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**Fig. S1.** The pristine surface state of biotite sample under electron microscope and AFM loading sample platform, of which (a) the SEM image of biotite sheet, (b) the biotite (001) surface, (c) the probe NP-S10 within contact mode, and (d) the probe MESP within tapping mode.



**Fig. S2.** The obvious features on the biotite (001) surface for 96 h incubation in pH 4.0  $H_2O$ , CA, and CA+NaCl solution at 25 °C, of which (a), (d), and (g) are height images, (b), (e), and (h) are phase images, (c), (f), and (i) are linear sections from the corresponding height images.



**Fig. S3.** The evolution of bulges and bumps on biotite (001) surface reacting with CA+NaCl solution. (a)–(f) are amplitude mode images and the iconography in (d) is phase image.



**Fig. S4.** FSEM images show the morphological changes of the biotite (001) surface after reacting with (a) pH 4.0  $H_2O$ , (b) CA, and (c), (d) CA+NaCl solutions for 168 h at 25 °C.



**Fig. S5.** Segments on the biotite (001) surface after reacting with pH 4.0 CA+NaCl solution for 168 h at 25 °C. (A) and (B) are amplitude mode image and height image, respectively and the scale bar is 15  $\mu$ m × 15  $\mu$ m. (i) and (ii) are linear sections from the corresponding sites.

Compound	Composition (%)
MgO	21.74
Al <sub>2</sub> O <sub>3</sub>	16.36
SiO <sub>2</sub>	42.44
K <sub>2</sub> O	10.71
TiO <sub>2</sub>	1.61
FeO	7.14
Total	100.00

Table S1. Chemical composition of the biotite used in this study, determined by electron probe microanalysis