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The Element Geochemistry of Coal Mine Drainage Pollution in Coal Mine from Northern Jiangsu Province

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Abstract The content of environmentally-sensitive elements released from coal gangue and the element geochemistry of coal mine drainage (CMD) in Kongzhuang coal mine were investigated on the basis of a soaking test. Results show that the content of volatile elements Hg and As released from the newly-explored gangue is higher than that from the old-gangue, and the Hg and Pb contents from the roof are higher than that from the floor. Hg, Se, P, Cd, Pb and SO_4^{2-} contents in CMD exceed the environment quality standards. Generally, the microorganism may play an important role in the pollution of CMD.
Keywords coal mine drainage, element geochemistry, pollution

Introduction

As a kind of fossil fuel, coal plays an important part in the industrial production and daily life in China, with about 74% of total primary energy and 60% of chemical materials derived from coal production (Dai et al.,2012). Coal, on one hand, prospers the economy, while on the other hand, leads to great environmental problems. It includes air pollution like $PM_{2.5}$ (Ren et al. 2009, Si et al. 2014), heavy-metals-induced soil infertility and water pollution like acid mine drainage (AMD) (Maccausland and McTammany 2007, Doulati Ardejani et al., 2010). At some mine sites, water quality downstream of the mine is the most challenging environmental factor during mine operations and after mine closure. There are several ways that mining could have a negative impact on downstream water quality including: the increase in suspended sediment, the release of environmentally significant trace element and the release of low pH acid mine drainage (Pope et al. 2010).

It is of vital importance to study the mechanism of hazardous elements released from coal gangue and coal, in that it is the main source of trace elements in coal mine drainage, especially AMD. This research explored the releasing behavior of environmentally significant elements and ions by an immersion test of four coal gangues and made an assessment of the pollution degree of coal mine water in Kongzhuang coal mine, Northern Jiangsu province.

Sampling and analytical method

Four kinds of coal gangue samples: newly-explored gangue, old-gangue, roof and floor were collected at Kongzhuang coal mine, Northern Jiangsu province, China. The specific description of the four samples is listed in Table 1.

Table 1 *Sampling description and experiment*

sample	Sampling location	Weight	Experiment	Description
New-explored gangue	Kongzhuang coal mine	4kg	Soaking test	Argillaceous, new surface
Old-gangue	Coal gangue hill	4kg	Soaking test	Sandy, oxidized surface
7435-roof	No.7 coal seam	3kg	Soaking test	Argillaceous, new surface
7435-floor	No.7 coal seam	3kg	Soaking test	Argillaceous, new surface

All samples were crushed to 200 mesh and then put into four 2500ml reagent bottles. By adding 2000ml deionized water with initial pH 7, a 7 day's soaking experiment was performed in China University of Mining and Technology. A XRF analysis was performed on the gangue samples and the water samples to determine the environmentally-sensitive elements and irons like SO_4^{2-}, F^-, As, Hg, Cr, Cd, Pb.

Coal mine water samples in Kongzhuang mine were also collected to compare with the environment standards. 6 kinds of coal mine water were collected including:
(1) mine water that directly extracted from coal mine;
(2) deposited water that was collected after the deposition of mine drainage;
(3) osmosis water that was collected after the reverse osmosis treatment;
(4) the upper washing water in the coal washing plant;
(5) the lower washing water in the coal washing plant;
(6) mine water in the pool of No.7 coal mine.

The liquid samples were analyzed in China University of Mining and Technology. 7 elements and ions including Hg, Pb, Cr, Cd, As, F^- and SO_4^{2-} were determined. Among them, Pb, Cr, Cd, As were determined by ICP-MS, Hg was determined by mercuryvapourmeter; F^- was determined by ion selective electrode for fluoride; SO_4^{2-} was analyzed by iodometry.

Results

The 7 days' soaking test of 7 elements is shown in fig.1. Among them Cd does not release until the 5th day. Also, the 7 day's total release value is compared with the GB5085.3-2007 in table 2. As we can see, elements and irons like Hg and SO_4^{2-} exceed the standards.

The comparison between element contents in mine water and environmental standards is shown in table 3. As can be seen, many elements like Hg, Se, P, Cd, Pb and SO_4^{2-} exceed the environmental standards.

Table 2 *Total release value of 7elements and irons in the 7 days soaking test*

Elements	New-explored gangue	Old-gangue	Roof	Floor	GB5085.3-2007
Hg	2.1672	1.7176	2.4344	1.4885	0.1
Pb	0.006	0.008	0.015	0.01	5
Cd	0.004	0.003	0.004	0.004	1
Cr	0.046	0.043	0.043	0.047	5
As	0.084	0.046	0.022	0.071	5
F^-	7.929	8.53	9.551	16.85	100
SO_4^{2-}	54.65	64.22	60.06	58.99	5

GB 5085.3-2007: Identification for extraction toxicity

Table 3 *Chemical analysis of coal mine water*

	K1	K2	K3	K4	K5	K6	GB 3838-2002
As	0.0373	0.0449	0.0279	0.0418	0.0387	0.0383	0.05
Hg	0.0079	0.0094	0.0054	0.0049	0.0072	0.0046	0.00005
Se	0.0544	0.0231	0.028	0.0419	0.05	0.0483	0.01
P	0.0411	0.0294	0.0227	0.0616	0.0402	0.0299	0.02
Zn	0.0143	0.0007	0.009	0.0053	0.0235	0.0026	0.05
Cd	0.0016	0.0014	0.0008	0.0017	0.0018	0.0013	0.001
Pb	0.0314	0.0374	0.0371	0.0494	0.0472	0.0363	0.01
Mn	0.0064	0.0007	0.001	0.0009	0.1285	0.012	0.1

Continued Table 3

	K1	K2	K3	K4	K5	K6	GB 3838-2002
Fe	0.0477	0.0005	0.0009	0.0003	0.0324	0.0281	0.3
Cr	0.003	0.003	0.0032	0.0035	0.0033	0.0022	0.01
Cu	0.0055	<0.0006	<0.0006	<0.0006	0.0154	<0.0006	0.01
Cl^-	156.28	194.5	13.9	233.56	215.34	130.25	250
SO_4^{2-}	629.75	629.75	bd	1065.22	1024.88	631.39	250
NO^{3-}	nd	1.91	0.5	3.3	nd	2.15	10
COD	nd	5.88	2.22	9.55	nd	7.72	15

Where, bd=below deadline; nd=no data; COD=chemical oxygen demand; K1, 2, 3, 4, 5, 6=liquid sample (1), (2), (3), (4), (5), (6), respectively; GB 3838-2002=environmental quality standard for surface water.

Discussion

Fig. 1 shows that the roof has a high release value of heavy metals like Hg and Pb, but releases slowly (from the 3^{rd} on, the release of Hg becomes the highest of the four gangues, while, for Pb, it is 6^{th} day). In terms of volatile elements like Hg and As, newly-explored gangue seems to have a higher release value. By contrast with the other three gangues, the release value of F in the floor is the highest. Pb and Cd show slow releasing speed in comparison with the other elements. Overall, the releasing behavior of most elements studied is almost consistent from the four gangues and all of the elements increase steadily in the 7 days and do not reach the releasing equilibrium.

Table 2 shows that the total release value of most 7 elements and irons does not exceed the extraction standards (GB5085.3-2007) but most environmentally-sensitive elements in mine water exceed the environmental standards (GB 3838-2002). This is because mine water is formed during a long time of geochemical reaction between coal, coal gangue and water, while 7 days soaking test does not reach the equilibrium of elements releasing.

$$4FeS_2+15O_2+2H_2O=2H_2SO_4+2Fe_2(SO_4)_3 \quad \Delta H=-6184.14 \text{ kJ/mol}, \ \Delta G=-4761.38 \text{kJ/mol} \qquad (1)$$

Those elements exceeding the standard except P show affinity with pyrite (Diehl et al. 2005; Wang et al. 2007; Kolker 2012), therefore, it is indicated that the pollution of mine drainage is associated with the oxidation of pyrite. Reaction (1) is the chemical equation of pyrite oxidation. From the chemical equation we can see that the oxidation of pyrite leads to the acidification of environment. Besides, many hazardous elements that exist in the crystal lattice of pyrite are easily to release into the environment. Also, it is noticeable that the release value of SO_4^{2-} in old gangue is the highest, but old gangue does not have a high release value of volatile elements like As and Hg. This is because the oxidation of pyrite has already happened and many elements are lost in the surrounding environment.

Lan et al. (2000) studied the oxidation of pyrite and found that the release value of SO_4^{2-} in sterile pyrite is only 3.9% that of bacteria-inoculated pyrite. This is because the activation energy of this reaction is fairly high and microorganism works as catalyst in this reaction. Also, it is confirmed that microorganisms play an important role in the oxidation of original minerals and the formation of exogenic minerals in coal (Dai et al. 2003, Yossifova et al. 2011). Moreover, the biodesulfurization that is widely used nowadays (Angel et al. 2001, Cardona and Márquez 2009) and the research on the role that microorganisms play in the formation of coal bed methane (Barnhart et al. 2013, Gallagher et al. 2013) together imply the significance of microorganism in coal.

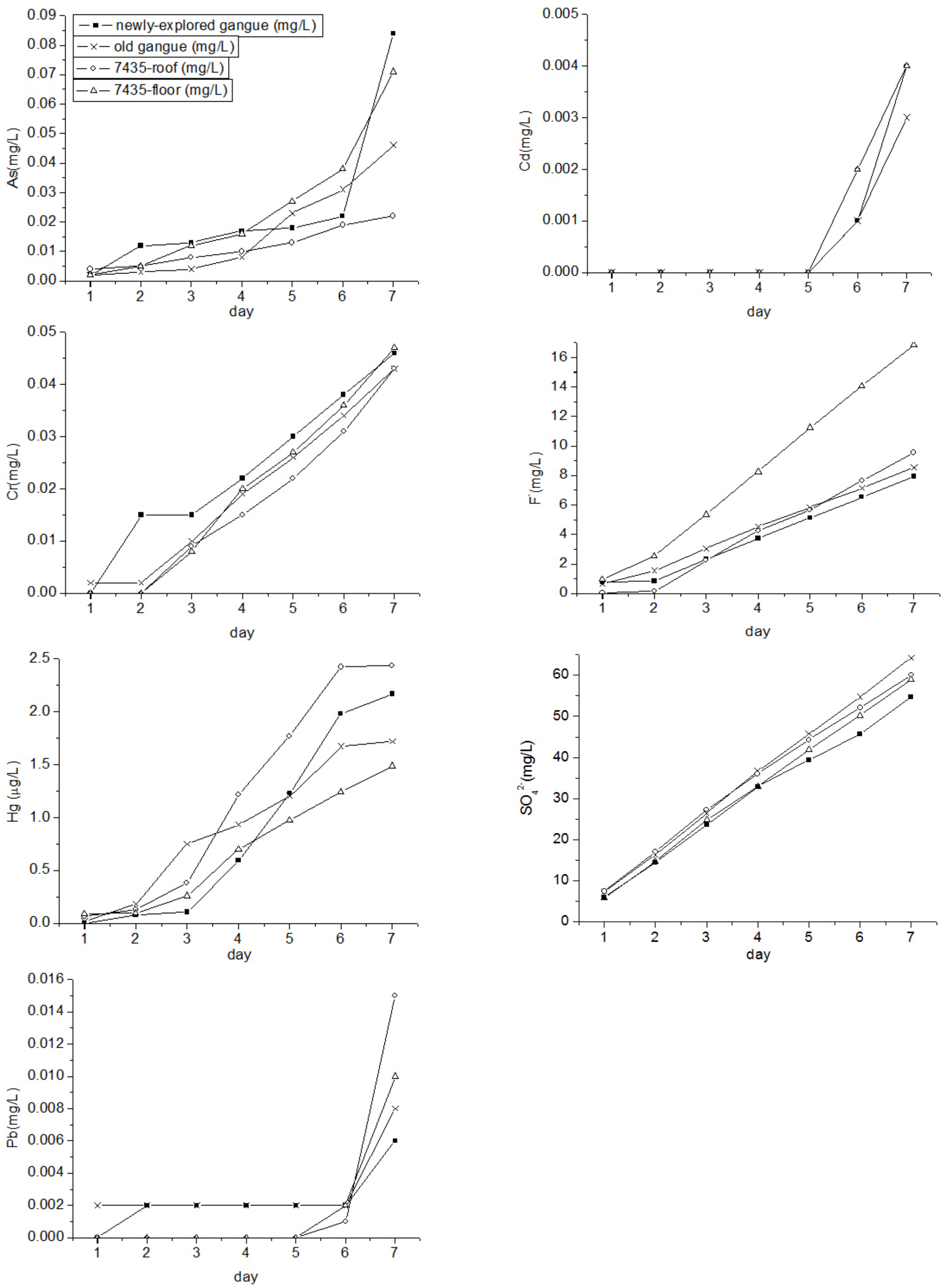

***Fig. 1** 7days' soaking result*

Conclusions

(1) The roof has a high release value of heavy metals like Hg and Pb, while for volatile elements like As and Hg, newly-explored gangue seems release more than the other gangues.
(2) The content of Hg, Se, P, Cd, Pb and SO_4^{2-} in Kongzhuang coal mine water is higher than the environmental standard and the pollution is attributed to the oxidation of pyrite.
(3) Microorganisms maybe play an important role in the oxidation of pyrite and the release of environmental sensitive elements in coal mine drainage.

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