

1. . Experimental

1.1 Materials and methods

Preparation of the activated coal gangue

The study was performed on the coal gangue of approximate dimensions of 2mm*2mm*2mm, which was dried to the air-dry state. It has shown that the spontaneous combustion coal gangue is partially activated (Meng et al. 2013) The raw materials were ground and washed with distilled water. The chemical activation was performed with HCl and KOH to obtain the modified CG. According to the literature, the concentrated large amount of acid helps to form a large surface area. The new mineral phase can form when the concentration of KOH is above 0.5 mol L⁻¹ (Becerro et al. 2009). Firstly, 25g of coal gangue was mixed with 70 mL of 36% HCl and 0.5 mol L⁻¹ KOH, respectively. The heterogeneous systems were under magnetic stirring and then allowed to stand for 24 hours in the solutions. After that, the materials were filtered off and washed repeatedly with distilled water until the neutral pH. The solids were dried in oven under 70 °C for 1 day, obtaining the activated coal gangue samples. The samples are referred to as follows.

CCG - Combusted coal gangue washed by distilling water

HCG (grey powder) - CCG s activated with HCl

KCG (red powder) - CCG activated with KOH.

1.2 Characterizations

Infrared spectra (4000–400 cm⁻¹, resol. 0.5 cm⁻¹) were recorded on a Varian 660 Fourier-transform infrared (FTIR) spectrometer using KBr pellets and the transmission

technique. Powder X-ray diffraction (PXRD) measurements were carried out on a Rigaku Miniflex X-ray diffractometer. The data were collected from 3° to 50° with a turning speed of 2.0 °/min. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) was performed using a NETZSCH Jupiter® STA 449F3 instrument. The measurements were done under air (20 mL·min⁻¹) at 35–800 °C with a speed of 10 K·min⁻¹. The elemental analysis and surface states of the samples were analyzed by X-Ray photoelectron spectroscopy (XPS), which were carried out on a Thermo Fisher K-Alpha spectrometer (ESCALAB 250Xi, Thermo Fisher, USA) using a monochromatized Al Ka radiation ($h\nu = 1486.6$ eV). Brunauer-Emmett-Teller (BET) specific surface area was investigated by N₂ adsorption-desorption method at 77 K on a ASAP 2020. The samples were activated at 300 °C overnight under vacuum. The scanning electron microscope (SEM) and energy-dispersive X-ray spectroscopy (EDS) analyses were performed on a Zeiss Sigma 300 scanning electron microscope.

1.3 Adsorption test

5 mg adsorbents and 30 mL U(VI) (50 mg·L⁻¹) solution with appropriate concentration and pH value were added to a 250 mL flask in an air bath oscillator (180 rpm) at the appropriate temperature. The pH was adjusted with 0.1 M negligible volume of HCl and NaCO₃. After adsorption of U(VI), the residual concentration of U(VI) in the solution was analyzed by a spectrophotometer at 650 nm (5 mL chloroacetic acid/sodium acetate buffer solution and 1 mL Arsenazo (III)) and ICP-MS (7700 Series, Agilent USA). The equilibrium adsorption capacity of U(VI) (q_e) and removal efficiency (%) was calculated as follows:

$$q_e = \frac{(C_o - C_e)V}{m} \quad (1)$$

$$\% = \frac{(C_o - C_e)}{C_o} \times 100\% \quad (2)$$

Where C_o and C_e are the initial and equilibrium concentrations of U(VI) (mg/L), respectively, V is the liquid phase volume (L), and m is the amount of adsorbent (g).

All the flasks were capped to avoid evaporation.

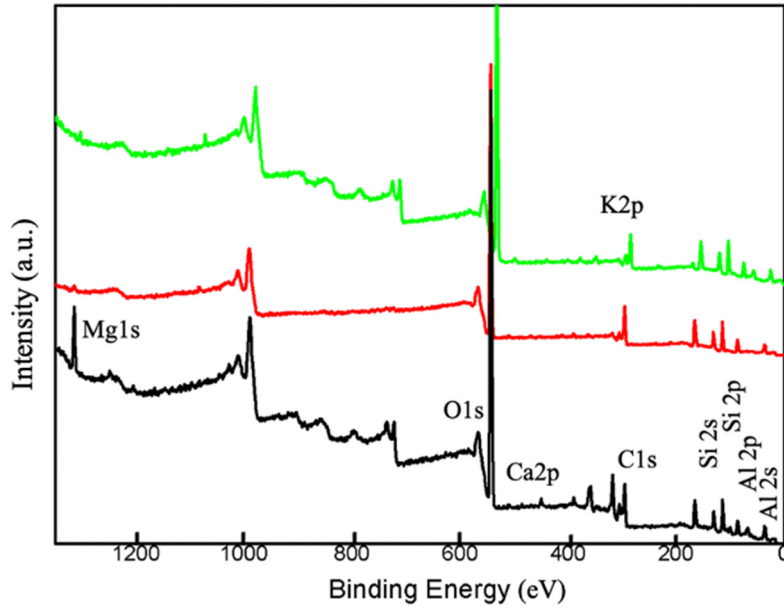


Figure. S1 The XPS of CCG (black), HCG (red) and KCG (green), showing the characteristic peaks.

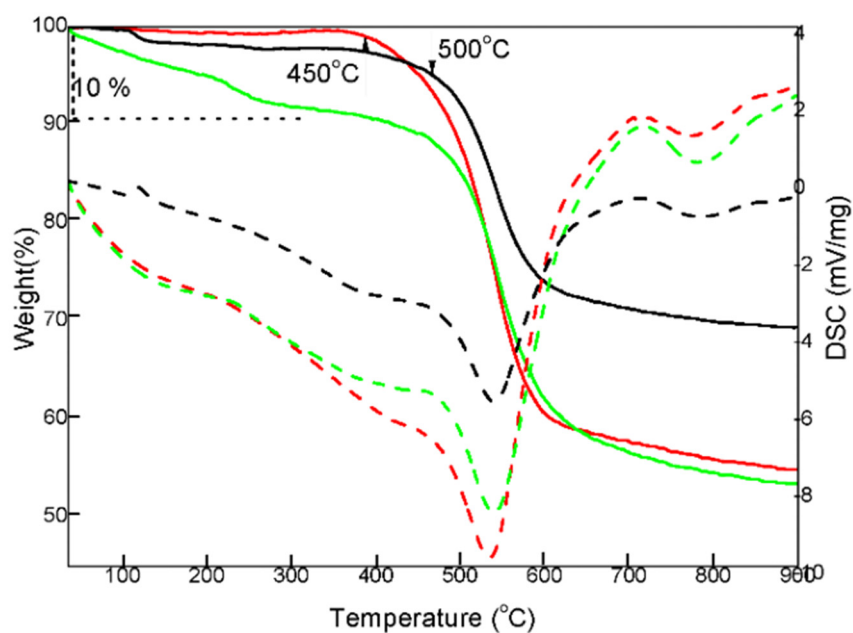


Figure. S2 The TGA (Line) and DSC (dots) of CCG (black), HCG (red) and KCG (green).

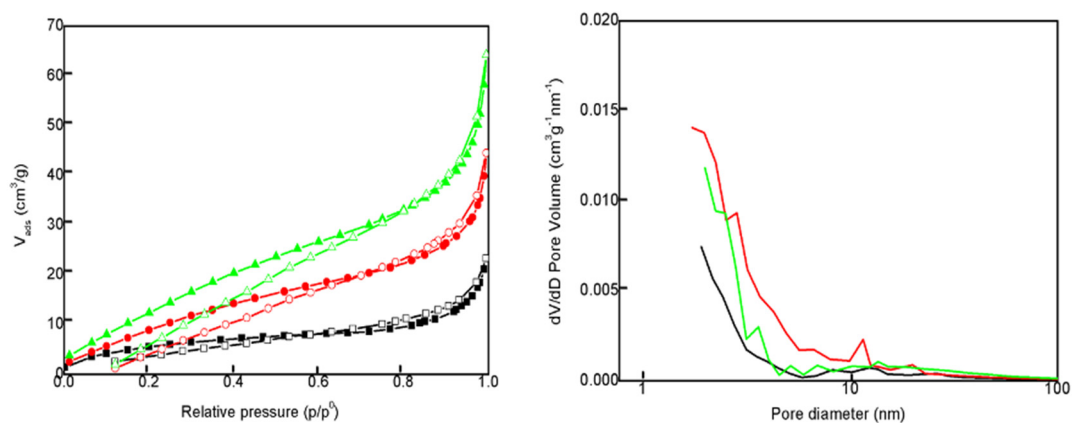


Figure. S3 The N₂ adsorption isotherms at 77K (Left) and the pore size distributions using BJH analysis (Right). CCG (black), HCG (red) and KCG (green)

Table S1 Adsorption capacity under variant pH condition (from 2 to 6) (mg/g)

Sample. \ pH	2	3	4	5	6
CCG	6.2	15.3	19.8	37.0	39.6
HCG	1.2	16.2	23.4	42.3	43.8
KCG	2.1	52.6	66.3	123.7	140

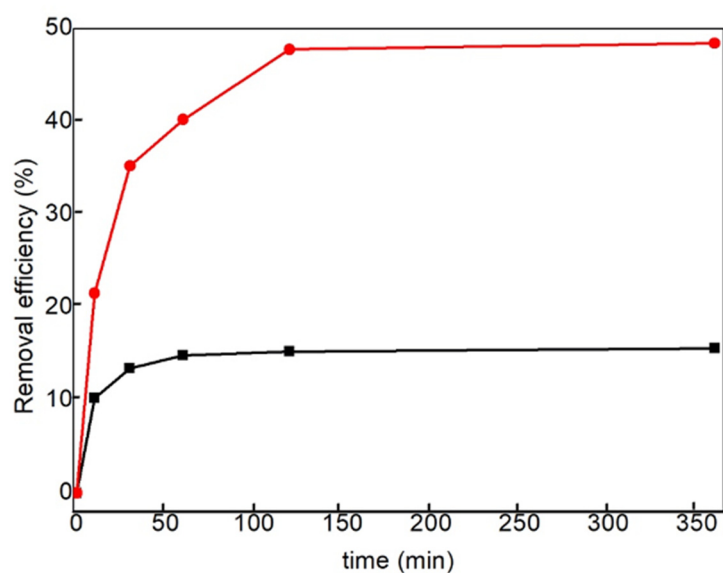


Figure. S4 Effects of adsorption contact time on the removal efficiency of U(VI) onto HCG (black) and KCG (red).

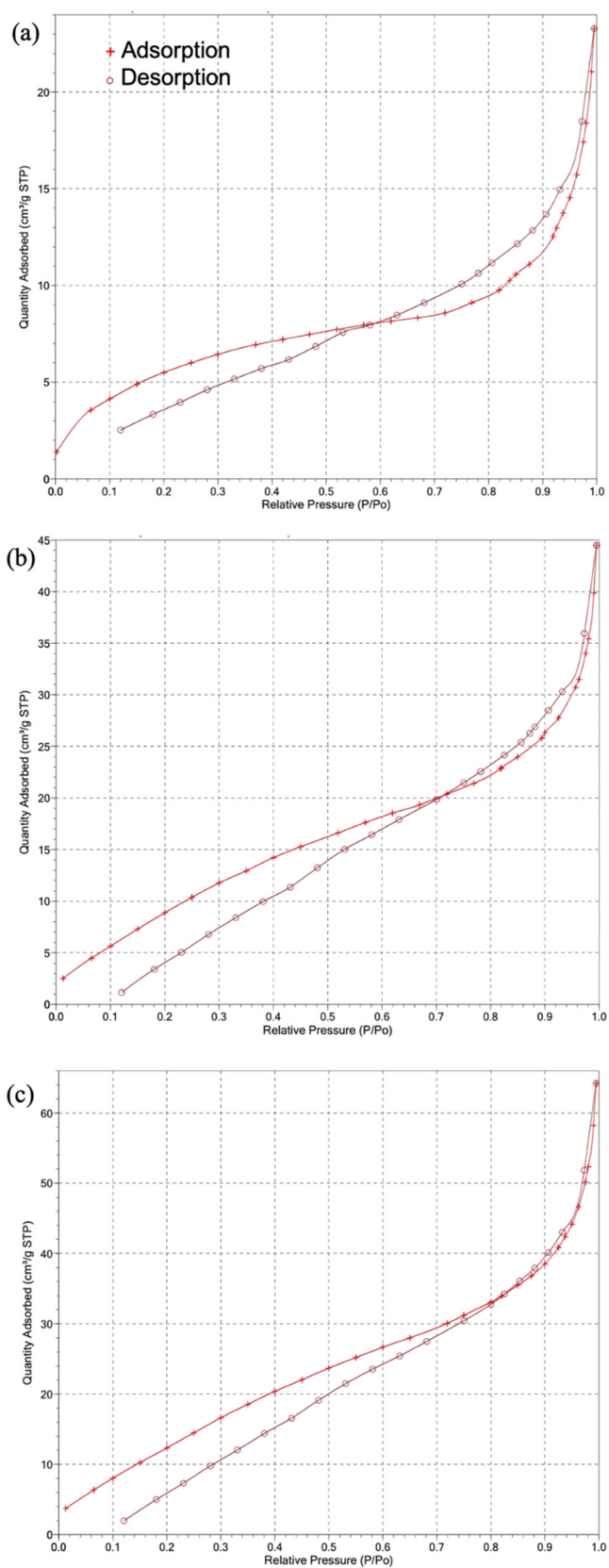


Figure. S5 The isotherm linear plots of CCG (a), HCG(b), and KCG(c).