



# Hydrothermal synthesis of pollucite, analcime and their solid solutions and analysis of their properties



Zhenzi Jing<sup>a,\*</sup>, Kunchuan Cai<sup>a</sup>, Yan Li<sup>a</sup>, Junjie Fan<sup>a</sup>, Yi Zhang<sup>a</sup>, Jiajun Miao<sup>a</sup>, Yuqian Chen<sup>a</sup>, Fangming Jin<sup>b</sup>

<sup>a</sup> Key Laboratory of Advanced Civil Engineering Materials, Ministry of Education, Tongji University, 4800 Cao'an Road, Shanghai 201804, China

<sup>b</sup> School of Environmental Science and Engineering, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240, China

## HIGHLIGHTS

- Pure pollucite barely exists in nature due to coexistence of Cs and Na.
- Pollucite, analcime and their solid solutions could be hydrothermally synthesized.
- Most formed solid solutions were found to have similar properties to pollucite.
- Even coexistence in nature, pollucite favors to form due to site preference for Cs over Na.

## ARTICLE INFO

### Article history:

Received 31 July 2016

Received in revised form

3 March 2017

Accepted 4 March 2017

Available online 7 March 2017

### Keywords:

Analcime

Pollucite

Solid solution

Hydrothermal synthesis

## ABSTRACT

Pollucite, as a perfect long-term potential host for radioactive Cs immobilization, barely exists in pure form naturally but in an isomorphism form between pollucite and analcime due to coexistence of Cs and Na. Pollucite could be hydrothermally synthesized with Cs-polluted soil or clay minerals which contain Cs and Na, and it is necessary to study the properties of the synthesis if Cs and Na contained. Pure pollucite, analcime and their solid solutions were hydrothermally synthesized with chemicals, and it was found that the most formed pollucite analcime solid solutions with Cs/(Cs + Na) ratios of 2/6–5/6 had very similar properties in mineral composition, morphology and size, structural water (Cs cations) and coordination environment to pollucite. This also suggests that even coexistence of Cs and Na in nature, pollucite favors to form due to site preference for Cs over Na, which leads to the property and the structure of the most solid solutions similar to that of pollucite.

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## 1. Introduction

Pollucite, a zeolite mineral belonging to the analcime zeolite family [1–4] in terms of their structure and chemical composition, is considered as one of the preferable final storage options for radioactive Cs [5–9] due to the fact that the channel system of pollucite is composed of six oxygen-containing rings of 2.8 Å; the diameter of Cs<sup>+</sup> in pollucite is 3.34 Å; once pollucite is formed, Cs<sup>+</sup> trapped inside is not released from the pollucite structure without breaking the whole framework [10,11]; moreover, the channel system can accommodate more than 40 wt% Cs into its structure.

The synthesis temperature of pollucite is rather high ( $\geq 1200$  °C)

[12,13] now, for which Cs evaporates and equipment corrodes readily. Therefore a low temperature hydrothermal synthesis of pollucite has been developed, and the pollucite could be converted in situ from Cs-polluted incineration ash [14] or Cs-polluted soil (clay mineral) [15] directly for immobilization of Cs in its structure.

Pure pollucite does not exist actually in nature, but in an isomorphism formation between pollucite and analcime (pollucite analcime solid solution) due to natural coexistence of Cs and Na. Therefore, the property and difference between pollucite, analcime and their solid solutions should be study further, especially for the case that Cs-polluted soil (clay minerals) or Cs-polluted incineration ash [14], which contains both Cs and Na, is used to convert into pollucite. On the other hand, it is very difficult to distinguish pollucite, analcime and their solid solutions by XRD [16] and Raman [17] analysis due to the pronounced structural similarity between analcime and pollucite. Therefore it is necessary to synthesize pure

\* Corresponding author.

E-mail address: [zzjing@tongji.edu.cn](mailto:zzjing@tongji.edu.cn) (Z. Jing).

pollucite, analcime and their solid solutions and to study their properties and differences.

Such a research has not been reported in the literature, and the aim of this study was (1) to investigate how to synthesize pure pollucite under hydrothermal conditions with chemicals; (2) to synthesize pure pollucite, analcime and their solid solutions with different ratios of Cs/(Na + Cs); (3) to investigate their properties and differences in mineral composition, morphology, structural water (Cs cations in structure) and coordination environment of the pollucite analcime solid solutions.

## 2. Experimental

### 2.1. Materials

Starting materials were prepared with cesium hydroxide (CsOH; AR, Zhongli Ltd, China) or/and sodium hydroxide (NaOH; AR, Sinopharm Co. Ltd, China), amorphous SiO<sub>2</sub> powder (AR, 100–200mesh, Sinopharm Co. Ltd, China), and aluminum powder (AR, 100–200mesh, Sinopharm Co. Ltd, China). The synthesis proceeded first by mixture of amorphous SiO<sub>2</sub> powder and aluminum powder in the solution prepared with different ratios of cesium hydroxide solution or/and sodium hydroxide, and then by syntheses at temperature of 200 °C for 12 h in a PTFE-lined autoclave. The synthesized materials were filtered in vacuum, washed with distilled water for three times and then dried at 80 °C for 24 h.

### 2.2. Characterization

The synthesized materials were characterized using X-ray diffractometry (XRD, Bruker D8 Advance X-ray diffractometer) with step scan covered angles of 10–55° (2θ) at a rate of 2°/min. Scanning electron microscopy (SEM, Sirion 200) with transmission electron microscopy (TEM, 2100F field emission) was conducted for the investigation of the microstructure and morphology of synthesized materials. TEM specimens were prepared by dispersing the powders in ethanol with ultrasonic agitation and dripping one drop of the suspension onto a copper grid with a supported carbon film and then allowing it to dry on filter paper. The chemical compositions were investigated through X-ray fluorescence spectroscopy (XRF, SRS3400). The thermal behaviors of the analcime, pollucite and solid solution were investigated by simultaneous differential scanning calorimetry and thermogravimetry (DSC/TG, TA Q600), specimens were analyzed at a heating rate of 10 °C min<sup>-1</sup> from room temperature to 1000 °C.

In order to investigate the coordination environments of the synthesized pollucite and its solid solutions, extended X-ray absorption fine structure (EXAFS) measurements were also performed at 4W1B endstation in Beijing Synchrotron Radiation Facility (BSRF), which runs 2.5 GeV with an average electron with current from 150 mA to 250 mA, in transmission mode for pollucite and in fluorescence excitation mode for pollucite analcime solid solution respectively because the content of Cs for the latter is quite low. The storage ring was run at 2.5 GeV with an average electron current of 250 mA. The incident X-ray was monochromatized by a double-crystal Si(111) monochromator, and all data were collected at ambient temperature.

## 3. Results and discussion

### 3.1. Hydrothermal synthesis of pollucite

Pollucite (CsAlSi<sub>2</sub>O<sub>6</sub>·H<sub>2</sub>O), with the stoichiometric Cs/Al/Si ratio of 1/1/2, was hydrothermally synthesized first with different contents of CsOH or SiO<sub>2</sub>, i.e. Cs/Al ratio ranging from 1/1 to 5/1 and Al/

Si ranging from 1/1 to 1/3 respectively.

#### 3.1.1. Effects of Cs/Al on pollucite formation

Fig. 1 shows XRD patterns of specimens synthesized at 200 °C for 12 h with different Cs/Al ratios of 1/1–5/1 at a fixed Al/Si ratio of 1/2. The all patterns with Cs/Al ranging from 1/1 to 5/1 were very similar and their peaks also well matched in position and relative intensity to a standard XRD pattern for pollucite phase (PDF2 files No.29-0407, ICDD) between 10° and 55° of 2θ. No other phases were found by XRD within the detection limit of the diffractometer. It should be noticed that the peaks of the stoichiometric specimen with Cs/Al/Si ratio of 1/1/2 is highest, standing for the best crystallinity.

#### 3.1.2. Effects of Al/Si on pollucite formation

The stoichiometric Cs/Al of 1/1 was chosen to discuss and the XRD patterns of specimens synthesized at 200 °C for 12 h with Al/Si changing from 1/1 to 1/3 are shown in Fig. 2. There were pollucite and zeolite-D (Cs) formed at Al/Si ratio of 1/1, in which the crystallinity of the formed pollucite was lower. The zeolite-D (Cs) forms

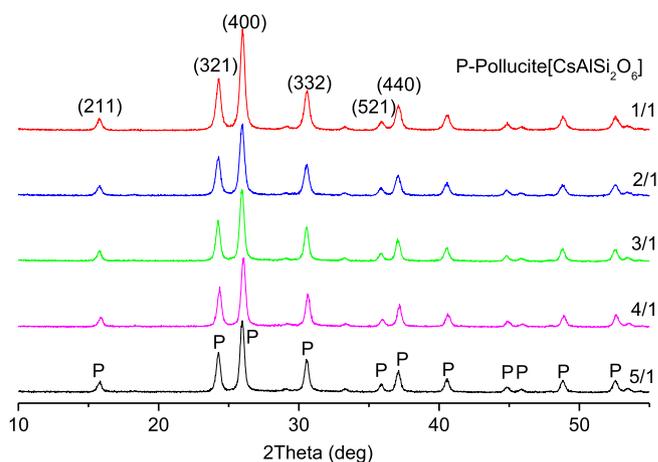


Fig. 1. XRD patterns of the specimens synthesized at 200 °C for 12 h with different Cs/Al ratios.

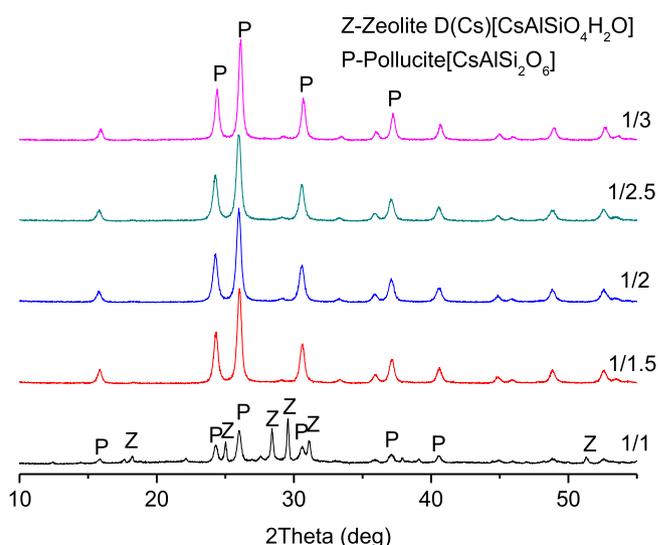
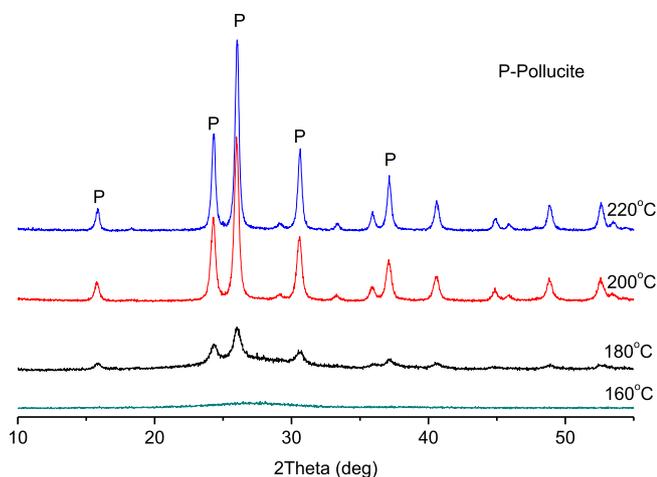


Fig. 2. XRD patterns of the specimens cured at 200 °C for 12 h with different Al/Si ratios.



**Fig. 3.** XRD patterns of the specimens synthesized at different temperatures for 12 h with a Cs/Al/Si of 1:1:2.

readily because the Cs/Al/Si ratio of 1/1/1 is same as that of zeolite-D (Cs) ( $\text{CsAlSiO}_4 \cdot \text{H}_2\text{O}$ ). With increasing Al/Si (increasing of  $\text{SiO}_2$  content), pure pollucite phases formed at Al/Si ratios of 1/1.5–1/3, and the crystallinity of the synthesized pollucite showed no obvious differences.

### 3.1.3. Effects of curing temperature on pollucite formation

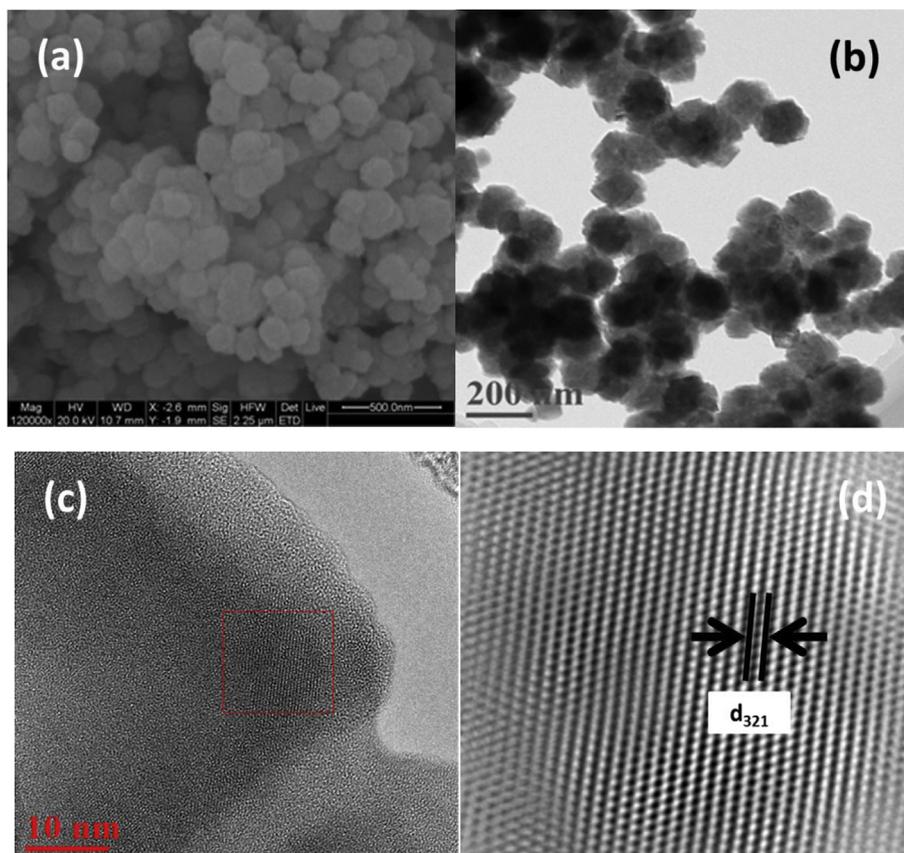
Temperature is an important factor in hydrothermal synthesis. Fig. 3 shows XRD patterns of specimens synthesized with the stoichiometric Cs/Al/Si of 1/1/2 at different temperatures for 12 h.

There was no peak found at 160 °C, showing no pollucite formed at a low temperature of 160 °C; while at 180 °C obvious peaks for pollucite were observed, showing that pollucite tended to form. When it comes to 200 °C, the peaks became more sharp, standing for a higher crystallinity of pollucite phase formed. A similar behavior could also be observed when temperature reached to 220 °C, showing the resemblance in crystallization.

Fig. 4a shows the SEM micrographs of the above specimen synthesized at 200 °C for 12 h with Cs/Al/Si of 1/1/2, and homogeneous irregular pollucite particles were observed with dimensions of 100–200 nm. In the TEM image (Fig. 4b), some formed cubic grains should correspond to the cubic syngony of pollucite. The cubic grains were further observed by HRTEM and the results Fig. 4(c and d) showed that even though the grains were prone to beam damage in the HRTEM analysis, some orderly lattices could still be observed clearly (Fig. 4c). The IFFT (Inverse Fast Fourier Transform) result after a FFT (Fast Fourier Transform) is shown in Fig. 4d. The atoms were highly ordered and the most clear interplanar spacing measured was 0.366 nm, which stands for the lattice plane (321) and corresponds directly with the second highest peak ( $2\theta = 24.3^\circ$ ) in the XRD pattern (Fig. 1).

### 3.2. Pollucite, analcime and solid solution formations with different ratios of Cs/Na

In order further to investigate the pollucite or/and analcime formations when  $\text{Cs}^+$  and  $\text{Na}^+$  coexisted in the strating materials, the stoichiometric (Cs + Na)/Al/Si of 2/1/2 was chosen to synthesize pollucite, analcime and their solid solutions. Fig. 5 shows the XRD patterns of specimens synthesized at 200 °C for 12 h with different



**Fig. 4.** SEM micrograph (a), TEM micrograph (b) and HRTEM results (c,d) of the specimen at 200 °C for 12 h with Cs/Al/Si of 1:1:2.

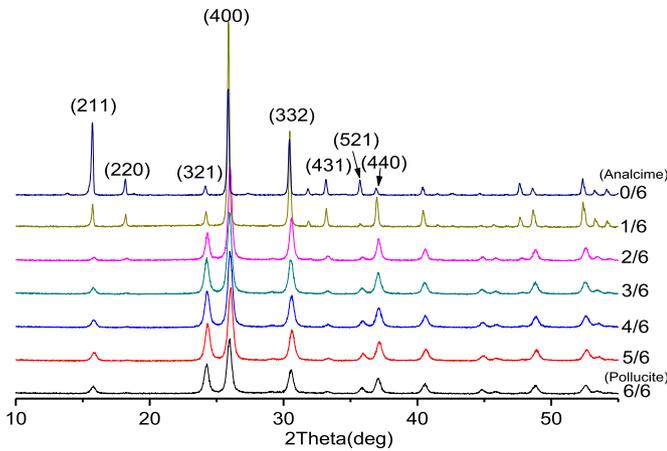


Fig. 5. XRD patterns of the specimens curd at 200 °C for 12 h with different Cs/(Na + Cs) ratios.

Cs/(Cs + Na) ratios changing from 0/6 (analcime) to 6/6 (pollucite). From Fig. 5, the diffraction peaks for both pollucite (6/6) and analcime (0/6) were very similar, and only a difference existed in the peak intensity, in which the strongest crystal face diffraction peak (400) and the second one (211) are located at  $2\theta$  of 26° and 16° respectively for analcime, while for pollucite the strongest peak

(400) and the second one (321) are located at  $2\theta$  of 26 and 24° respectively [17,18]. Both pollucite and analcime have a similar structure, and the only difference is in that for pollucite the Cs cation cannot occupy the Na site of analcime due to its large size, but the H<sub>2</sub>O site instead [18]. It should be noted that only the peak intensity with the ratio of Cs/(Cs + Na) of 1/6 was similar to that of analcime (0/6), while the others (2/6–5/6) seemed very similar to that of pollucite (6/6). This might reflect a structural similarity to pollucite for the most formed solid solutions (2/6–5/6) or a priority for pollucite formation when Cs and Na cations coexist. In order further to reveal the similarity to pollucite for the most formed solid solutions with Cs/(Cs + Na) ratios of 2/6–5/6 in crystal morphology and size, structural water content (Cs cation amount) and coordination environment in structure, the solid solution with Cs/(Cs + Na) ratio of 3/6 was selected, which contains half Cs and half Na, to represent the solid solutions with Cs/(Cs + Na) ratio of 2/6–5/6 for convenience in the subsequent studies.

### 3.3. Structural differences

#### 3.3.1. SEM microstructures

Fig. 6a shows the SEM micrograph of the specimen synthesized at 200 °C for 12 h with a Cs/(Cs + Na) ratio of 0/6 (analcime), the uniform and spherical grains were observed clearly, with the diameter of ~20 μm. For Cs/(Cs + Na) ratio of 1/6 shown in Fig. 6b, similar spherical particles with size of 5–20 μm were found, and its

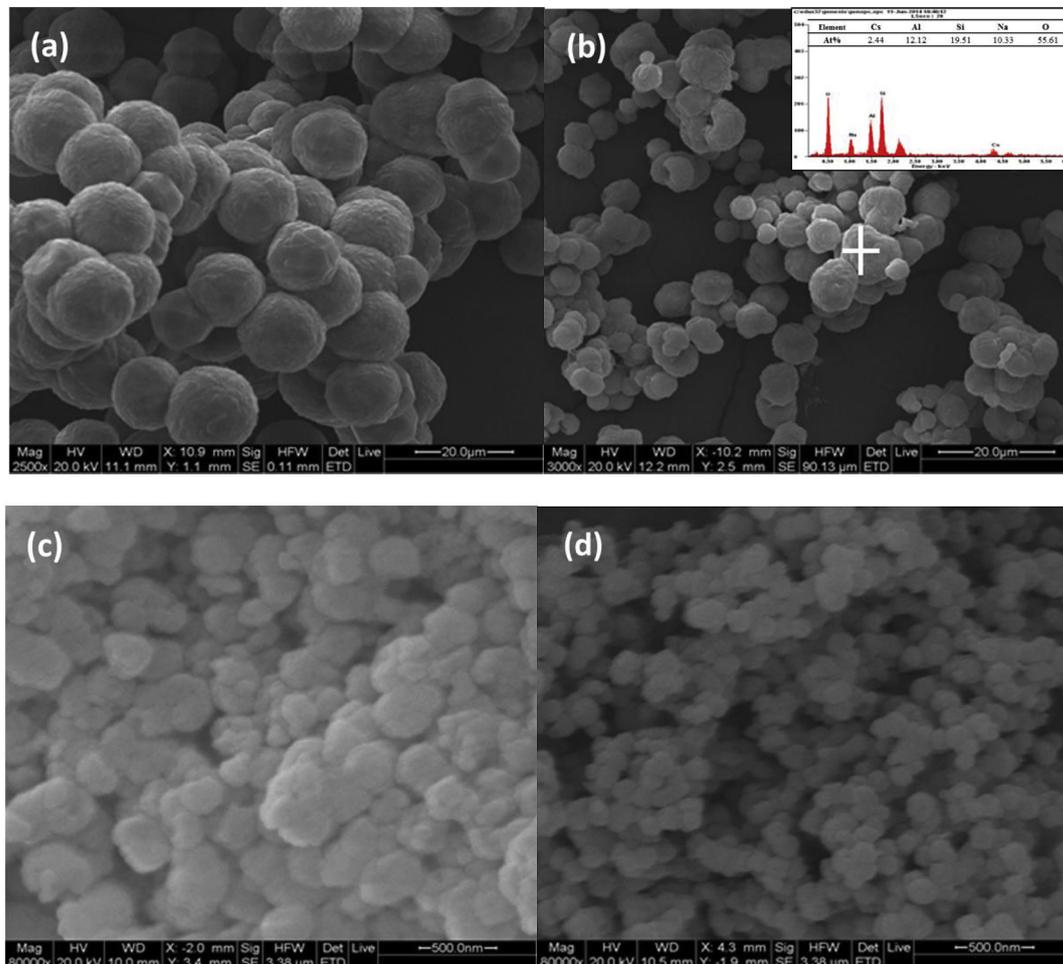


Fig. 6. SEM micrograph of the specimen at 200 °C for 12 h with a Cs/(Cs + Na) ratio of (a) 0/6; (b) 1/6 and the energy dispersive X-ray (EDX) spectrum on the marked spot; (c) 3/6; and (d) 6/6.

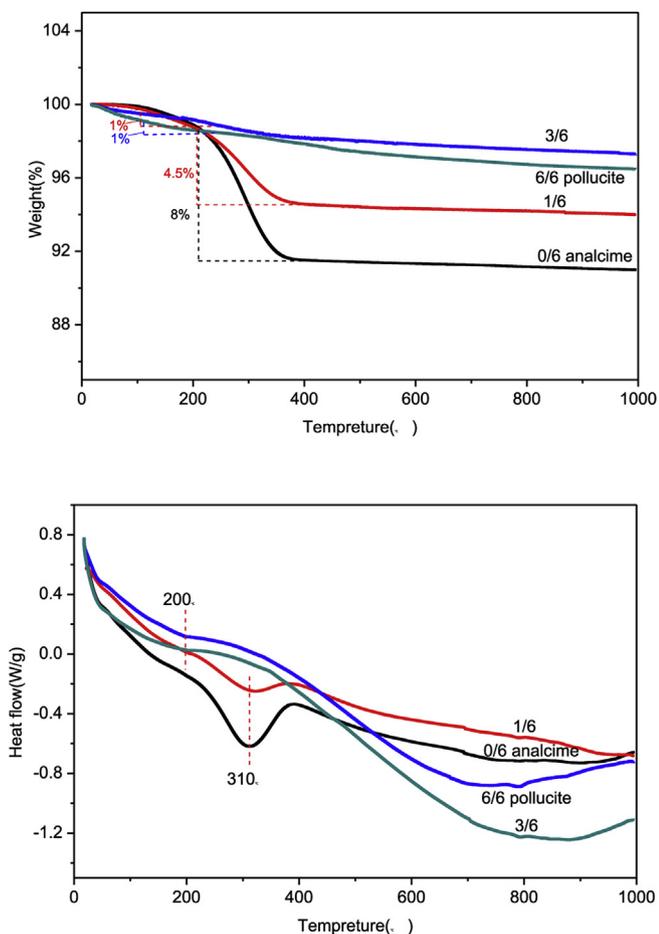


Fig. 7. TG and DSC curves of the specimens with different Cs/(Cs + Na) ratios.

EDX spectrum revealed that a Cs/Na atom ratio of 0.23 (2.44/10.33) for the crystallized material was larger than Cs/Na of 0.2 (1/5) in the starting material, which suggests that although Na is confirmed to enter into the structure, Cs seems to be preferred to be incorporated in the structure even though there is more Na than Cs in absolute

terms. The small difference in crystal size might be due to Cs cations have been entered into the structure. As Fig. 6c shown, the shape of the particles with a Cs/(Cs + Na) ratio of 3/6 was irregular, and the particle size with dozens to hundreds of nanometers was much smaller than that with Cs/(Cs + Na) ratio of 1/6 (Fig. 6b), but similar to that of pollucite shown in Fig. 6d, which reflects similar crystal morphology and size to that of pollucite (6/6).

### 3.3.2. TG-DSC analysis

The mutual substitution of Cs, Na and H<sub>2</sub>O in the structure results in analcime (NaAlSi<sub>2</sub>O<sub>6</sub>·H<sub>2</sub>O), pollucite (CsAlSi<sub>2</sub>O<sub>6</sub>·H<sub>2</sub>O) and pollucite analcime solid solution ((Cs,Na)AlSi<sub>2</sub>O<sub>6</sub>·nH<sub>2</sub>O) formations, however the larger Cs cation cannot occupy the Na site of analcime but the H<sub>2</sub>O site instead for pollucite. With increasing Cs cation, more and more sites of water molecules would be occupied by Cs cations, and thus the structural water could reflect the Cs cation amount in the structure. In order to study the similarity degree to pollucite or analcime for the synthesized solid solutions, the structural water was investigated by TG-DSC. As seen from TG curves shown in Fig. 7, trace of structural waters (~1%) exist in pollucite and the solid solution made from Cs/(Cs + Na) ratio of 3/6, suggesting that they have almost same Cs cation amount in the structures. Much more water (8%) exists in analcime, and a similar phenomenon (4.5% lost water) could be seen for the case of Cs/(Cs + Na) ratio of 1/6, suggests that some sites of water in the structure have been occupied by Cs cations for the case of 1/6. The dehydrating temperature (~200 °C) of DTG curve (Fig. 7) for both pollucite and Cs/(Cs + Na) ratio of 3/6 are much lower than that for analcime and Cs/(Cs + Na) ratio of 1/6 (310 °C) due to the less stable structure for pollucite than the original one for analcime. The consistency in structural water content for the solid solutions with Cs/(Cs + Na) ratios of 3/6 and pollucite suggests that almost same Cs cations (structural water) enter into the structures of pollucite and the most solid solutions (2/6–5/6).

### 3.3.3. EXAFS analysis

EXAFS spectra can provide information on the coordination environment for a certain atom structure [19,20], and the fluorescence Cs L<sub>III</sub>-edge EXAFS spectra in k-space and the corresponding Fourier transformed EXAFS spectra in R-space are shown in Fig. 8. The EXAFS curves of the solid solutions with Cs/(Cs + Na) ratios of 1/

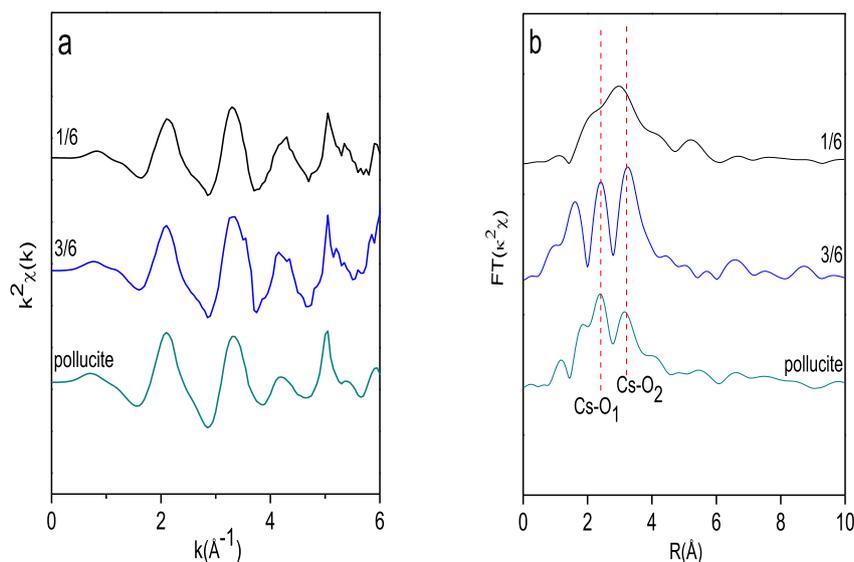


Fig. 8. EXAFS spectra in k-space (a) and corresponding fourier transformed spectra in R-space (b) of pollucite and pollucite-analcime solid solution.

**Table 1**  
Chemical compositions for specimens with different ratios of 0/6, 3/6 and 6/6.

	Analcime (0/6)	Cs/(Cs + Na) = 3/6	Pollucite (6/6)
Na <sub>2</sub> O	15.8	1.94	–
Al <sub>2</sub> O <sub>3</sub>	21.9	14.1	17.0
SiO <sub>2</sub>	55.6	39.5	38.5
Cs <sub>2</sub> O	–	41.2	41.1

6 and 3/6 presented in k-space (Fig. 8a) look similar to that of pollucite (6/6), suggesting that the coordination environments around Cs atom for pollucite and their solid solutions are similar. The EXAFS curves in R-space ( $R = 2\text{--}4\text{Å}$ ) (Fig. 8b) show two independent peaks for pollucite (6/6), i.e., the first peak at 2.5 Å assigned to the Cs–O<sub>1</sub> shell and the second peak at 3.5 Å to the Cs–O<sub>2</sub> shell, which agrees with that reported by Naray-Szabo [21], indicating that Cs is indeed embedded in the structure of the synthesized pollucite and combined with oxygen atoms rather than physically adsorbed in the pores. The position of two obvious peaks of the solid solution with a Cs/(Cs + Na) ratio of 3/6 is almost same as that of pollucite, suggesting very similar coordination environments for them. However, the position of two peaks for the one with a Cs/(Cs + Na) ratio of 1/6 shifts left a little, suggesting some changes in the (Al,SiO<sub>4</sub>) tetrahedron framework (both Cs–O<sub>1</sub> shell and Cs–O<sub>2</sub> shell become short). In pollucite, Cs-sites in the structure are symmetrical [3]; while Cs-sites in the pollucite analcime solid solution with ratio of 1/6 are partially occupied by Na and H<sub>2</sub>O, and because of weaker Na–O bonds, the oxygen is more close to Cs, which results in shorter Cs–O bonds. The results suggested that the most solid solutions (Cs/(Cs + Na) ratio of 2/6–6/6) have almost same coordination environments with pollucite, while the only one with Cs/(Cs + Na) ratio of 1/6 seem to have some changes in coordination environment. According to the above XRD, SEM (TEM), TG–DSC and EXAFS analysis results, the most formed solid solutions with Cs/(Cs + Na) ratios of 2/6–5/6 were proved to have very similar properties to that of pollucite, which also suggests that even coexistence of Na and Cs, Cs favored to enter into the structure of the pollucite analcime solid solutions due to site preference for Cs over Na.

### 3.3.4. XRF analysis

The chemical compositions of the synthesized analcime, pollucite and solid solution with a Cs/(Cs + Na) ratio of 3/6, are presented in Table 1, which clearly show the differences in composition among them. It should be noted that the content of Cs<sub>2</sub>O of the solid solution with a Cs/(Cs + Na) ratio of 3/6 (41.2%) is nearly same as that of pollucite (41.1%), which shows that almost same Cs cations have been entered into the structure of the solid solution of 3/6 as that in pollucite. This suggests that although the same content of Cs<sub>2</sub>O and Na<sub>2</sub>O were added to form the pollucite analcime solid solutions, much more Cs<sub>2</sub>O (41.2%) than Na<sub>2</sub>O (1.94%) was found to enter into the structure of the solid solution with a Cs/(Cs + Na) ratio of 3/6, which is almost same as that in pollucite (41.1%). This also reflects that Cs cation participates in the reaction more readily than Na cation. The site preference for Cs over Na has resulted in the most pollucite analcime solid solutions with a Cs/(Cs + Na) ratios of 2/6–5/6 having very similar coordination environment, morphology, structural water and mineral property to that of pollucite.

## 4. Conclusions

Pure pollucite, analcime and their solid solutions have been synthesized by a low temperature hydrothermal technology and their properties and differences were investigated by XRD, SEM (TEM), TG–DSC and EXAFS. The experimental results can be summarized as follows:

Pollucite could be synthesized at relatively low temperature (<220 °C) by hydrothermal synthesis with chemicals. Cs/Al ratios seemed to exert a small influence on the pollucite formation in range of 1/1–5/1, while a larger Al/Si ratio ( $\geq 1/1.5$ ) and a higher temperature ( $\geq 200$  °C) favored to form a purer pollucite. A well crystallized pollucite, with dimensions of 100–200 nm, could be obtained at 200 °C for 12 h.

Pollucite, analcime and their solid solutions could be synthesized with different Cs/Na ratios. XRD analysis showed that the most formed solid solutions with Cs/(Cs + Na) ratios of 2/6–5/6 had very similar mineral compositions to pollucite (ratio of 6/6), while only the one with Cs/(Cs + Na) ratio of 1/6 was similar to that of analcime (0/6).

SEM (TEM), TG–DSC and EXAFS analyses further revealed that the morphology and size, the structural water contents (Cs cations in structure), and the coordination environments of the most solid solutions (2/6–5/6) were very similar to that of pollucite, which suggests a similarity not only in mineral composition but also in structure. XRF results also indicated that much more Cs cations than Na cations was found to enter into the structure of the solid solution with a Cs/(Cs + Na) ratio of 3/6 (2/6–5/6), which suggested Cs cation participates the reaction to form solid solution more readily than Na cation during pollucite analcime solid solutions formation.

As such, even coexistence of Na and Cs, Cs favors to participate in the reaction due to site preference for Cs over Na, which results in the properties of the most formed pollucite analcime solid solutions similar to that of pollucite.

## Acknowledgement

The work reported here was supported by and the National Natural Science Foundation of China (Nos. 51272180, 51072138). The EXAFS beam time was granted by 4W1B end station of Beijing Synchrotron Radiation Facility, Institution of High Energy Physics, Chinese Academy of Sciences.

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