ground water composition: Sr<sup>2+</sup> (0.042 ppm), Na<sup>+</sup> (131 ppm), Ca<sup>2+</sup> (24 ppm), Mg<sup>2+</sup> (10 ppm), K<sup>+</sup> (7 ppm). Batch condition 1 L/g, equilibrium time 12 h, at 25 °C.

**Table S5.** Sr<sup>2+</sup> capture from groundwater at various [Sr<sup>2+</sup>]<sub>i</sub> using Na-SGU-7.

Initial concentration (ppm)	Final concentration (ppm)	Sr <sup>2+</sup> removal (%)	K <sub>d</sub> (mL/g)	Log K <sub>d</sub>
0.043	0.0003	99.29	140667	5.15
0.118	0.0006	99.50	199678	5.30
0.556	0.0019	99.66	290695	5.46
1.166	0.0038	99.67	302646	5.48
5.628	0.0297	99.47	188751	5.28
10.696	0.1070	99.00	98963	5.00
21.272	0.4580	97.85	45445	4.66
31.444	1.5480	95.08	19313	4.29

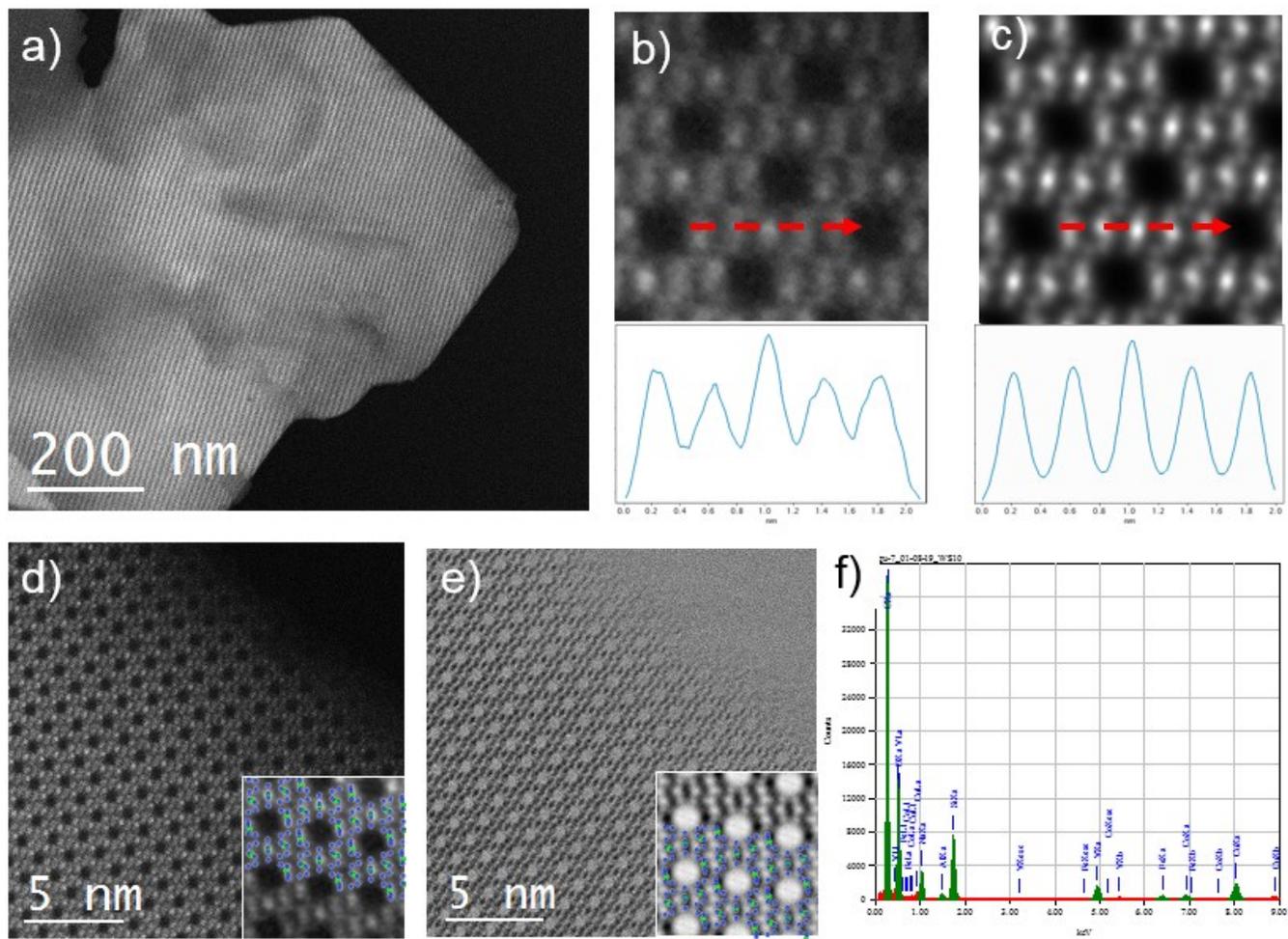
Ground water composition: Na<sup>+</sup>: 131, Ca<sup>2+</sup>: 24, Mg<sup>2+</sup>: 10, K<sup>+</sup>: 7 ppm. Batch condition 1 L/g, equilibrium time 12 h, at 25 °C.

**Table S6.** Comparison of Sr<sup>2+</sup> removal under acidic condition

Sample	pH	Initial concentration (ppm)	Final concentration (ppm)	Sr <sup>2+</sup> removal (%)	$K_d$ (mL/g)	Log $K_d$
Na-SGU-7 <sup>a</sup>	~3.2	4.10	0.003	99.93	1365667	6.13
KMS-1*	~3.2	4.09	0.153	96.26	24986	4.40

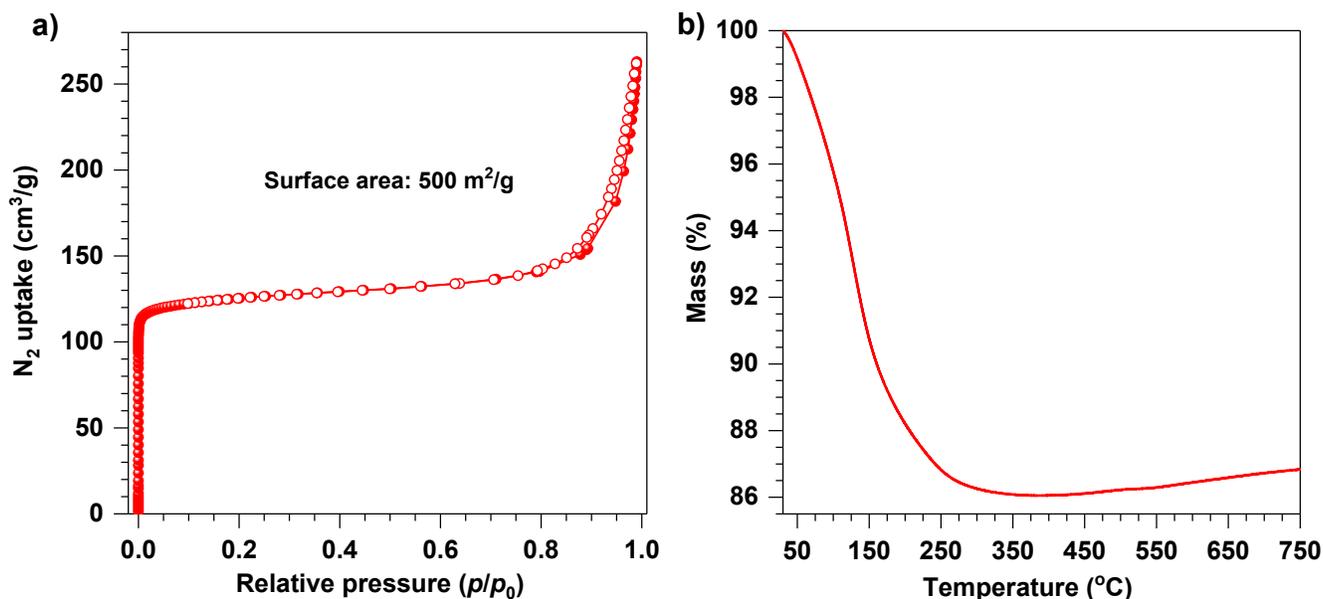
<sup>a</sup>Batch condition 1 L/g, equilibrium time 12 h, at 25 °C. \*data taken from [M. J. Monos, N. Ding and M. G. Kanatzidis, *Proc. Natl. Acad. Sci. U.S.A.*, 2008, **105**, 3696-3699.]

**SI 1. STEM images of Na-SGU-7 along [010] and EDS spectrum.**



**Fig. S1** STEM images of Na-SGU-7 and EDS analysis. (a) Low-mag ADF, (b) experimental high-resolution ADF image with intensity profiles along the red arrow, (c) simulated high-resolution ADF image with intensity profile, (d) experimental ADF images, (e) experimental ABF image, and (f) EDS spectrum. The insets in (d) and (e) are filtered images (overlaid with atomic structural model) after applying  $p2$  symmetry. The oxygen and cations in the structural model have been omitted for clear comparison. Cu, Fe and Co in EDS spectrum come from the equipment, grid and holder.

## SI 2. N<sub>2</sub> adsorption isotherms and thermogravimetric analysis (TGA).

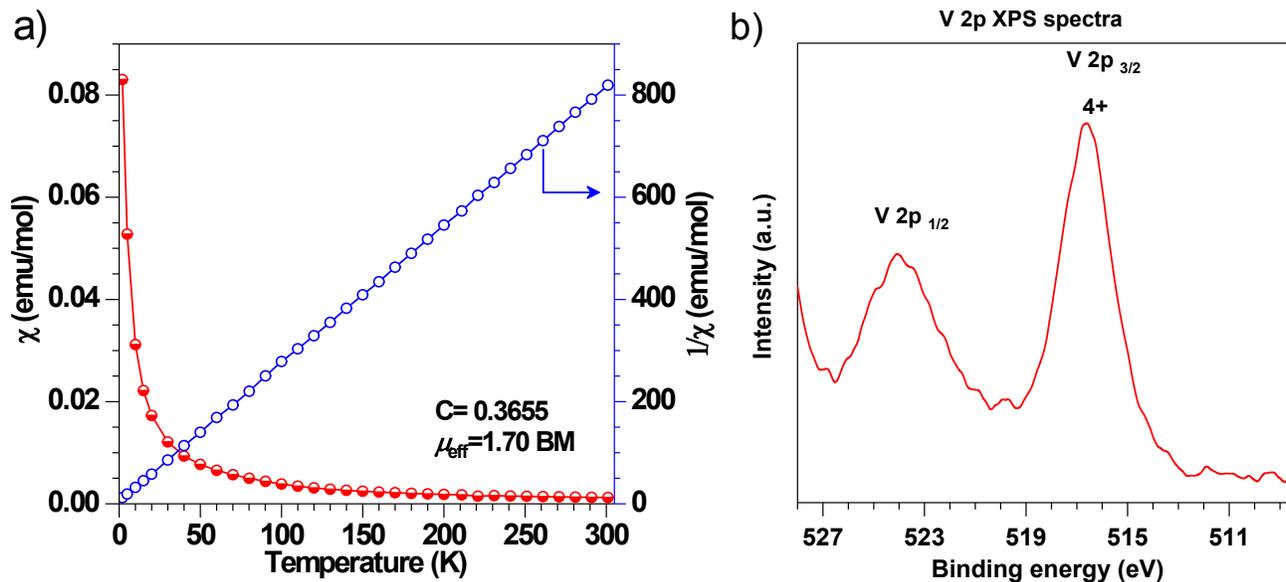


**Fig. S2** (a) N<sub>2</sub> adsorption and desorption isotherms of Na-SGU-7 measured at 77 K. (b) Thermogravimetric analysis (TGA) of Na-SGU-7 under air.

(a) Na-SGU-7 shows Type I isotherm according to the classification of Brunauer, Emmett, and Teller (BET), which is typical for microporous materials. The obtained total pore volume, BET surface area, and Langmuir surface areas are 0.406 cm<sup>3</sup> g<sup>-1</sup>, 500 m<sup>2</sup> g<sup>-1</sup>, and 520 m<sup>2</sup> g<sup>-1</sup>, respectively.

(b) Thermogravimetric analysis (TGA) of Na-SGU-7 was measured between room temperature and 750 °C with air as the eluting gas. Water loss starts from 25 °C and continued up to at 350 °C. The total weight loss was ca. 14%. Weight increase starts at 375 °C, owing to the oxidation of V<sup>4+</sup> to V<sup>5+</sup>.

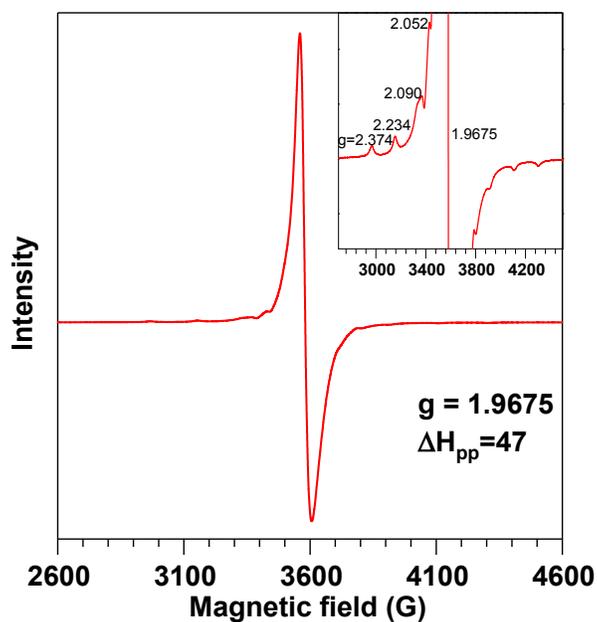
### SI 3. Magnetic susceptibility and X-Ray Photoelectron Spectroscopy (XPS) of Na-SGU-7



**Fig. S3** (a) Plots of magnetic susceptibility ( $\chi$ ) and inverse magnetic susceptibility ( $1/\chi$ ) of pristine SGU-7 with respect to temperature ( $T$ ) from 2 to 300 K. The calculated effective magnetic moment ( $\mu_{\text{eff}}$ ) is 1.70 BM. The linear relationship established between  $1/\chi$  and  $T$  demonstrates that the V ions in SGU-7 are Curie-like purely paramagnetic. (b) V 2p core-level XPS spectra of Na-SGU-7 recorded at room temperature.

Note that the magnetic susceptibility ( $\chi$ ) sharply increases at the low temperature region, indicating the ferromagnetic properties of Na-SGU-7. From the linear plot shown in the temperature region between 2 to 300 K,  $C = 0.3655 \text{ mol K/emu}$  was deduced from the Curie equation of  $\chi = C/T$ , where, and the effective magnetic moment can be determined from the equation  $\mu_{\text{eff}} = 2.828 \sqrt{C}$ . An effective magnetic moment of  $1.70 \mu_{\text{B}}$  was obtained from Na-SGU-7 sample. This indicates that V atoms in Na-SGU-7 exist in  $\text{V}^{4+}$  oxidation state.

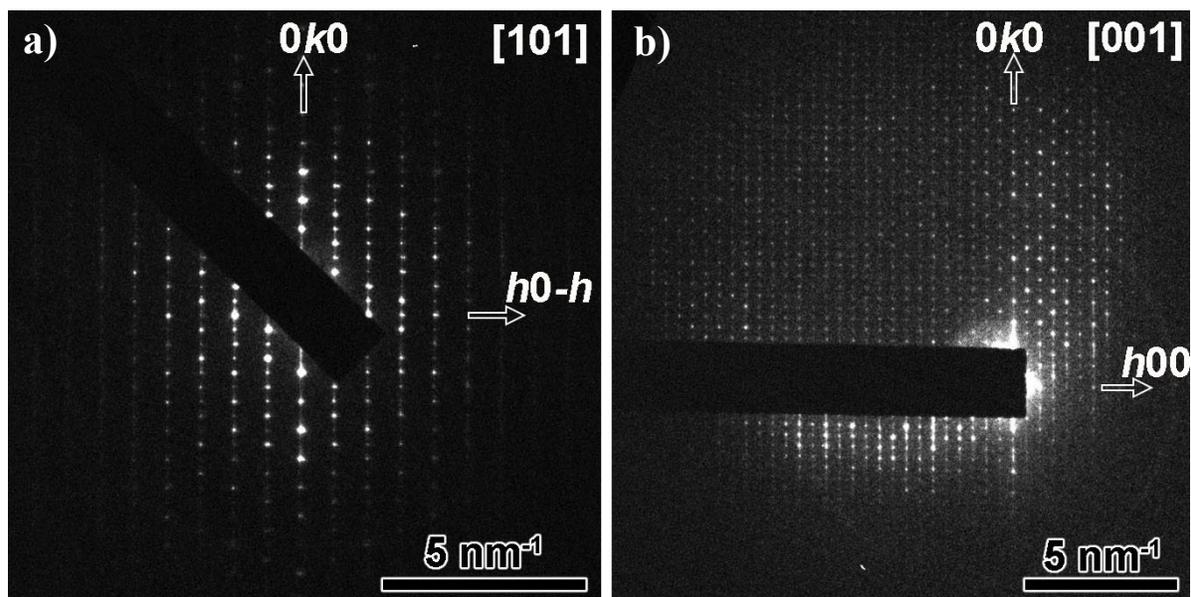
#### SI 4. ESR spectra of Na-SGU-7



**Fig. S4** X-band ESR spectrum of Na-SGU-7 measured at room temperature showing the hyperfine interaction.

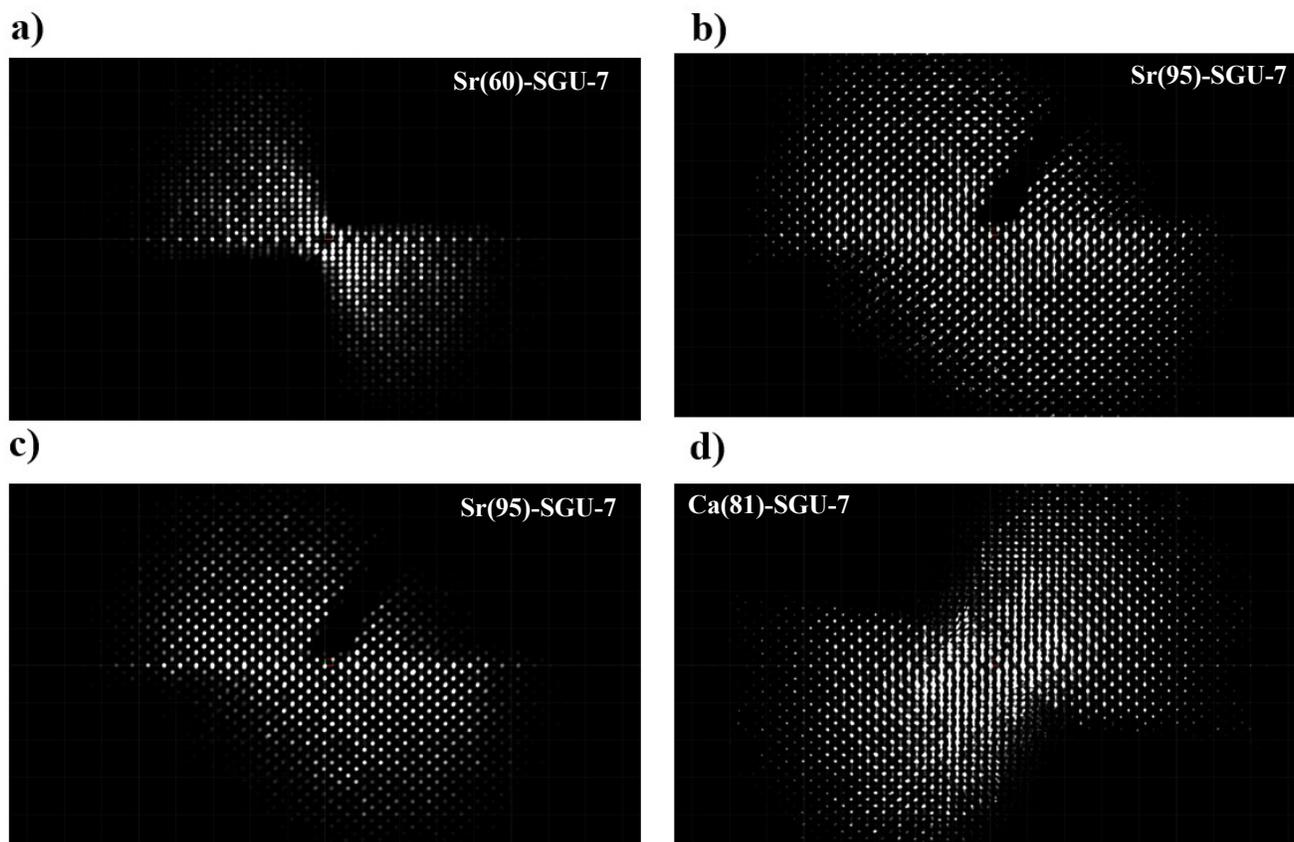
The spectrum showed a strong signal of  $V^{4+}$  with the  $g$  value of 1.9675 and  $\Delta H_{pp}$  of 47 G, consistent with the presence of an unpaired electron in each  $V^{4+}$  ion. The hyperfine structure is observed from Na-SGU-7 sample (inset).

SI 5. Selected area electron diffraction (SAED) patterns of Na-SGU-7 along [101] and [001] directions.



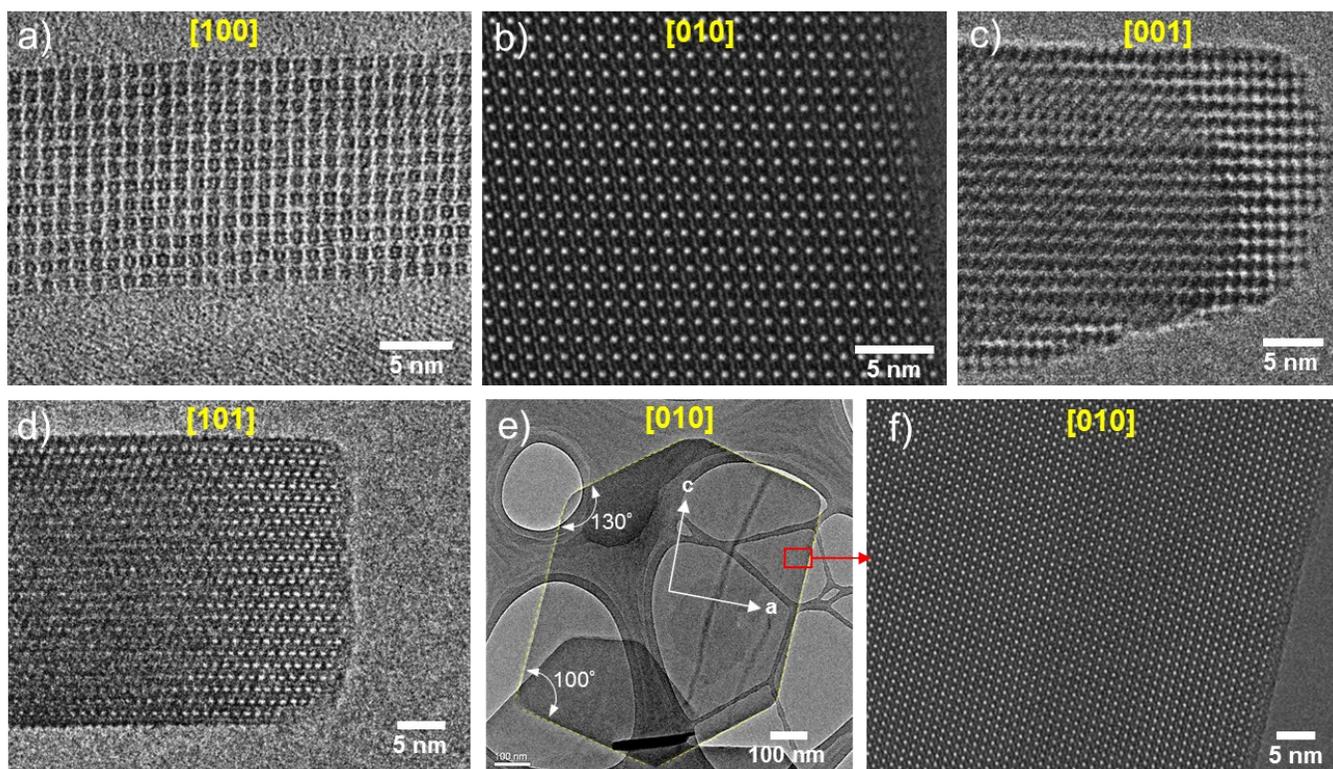
**Fig. S5** SAED patterns of Na-SGU-7 along (a) [101] and (b) [001] directions. Reflection conditions of  $0k0$ :  $k = 2n$  can be observed. Some residual intensities of  $0k0$  ( $k$  is odd) reflections in the right pattern might come from the dynamical scattering.

**SI 6. 3D EDT datasets of Sr- and Ca- ion exchange SGU-7 samples revealing the phase change from P21/a to C2/m, during monovalent Na-ions are replaced by divalent Sr- or Ca-ions.**



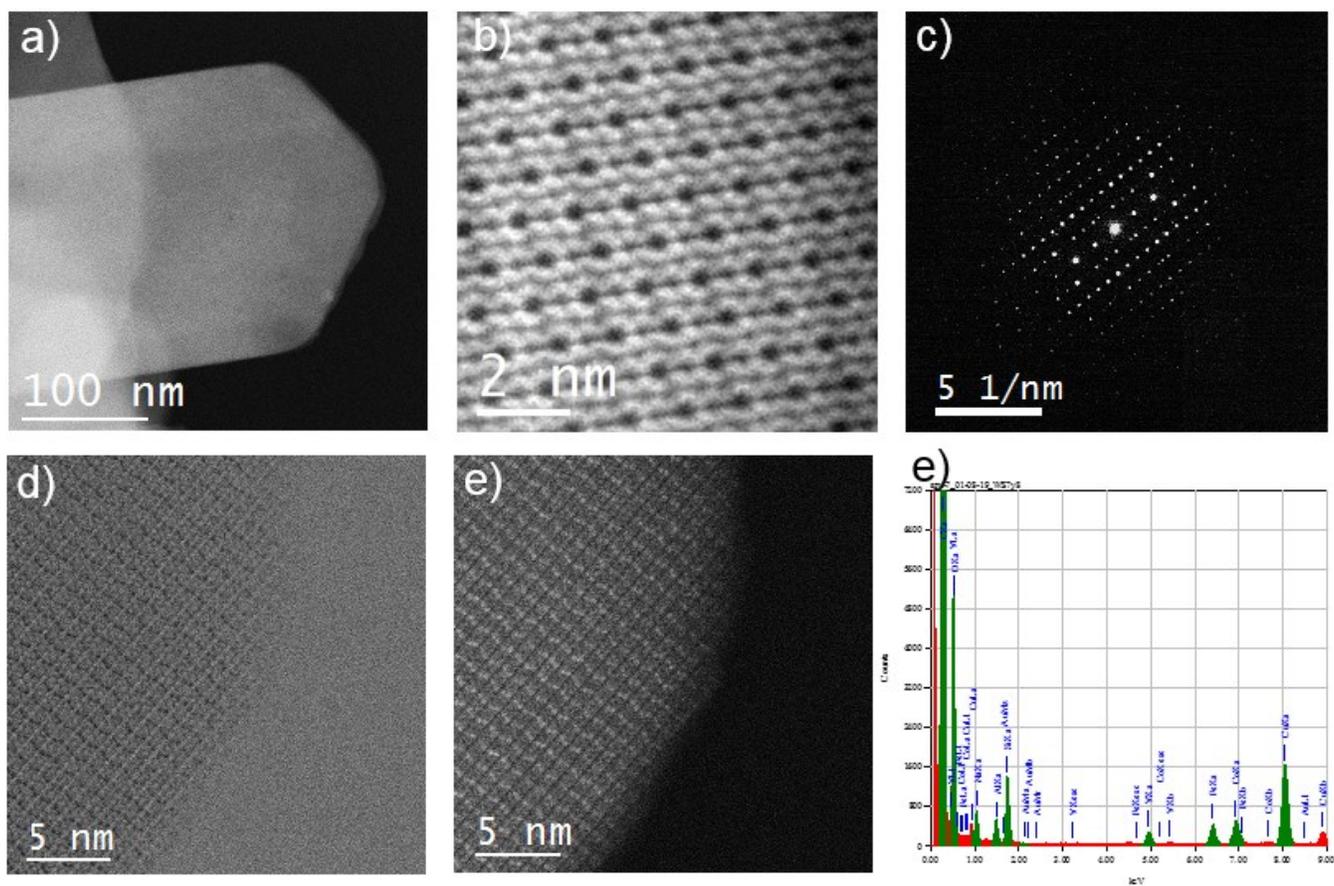
**Fig. S6** 3D EDT datasets collected from different samples: (a) Sr(60)-SGU-7, (b) and (c) Sr(95%)-SGU-7, and (d) Ca(81)-SGU-7, respectively.

SI 7. High resolution transmission electron microscopy (HRTEM) images of Na-SGU-7



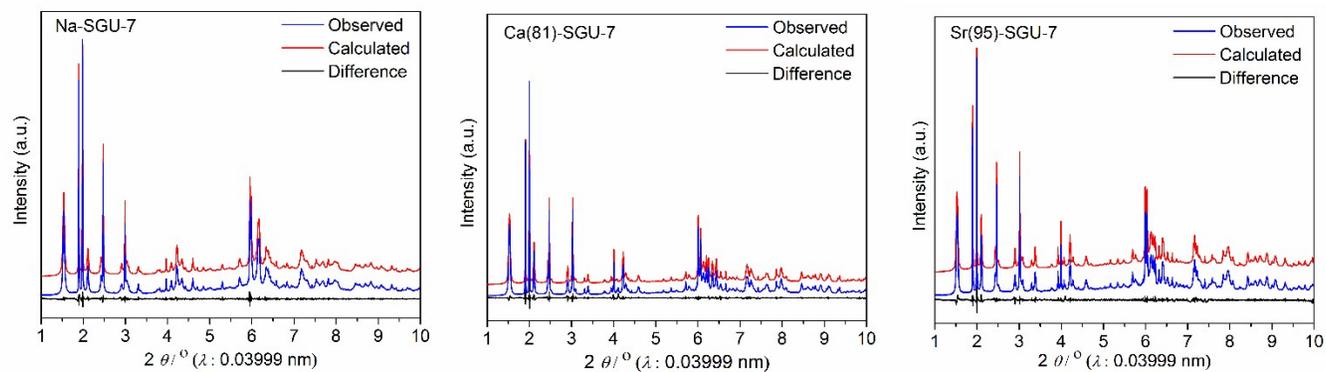
**Fig. S7** HRTEM images of Na-SGU-7. The images taken along different zone axes, (a) [100], (b) [010], (c) [001], and (d) [101]. (e) Image showing the hexagonal 2D nanosheet morphology with indicated crystal axis, and (f) the corresponding TEM image from indicated area.

## SI 8. STEM images of minor phase



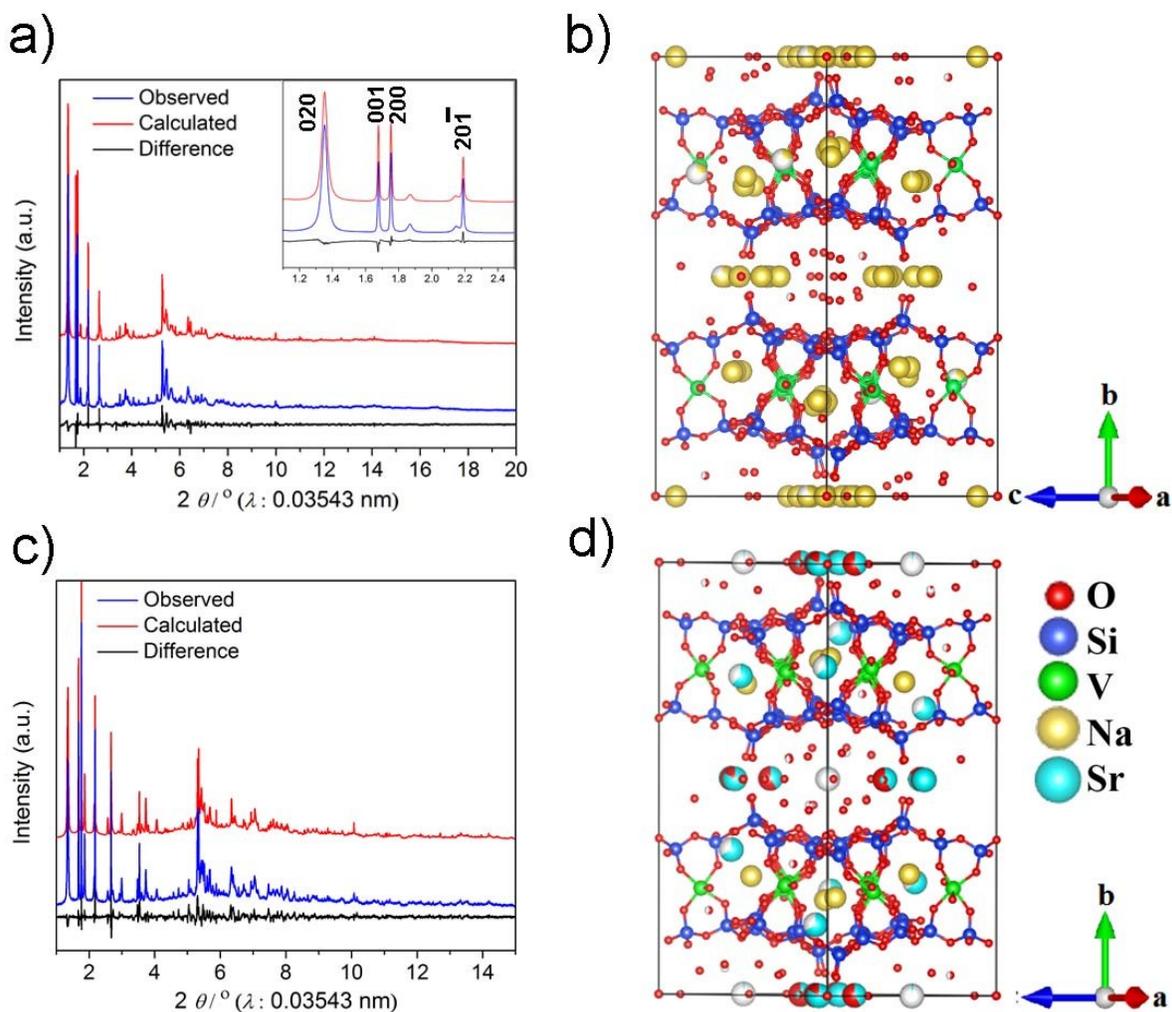
**Fig. S8** STEM images and EDS spectrum of minor impurities in the sample. (a) Low-mag ADF, (b) High-resolution ADF, (c) SAED pattern, (d) high-resolution ABF, (e) ADF images, and (e) EDS spectrum of another phase in the sample. The EDS spectrum shows this impurity is also vanadium silicate but different from SGU-7.

## SI 9. The Pawley unit cell refinement of synchrotron PXRD profiles from SGU-7 samples



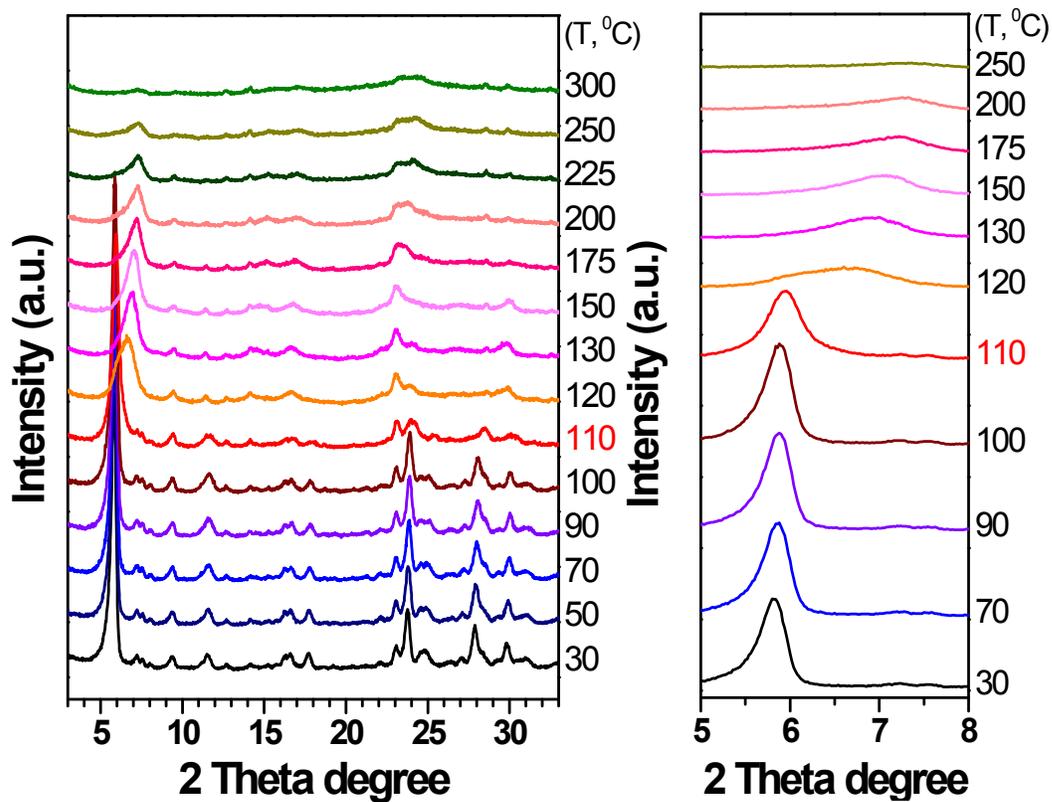
**Fig. S9** Pawley refinements of SGU-7 samples exchanged with different cations. The unit cell parameters are:  $a = 23.5144(2) \text{ \AA}$ ,  $b = 29.924(8) \text{ \AA}$ ,  $c = 12.2871(1) \text{ \AA}$ ,  $\beta = 100.772(9)^\circ$  ( $R_{\text{exp}} = 0.013$ ,  $R_p = 0.06$ ,  $R_{\text{wp}} = 0.085$ ) for Na-SGU-7;  $a = 23.349(1) \text{ \AA}$ ,  $b = 29.941(2) \text{ \AA}$ ,  $c = 12.2479(6) \text{ \AA}$ ,  $\beta = 101.520(1)^\circ$  ( $R_{\text{exp}} = 0.014$ ,  $R_p = 0.032$ ,  $R_{\text{wp}} = 0.048$ ) for Ca(81)-SGU-7;  $a = 23.4286(7) \text{ \AA}$ ,  $b = 29.971(2) \text{ \AA}$ ,  $c = 12.3063(8) \text{ \AA}$ ,  $\beta = 101.441(5)^\circ$  ( $R_{\text{exp}} = 0.015$ ,  $R_p = 0.057$ ,  $R_{\text{wp}} = 0.077$ ) for Sr(95)-SGU-7. The chemical compositions of three samples are  $\text{Na}_{38}\text{H}_2\text{Si}_{80}\text{V}_{20}\text{O}_{220}\cdot 55\text{H}_2\text{O}$ ,  $\text{Ca}_{16.4}\text{Na}_{7.2}\text{Si}_{80}\text{V}_{20}\text{O}_{220}\cdot 74\text{H}_2\text{O}$  and  $\text{Sr}_{19}\text{Na}_2\text{Si}_{80}\text{V}_{20}\text{O}_{220}\cdot 82\text{H}_2\text{O}$ .

## SI 10. Rietveld refinement of Na-SGU-7 and Sr(95)-SGU-7



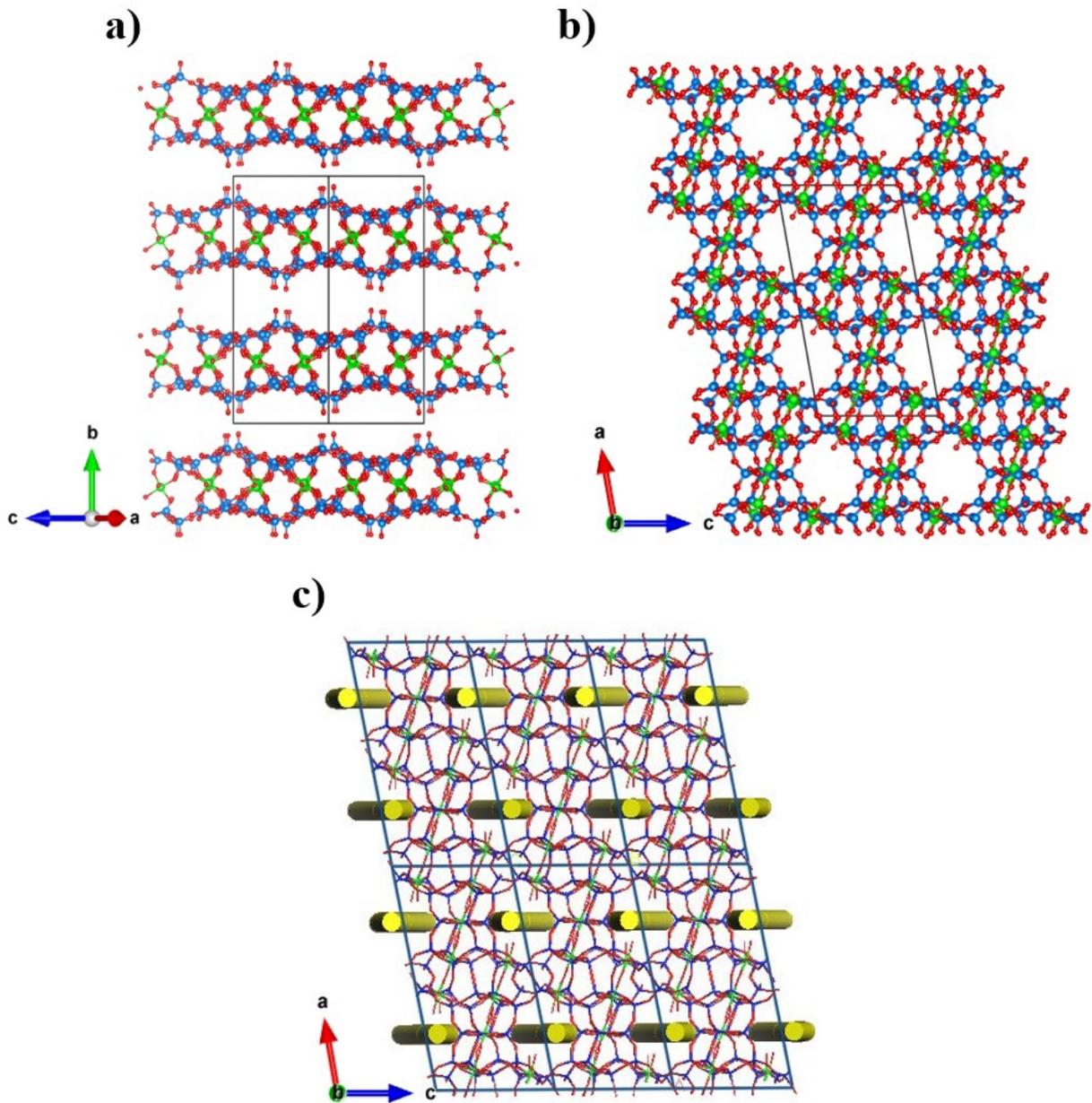
**Fig. S10** (a) Rietveld refinement of Na-SGU-7 (the inset figure shows peak shape anisotropy). The refinement result:  $R_{\text{exp}} = 0.026$ ,  $R_p = 0.06$ ,  $R_{\text{wp}} = 0.086$ ,  $R_{\text{bragg}} = 0.042$ . (b) Refined structural model of Na-SGU-7. (c) Rietveld refinement of Sr(95)-SGU-7 sample ( $R_{\text{exp}} = 0.027$ ,  $R_p = 0.057$ ,  $R_{\text{wp}} = 0.077$ ,  $R_{\text{bragg}} = 0.036$ ) and (d) Refined structure model of Sr(95)-SGU-7 (Yellow: Na, cyan: Sr and red: O. Some cations show partially white color because of partial occupancies).

SI 11. *In situ* PXRD patterns of Na- SGU-7.



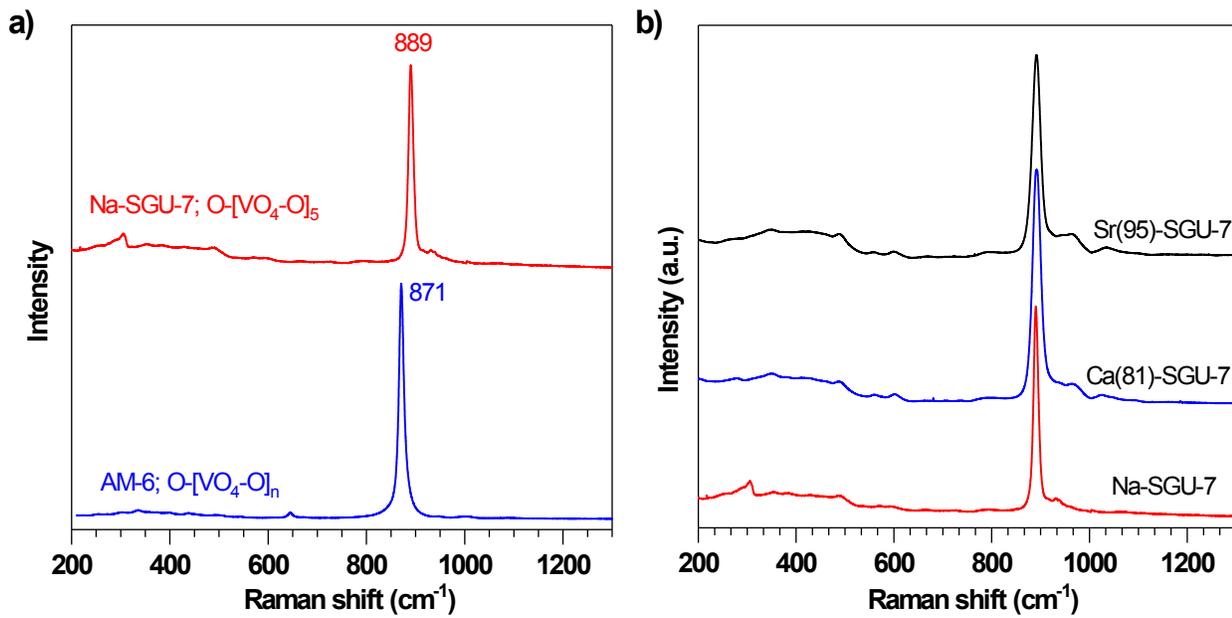
**Fig. S11** *In situ* X-Ray powder diffraction patterns of Na-SGU-7 at mentioned temperature under air. ( $\lambda$ :0.15406 nm)

## SI 12. Crystal structural model of Na-SGU-7



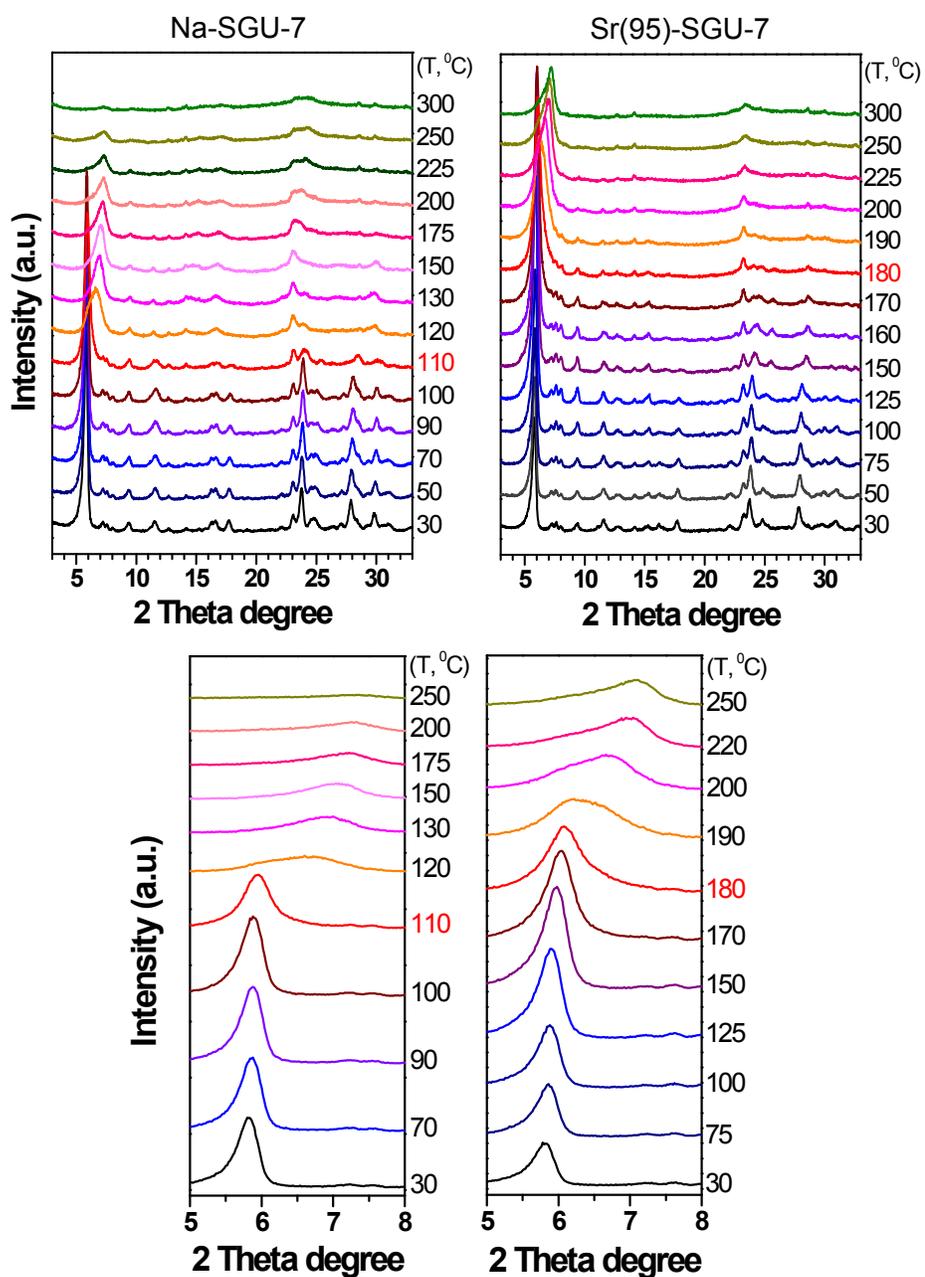
**Fig. S12** Crystal structures of Na-SGU-7 viewed along (a) [101], (b) and (c) [010] directions, cations are omitted for clarity. The green, blue and red spheres represent vanadium, silicon and oxygen atoms, respectively.

### SI 13. Raman spectra



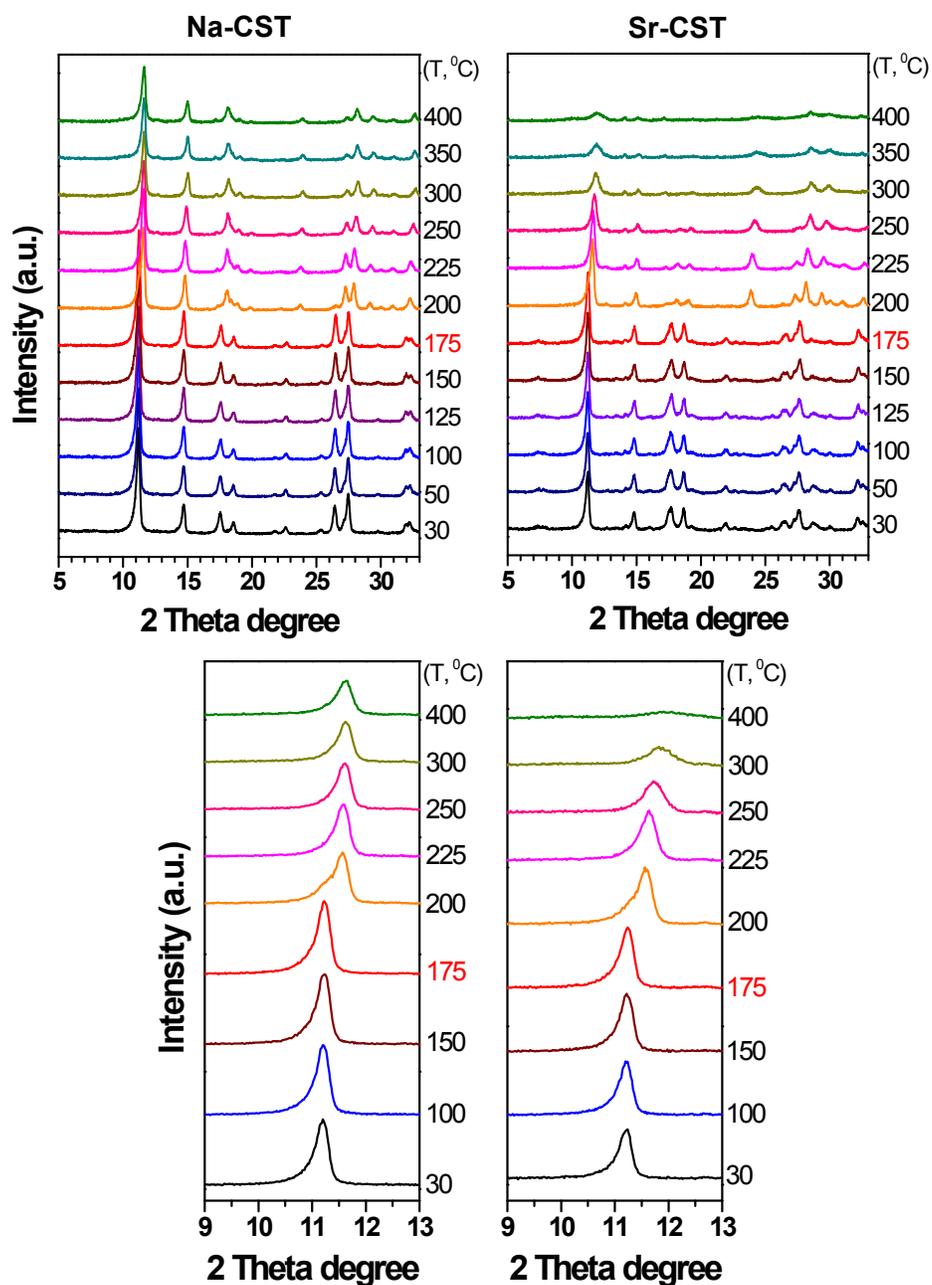
**Fig. S13** (a) A Comparison of Raman spectra of Na-SGU-7 and AM-6 in the 200-1300  $\text{cm}^{-1}$  region. (b) Raman spectra of Na-SGU-7, Ca(81)-SGU-7, and Sr(95)-SGU-7, respectively.

SI 14. *In situ* PXRD patterns of Na-SGU-7 and Sr(95)-SGU-7.



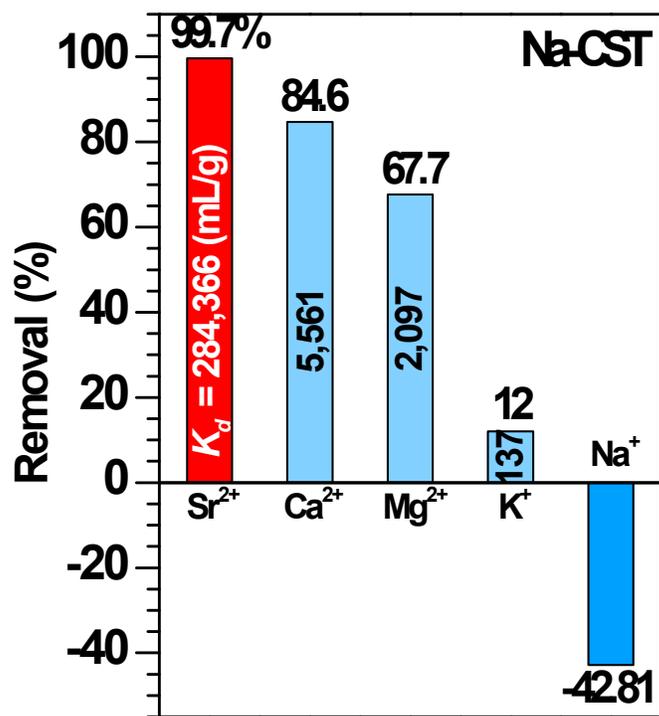
**Fig. S14** *In situ* X-Ray powder diffraction patterns of Na-SGU-7 (left column) and Sr(95)-SGU-7 (right column) at mentioned temperature under air. ( $\lambda$ :0.15406 nm).

SI 15. *In situ* PXRD patterns of Na-CST and Sr-CST



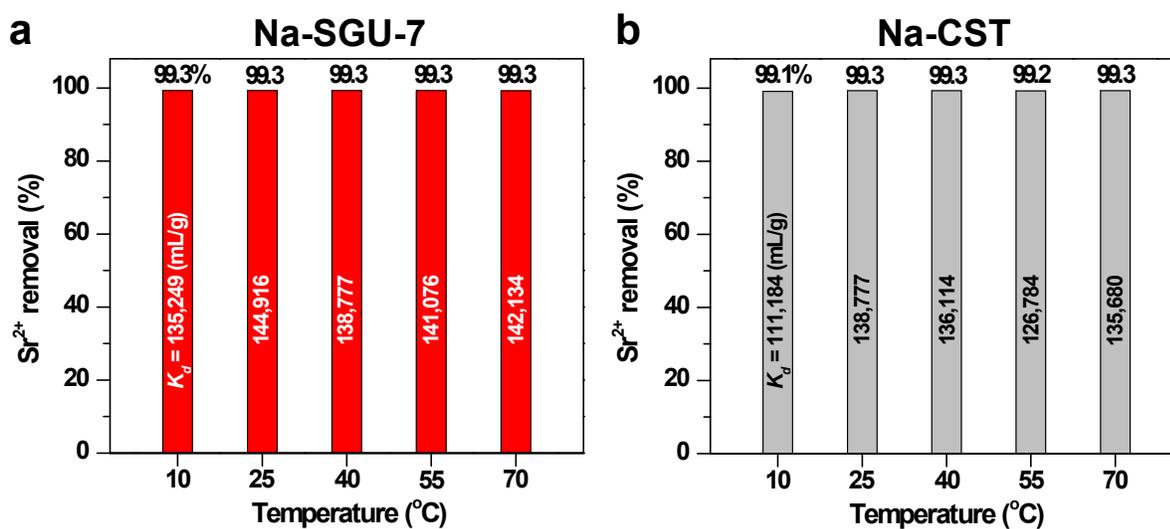
**Fig. S15** *In situ* X-Ray powder diffraction patterns of Na-CST (left column) and Sr-CST (right column) at mentioned temperature under air. ( $\lambda$ :0.15406 nm).

SI 16. The effects of competitive ions on Sr<sup>2+</sup> removal with Na-CST as the Sr<sup>2+</sup>-remover.



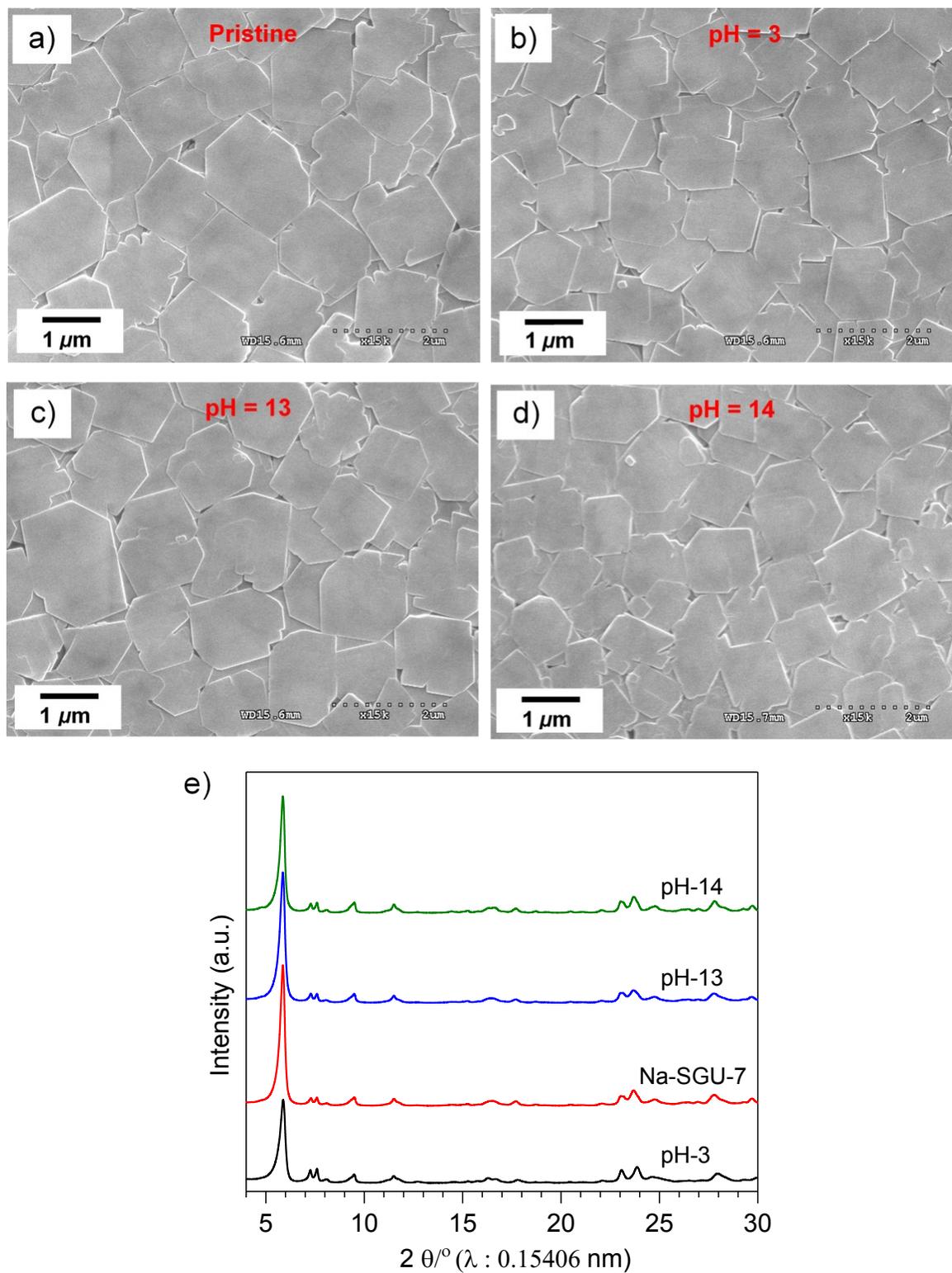
**Fig. S16** Plot of the removed amounts of Sr<sup>2+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, and K<sup>+</sup> and the increased amount of Na<sup>+</sup> in groundwater (in % with respect to the initial amount of each cation) during the removal of Sr<sup>2+</sup> from groundwater with Na-CST under the 1 L/g condition with [Sr<sup>2+</sup>]<sub>i</sub> = 42 ppb after 12 h.

SI 17. Effect of temperature on the performances of Na-SGU-7 and Na-CST.



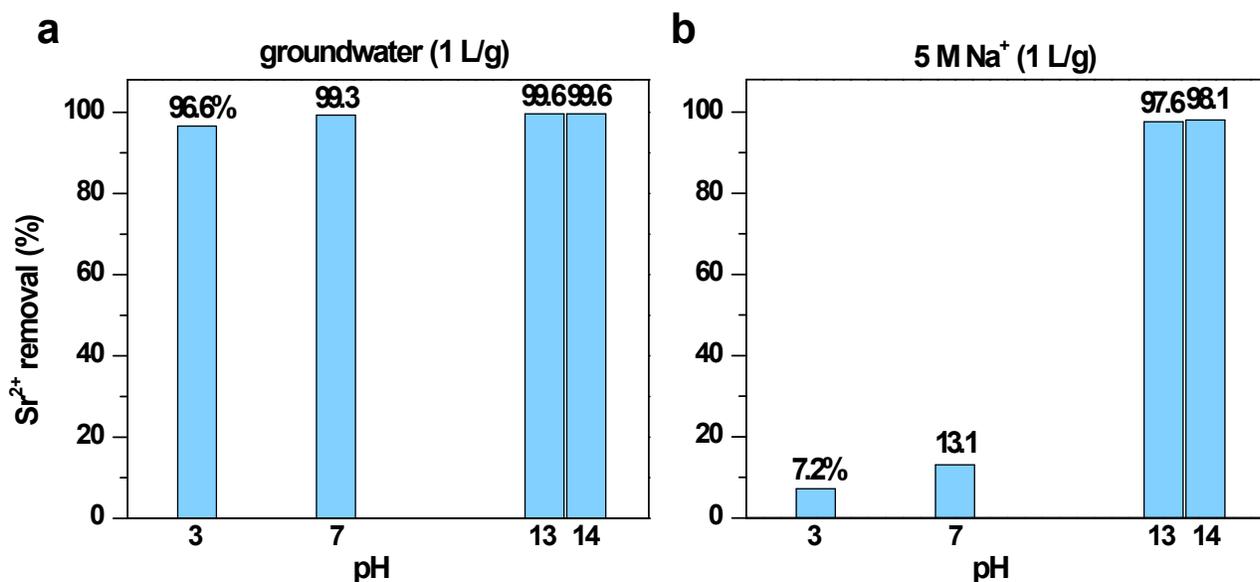
**Fig. S17** Plots of the degree of Sr<sup>2+</sup> removal from groundwater with respect to temperature from 10 and 70 °C after 12 h with (a) Na-SGU-7 and (b) Na-CST as Sr<sup>2+</sup>-removers under the condition of 1 L/g with [Sr<sup>2+</sup>]<sub>i</sub> = 43 ppb.

### SI 18. Stability of Na-SGU-7 under the acidic and basic conditions



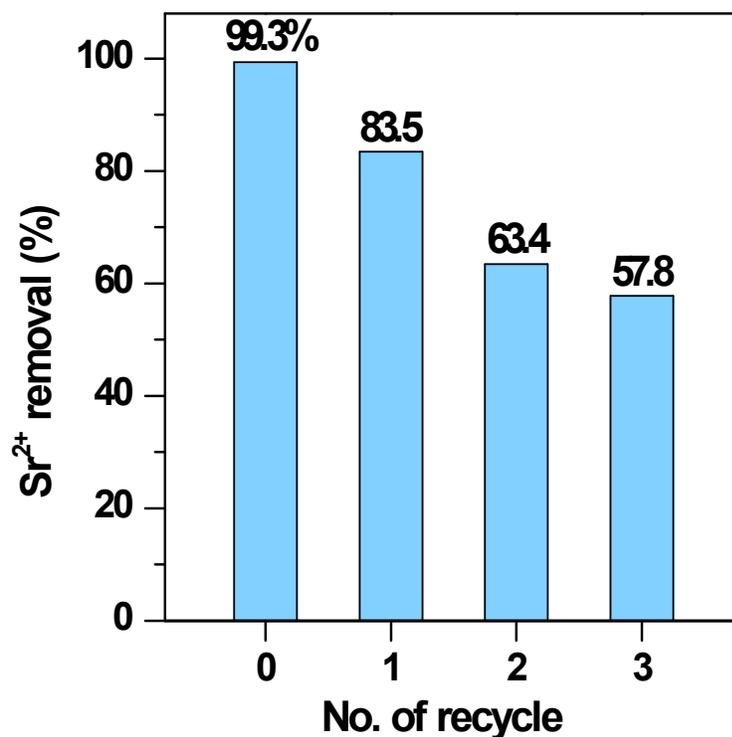
**Fig. S18** Stability of Na-SGU-7 under various pH conditions. SEM images of (a) Pristine Na-SGU-7, after immersion (b) in acidic (pH = 3) and basic solutions of (c) pH = 13 and (d) 14 for 12 h, and (e) their corresponding X-ray diffraction patterns.

SI 19. Effect of pH on the performances of Na-SGU-7 and Na-CST.



**Fig. S19** (a) Plot of the degree of Sr<sup>2+</sup> removal (in %) from groundwater with Na-CST as the Sr<sup>2+</sup>-remover with respect to pH (3, 7, 13, and 14) under the 1 L/g condition with [Sr<sup>2+</sup>]<sub>i</sub> = 1.03 ppm. (b) Plot of the degree of Sr<sup>2+</sup> removal (in %) from 5 M Na<sup>+</sup> solution with Na-CST as the Sr<sup>2+</sup>-remover with respect to pH (3, 7, 13, and 14) under the 1 L/g condition with [Sr<sup>2+</sup>]<sub>i</sub> = 1.03 ppm.

SI 20. Recycle of Na-SGU-7.



**Fig. S20** Plot of the degree of Sr<sup>2+</sup> removal (in %) at room temperature from groundwater with used Na-SGU-7 under the 1 L/g condition with [Sr<sup>2+</sup>]<sub>i</sub> = 42 ppb with respect to the number of recycle. Here, regeneration of used Na-SGU-7 was conducted by washing the previously used Na-SGU-7 (0.5 g) with a fresh 5 M NaCl solution with the volume of 500 mL for 2 h. As noted, the degree of Sr<sup>2+</sup>-removal decreased as the number of recycle increases.