The distribution and origin of hydrogen sulfide abnormal enrichment coal mines in China

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ABSTRACT

Coal mines that contain hydrogen sulfide (H_2S) are widely distributed in China. At present they are mainly located in Zaozhuang in east China, Xin'an, Anyang, and Shuangfeng in central China, Qinshui basin, Taiyuan - Liulin and Wuhai in north China, and Tongchuan-Xianyang area in northwestern China, as well as places such as Shizuishan, Houdon, Changji, Liuhuanggou, and Sikeshu which belong to Xinjiang, mainly in northwest China. These coal mines are showing a rapid increase. The contents of H_2S in coal mines has changed greatly, from 10 ppm to 5000 ppm. The abnormal coal seams are mainly distributed in the Permian Longtan Formation of southwest China, Permian Shanxi Formation and Carboniferous Taiyuan Formation of east, north and central China, and Jurassic Xishanyao and Yan'an Formation of northwest China. The abnormal areas have a favourable overlying strata, and the hydraulic connections between the underground water and other aquifers are weak, therefore, they are good thick reservoir-seal combinations, which are beneficial to the origin of Bacterial Sulfate Reduction (*BSR*) or Thermochemical Sulfate Reduction (*TSR*). The main origin of H_2S in coal mines are *BSR* or *TSR*, and the origin model can be identified synthetically via the tectonic evolution, coal thermal evolution history, isotopic characteristics of carbon and sulfur, methane gas component testing, sulfate sources, and sulfate-reducing bacteria activity features.

KEYWORDS: coal mine; hydrogen sulfide; distribution; origin; *BSR*; *TSR*

1. INTRODUCTION

Disasters and the potential hazards caused by the abnormal enrichment of hydrogen sulfide (H_2S) frequently occur in coal mines in China as well as abroad. In recent years, more than 30 sudden releases of H_2S has been recorded in 10 provinces in China, such as in Guizhou, Sichuan, Shandong, Henan, Shanxi, Shaanxi, Inner Mongolia and Xinjiang, etc. These incidents have caused more than 20 casualties. In addition, a considerable number of coal mines experiencing the abnormal enrichment of the low content of H_2S do not cause serious casualties, and thus do not call up widespread concern. H_2S acts as a kind of potent nerve poison upon contact. The main target organs, on which its toxicity has an effect, are the central nervous system and the respiratory system. The tissues that are the most sensitive to the toxicity are the brain and the contact sites of mucous membranes, thus, H_2S can cause systemic poisoning, or even respiratory arrest (Ma et al., 2008; Liu et al., 2011). In the long term, it poses a serious threat to the miners' occupational health and safety. Therefore, conducting research like geological surveys, distribution characteristics, and causes of coal mines

that are rich in H_2S is an urgent issue and has important significance.

2. DISTRIBUTION OF COAL MINES OF HYDROGEN SULFIDE ABNORMAL GATHERING IN CHINA

In China, coal mines that are rich in H_2S are widely distributed. The statistics concerning partial coal mines containing H_2S are as shown in Table 1.

Table 1: Distribution characteristics of hydrogen sulfide abnormally accumulated in China's coal mines.

Coal mines	Coal forming environment	Reservoir layer	H_2S (ppm)	S (%)	R_0 (%)	Abnormal zone
Xintian of Guizhu	Sealand	Longtan	20	0.98~7.91	3.50~3.80	Drilling process
Longtan of Sichuan	Marine facies	Longtan	1300	2.14~4.52	1.44~1.88	Working surface
Guang'an of Sichuan	Marine facies	Longtan	2500	2.01~5.26	1.21~1.79	Aquifer of roof
Anyang of Henan	Littoral zone	Shanxi	--	0.51~0.98	1.94~3.22	Working surface
Xin'an of	Littoral	Shanxi	--	1.80~2.30	1.02~	Drilling

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Henan	zone				2.32	process
Cuizhuang of Shandong	Sealand	Shanxi	5000	0.65~0.83	0.65~1.25	III mining area
Shandong Bayi	Sealand	Shanxi	1500	2.68	0.59~1.24	Well area
Xiqu of Shanxi	Sealand	Taiyuan	350	Low-medium	0.98~1.60	Working surface
Wuda of Inner Mongolia	Sealand	Taiyuan	400	1.56~6.69	0.81~1.22	Mining area
Xishan of Urumqi	Continental facies	Xishanyao	1500	Low-medium	0.64~0.79	Mining area
Tingnan of Shaanxi	Sealand	Yan'an	56	0.13~2.90	0.51~0.72	Working surface

Currently, the known coal seams that contain high levels of H_2S mainly are Permian Longtan Formation in southwest China, Permian Taiyuan and Carboniferous Shanxi Formations which are in east, north, and central-south China, and Jurassic Xishanyao and Yan'an Formations in northwest China. Xinjiang is not only the region with the most coal mines that contain H_2S in China, but also the region which has the largest number of mines with an abnormal gathering of H_2S in China. In China, most coal mines are hydrogen sulfide-bearing gas reservoirs or micro hydrogen sulfide-bearing gas reservoirs (Dai, 1985).

3. CHARACTERISTICS OF TYPICAL HYDROGEN SULFIDE ABNORMAL GATHERING COAL MINES IN CHINA

The three necessary conditions for the production of BSR are plenty of organic matter, abundant sulfates, and conditions that suit sulfate reducing bacteria to grow and reproduce (Machel, 2001; Cai et al. 2003; Oayer, 1999). The $\delta^{34}S$ value of H_2S in the coal mines mainly depends on the characteristics of sulfur isotopic composition of the sulfur source itself, the abundance of sulfur in the sulfur source, and the genesis model of H_2S in the gas generation medium. The $\delta^{34}S$ value of H_2S and pyrite is generated by *BSR,* as the alienation effect of sulfate-reducing bacteria can result in the large-scale sulfur isotope fractionation, so the isotopic composition of this sulfur is usually negative. The fractionation of the H_2S $\delta^{34}S$ value usually can reach the range of -30‰ - -15‰, which is generally around 20‰ lower than the sulfur isotope of sulfate. Accompanying H_2S, the CO_2 gas component is generally lower, usually less than 5%. The carbon isotope of CO_2 has a lighter characteristic and the δ^{13}_{CO2} value is generally less than -5‰. The shape of the byproduct (pyrite) generated by *BSR* is generally spherical berry. The main hydrocarbon component in

methane gas is CH_4. The CH_4 usually has biogenic features and its accompanying hydrocarbon may have many heavy hydrocarbon components (C_{2+}). The generation and preservation reservoirs of H_2S may have traces of biological effects.

The three basic conditions for the origin of TSR are the higher temperature that coal and rock strata experience, plenty of hydrocarbon organic matter, and the abundant supply of sulfates (Liu et al. 2011; Machel, 2001; Cai et al., 2003; Oayer, 1999; Zui et al. 2006). The $\delta^{34}S$ value of H_2S generated by *TSR* is usually larger. The sulfur isotopic compositions of H_2S are slightly less than or equal to the $\delta^{34}S$ value of the homologous sulfate, and the $\delta^{34}S$ value of H_2S is generally between 10‰ and 20‰. The coal and rock strata generally experience environments of high temperatures above 120°. Accompanying H_2S, the carbon isotope composition of CO_2 is heavier and usually around 0‰. It is often similar to the carbon isotopic composition of carbonates in strata. The shape of pyrite generated by *TSR* is usually cubic or columnar. Since *TSR* is a process which gives priority to the consumption of heavy hydrocarbons, the dry coefficient of the methane gas is usually higher. The content of heavy hydrocarbons in the gas is less, but the content of non-hydrocarbon generally increases.

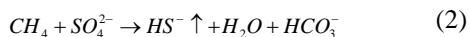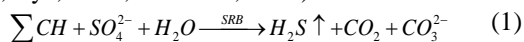*3.1 Characteristics of H_2S in Xiqu coal mine of Shanxi Province*

The location map of Xiqu coal mine of Shanxi Province is shown in Figure 6. The No. 9 coal seam is located at middle and lower part of the Taiyuan formation and is high grey, medium-low sulfur. The average sulfur content in raw coal is 1.37%. The main type of coal is coking coal, and the average value of the maximum vitrinite reflectance of coal ($R_{o,max}$) is 1.60% (Liu et al. 2012). The roof and floor is mostly sandy shale or mud sandstone. The abnormal area of H_2S in the No. 9 coal seam is located in a wide and gentle folding wing of the 19101 and 19102 working face in the south panel. It presents the zonal distribution and its width is about 400 m. In the leeward side of the mining machine, the maximum concentration of H_2S measured is up to 350 ppm[10]. In the gas produced by spontaneous combustion of the ground coal gangue piles, the content of H_2S reaches 89 ppm (Zeng et al. 2012). Within the area of the above two working faces, the geological structure is simple and it has no faults collapse column, but in the northwest side of the faces, it develops a large number of faults and collapsed pillars. The biggest slip of fault is 5.0 m and the slip of the small secondary fault is in the range of 1.0 m - 2.0 m. The inside of the collapsed pillars which formed in Yanshan movement period are often found to be filled by the later generated

epigenetic calcite and pyrite crystals. The subsided columns are finely cemented, and less permeable to water in the deep.

The overlying seam of the abnormal area of H_2S in the No. 9 coal seam is the gob area of No. 8 coal seam whose thickness is 8 m – 12 m. The average sulfur content in No. 8 coal seam is 2.57% (pyrite accounts for 1.58%, organic sulfur accounts for 0.98%) (Song et al., 2000). The distribution of Pyrite is in an irregular grain shape (tetrahedral) and mouldiness. The roof of No. 8 coal seam is composed of limestone, mudstone, carbonaceous mudstone or sandy mudstone. Its floor is composed of sandy mudstone or shale, and some is carbonaceous mudstone or sandstone.

The groundwater of coal mine is acidic, where sulfate ions are abundant and the content is up to 1928.9 mg/L, much higher than that in other water bodies, such as the deep groundwater and the Fen River Reservoir (Yang et al., 2013). The content of Na^+ increases significantly, and the content of HCO^{3-} declines dramatically. The total hardness of water is high and the type of water is $Na-Ca-SO_4$.

Sulfate-reducing bacteria reduction is a process which consumes sulfate ions constantly. The process that generates H_2S is as shown by formulas (1) and (2) (Cai et al., 2003; Oayer et al., 1999; Pan et al. 1992; Rye, 2005; Dai et al., 2000).

$$\sum CH + SO_4^{2-} + H_2O \xrightarrow{SRB} H_2S \uparrow + CO_2 + CO_3^{2-} \quad (1)$$

$$CH_4 + SO_4^{2-} \rightarrow HS^- \uparrow + H_2O + HCO_3^- \quad (2)$$

Formulas (1) and (2) shows that if the H_2S is caused by *BSR*, during the reduction process, the sulfate ions in the water body will usually reduce and the content of CO_3^{2-} and HCO^{3-} will increase. This is in contradiction to the fact that in this area, sulfate ions obviously increase, and the content of HCO^{3-} decreases. Therefore, the *BSR* cause can be initially ruled out. In the groundwater of the original Gujiao No. 2 and No. 3 mines, the values of $\delta^{34}S$ are in the range of 8.2‰ - 11.6‰ (Pan, 1989), with all of the values being positive. They do not have the characteristics of *BSR*. In the process of coal and rock formation, this area does not exhibit magma intrusion, so the magmatic origin can be excluded.

The metamorphic degree of regional coal is high metamorphic focal smoke, and the average value of the maximum vitrinite reflectance of coal ($R_{o,max}$) is over 1.5%. According to the corresponding relation between vitrinite reflectance and temperature, the coal and rock should has experienced a thermal evolution environment with temperatures of over 120°, in order to have the temperature conditions of *TSR*. Regional water and coal petrography are rich in sulfate ions. The roof of No. 8 coal seam is limestone (good reservoir stratum), and it has plenty of

hydrocarbon compounds. Every condition that is necessary for the occurrence of *TSR* is met. The $\delta^{34}S$ value in groundwater is about 10‰, has the characteristics of *TSR* genesis. Therefore, according to the changes of temperature, sulfur isotope characteristics, and sulfate ions in water bodies, it is possible that the generation of H_2S in this area is caused by *TSR*. However, it should not be ruled out that, due to the effect of microbe assimilation reduction in the gob area of No. 8 coal seam which is the overlying adjacent layer, the H_2S generated under the role of corrupt migrated to the abnormal enrichment zone of No. 9 coal seam by the mining fissure.

3.2 Characteristics of H_2S in Xishan coal mine of Xinjiang Uygur Autonomous Region

Xishan coal mine of Xinjiang is located in the binding sites which combine the middle of the Southern Junggar Basin with the northern Tianshan. Its location map is shown in Figure 6. The main coal bearing strata of mines are the lower segments of Xishanyao formation of Jurassic system middle Triassic, and belong to coal formations in terrestrial facies. Most of the coals belong to the range of middle - ultra low sulfur. The reflectance of coal vitrinite (R_o) is in the range of 0.64% - 0.79%, so coals belong to the low rank bituminous coal. Each coal seam in this region exhibits the abnormal enrichment of H_2S. The concentration of H_2S is not distributed evenly, having distinct subdivision and zoning phenomenon. The underground water of mines is rich in H_2S. The methane gas in coal seams is mainly composed of N_2 and CH_4, accompanied with CO_2 and heavy hydrocarbon components, like C_2H_6, C_3H_8, and H_2S. In the gas component, CH_4 accounts for 2.42% - 86.5%, N_2 accounts for 15.91% - 73.68%, CO_2 accounts for 0.81% - 12.31%, H_2S accounts 0 ppm - 1500 ppm. One hour after blasting, the concentration of H_2S in the air flow reached 100ppm. At 0:00 on November 5, 2011, after blasting in the working face of the track uphill, due to the impact of the vibration, a lot of water and harmful gases suddenly rushed out from the roof of No. B_{19} coal seam. The H_2S concentration in the air flow was up to 400ppm, and the gas concentration was up to 19.5%. Since in the process of H_2S generation, heavy hydrocarbons are preferentially generated, *TSR* thereby leads to the increase of the gas dry coefficient. Methane gas of Xishan coal mine is primarily wet gas, indicating that the genetic features of H_2S by *TSR* in the coal seams are not distinct.

The changes of H_2S concentration which are monitored daily in each point of mine are shown in Figure 1.
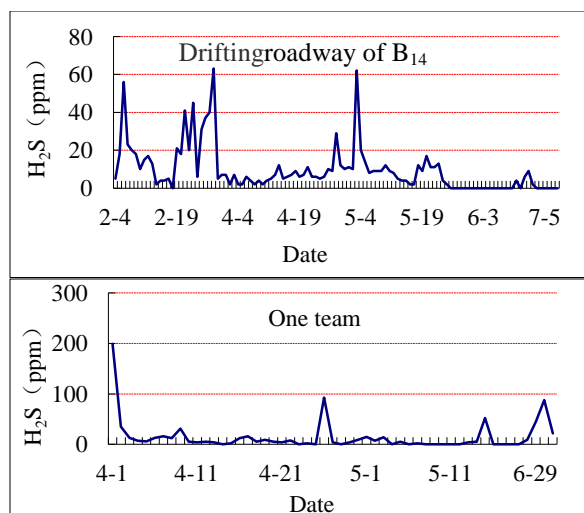
Figure 1: The monitored concentration of H_2S in the each point.

The testing of gas composition in No. B_7 coal seam by exploration holes was conducted in 2011 for Xishan coal mine. The results are shown in Figure 2.
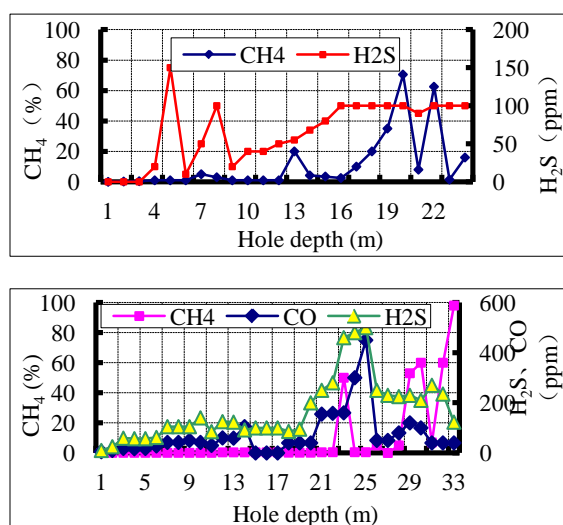

Figure 2: Gas composition of 1# and 3# exploration holes.

Figure 2 shows that, the maximum volume component of CH_4 measured by exploration holes is 98.0%. The maximum concentration of H_2S is 500ppm, and the maximum concentration of CO is 450ppm. The number of sulfate-reducing bacteria in the inspected samples of mines and adjacent mines is shown in Table 2 (Cheng et al., 2009). It is known that the *SRB* breed acts fiercely, so a large amount of H_2S will be generated by *BSR*.

Table 2: Distribution of *SRB* in the each mine of region.

Stage positions	Depth (m)	T (℃)	*SRB* (A/g sample)
Beishan coal mine-1	272.8	30	100
Xiaolongkou coal mine-1	312.0	55	3500
Beishan coal mine-2	209.5	55	650
Qitai coal mine-1	463.5	30	234
Xishan coal mine	563.0	25	235
Xishan coal mine	750.0	28	528

The values of carbon isotope $\delta^{13}C_{CO2}$ of regional CO_2 change within the range of -18‰ - -11‰ (Dai, 1985), indicating that CO_2 in the region is a product of the carbonate decomposition leaching effect as well as the organic decomposition oxidizing effect. The CO_2 represents characteristics of organic origin, and also represents the genesis features of *BSR*. The $\delta^{34}S$ of pyrite in the each coal seam of region has a distribution range of -15.3‰ - +3.7‰, further confirming that the H_2S in the each coal seam of the study area is generated by *BSR*.

4. GENESIS CHARACTERISTICS OF HYDROGEN SULFIDE IN CHINA'S COAL MINE

Due to the geological movement and various chemical reductions in China, H_2S that is generated by biodegradation under the early accumulation of peat in the oxygen-rich conditions of the early coal-forming period is impossible to preserve in the coal rock strata.

The content of H_2S in the coal rock strata is concerned with the sulfur content in coal, and has a positive correlation with the content of sulfur in pyrite. Coals of the Permian Longtan group in the Province of Guizhou, Sichuan and Chongqing are mostly high-sulfur coal, and pyrite is widely developed. The sulfur content in Guiding coal of Guizhou Province is up to a range of 9.49% - 10.46%, and more than 70% of coals in Sichuan Province are high-sulfur coal. According to the Ministry of Environmental Protection of the People's Republic of China (http://www.zhb.gov.cn), acid rain mainly focuses on the south of the Yangtze River, Sichuan, and eastern Yunnan, including the Zhejiang, Fujian, Jiangxi, Hunan, etc., and cities of the Yangtze River, the Pearl River Delta region. Figures 3 and 4 show that high-sulfur coal producing areas have characteristics consistent with the distribution of acid rain in China (Deng et al., 2013).
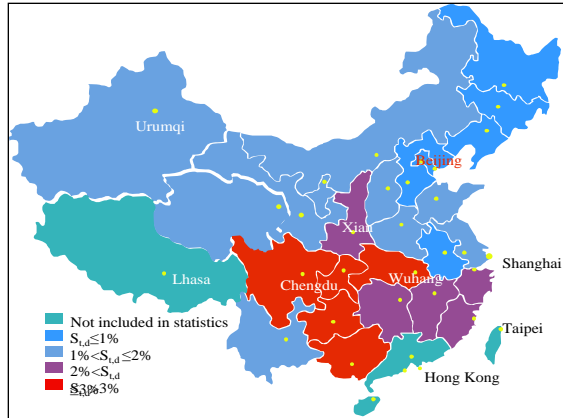
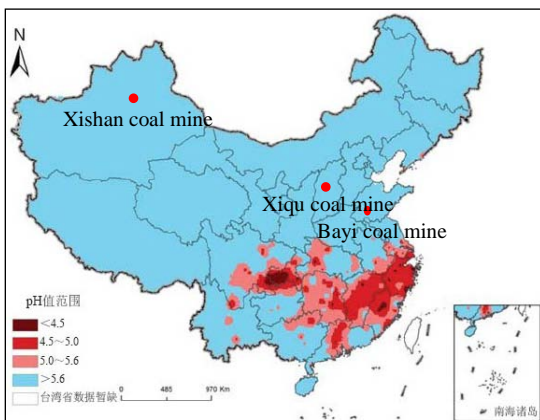Figure 3: The distribution of sulfur in coal in China.

Figure 4: The contours of the annual average pH value in China in 2014.

In the high sulfur coal of the late Permian Longtan Formation in Guizhou Province, the $\delta^{34}S$ values of pyritic sulfur and organic sulfurare in the range of -30.30‰ - 62.69‰ and -13.43‰ ~ \-10.87‰ respectively. The peak values of $\delta^{34}S$ in pyrite are in the range of -30.30‰ - -9.18‰ and 9.45‰ - 38.40‰ (Ni et al., 1999; Luo et al., 2005). These data suggest that the formation of the regional H$_2$S may experience the double genesis of *BSR* and *TSR*.

The sulfur content in coal has an increasing trend from the east to the west in China. In the three northeast provinces, coals have the lowest sulfur content and currently, coal mines in the region have not reported that H$_2$S is abnormal. The sulfur content in coal of the Carboniferous Taiyuan formation in the southwest of Shandong Province is mostly over 1%, and some are up to 5%. The maximum sulfur content in coal of the Permian Shanxi formation in the southwest of Shandong Province and in the Yongxia coal field which belongs to Henan Province is up to 3%. In the methane gas of the above regional coal

seams, sulfur isotopes have the characteristics of positive end, whose value is closer to the $\delta^{34}S$ of sulfate. It may be that under the action of thermal properties of the magmatic rock intrusion, the coal, sulfur-bearing organic matter, and sulphate rocks in the surrounding rock conduct the thermal chemical decomposition and the thermal chemical reduction to generate H$_2$S. For carboniferous coal seams in north China, the high sulfur coal is widespread. Types of coal are mostly coking coal and above. The average maximum vitrinite reflectance $R_{o,max}$ of coal from part of coal seams is over 1.2%, having the genesis conditions of *TSR*. For example, the coal rock strata of Qinshui basin in Shanxi Province may have experienced high temperatures of more than 268°, with the temperature generally being above 150° (Cheng et al., 2009). Through measurement of gas samples from the exhaust gas of the block surface well head in Fanzhuang and Panzhuang, the content of H$_2$S in the coal seam gas component is 0.0001% - 0.001%. The H$_2$S content in the desorbed gas of two coal samples from Sihe coal mine is 0.0001% and 0.0002%, respectively (Ma et al., 2008). Since H$_2$S can easily react with metal, the actual content of H$_2$S is much larger than the above data. The distribution of coal mines in which H$_2$S is caused by *BSR* is broader, such as Wuda of Inner Mongolia, Tingnan of Shaanxi Province and the vast region of Xinjiang Autonomous Region.

5. DISCUSSION AND PROSPECTS

The H$_2$S content in the coal rock strata is concerned with the sulfur content in coal, and has a positive correlation with the content of sulfur in pyrite. In the coal-forming environment of marine facies, the sulfur content in the coal rock strata is generally higher and H$_2$S is easy to generate and preserve. Due to the insufficient sulfur ion abundances and sufficient heavy metals, H$_2$S of coal formation in terrestrial facies is relatively difficult to generate, reserve and accumulate. The coal seams of Xishanyao Formation in Xinjiang Autonomous Region belong to terrestrial facies of coal. The H$_2$S in this region is abnormally accumulated and widely distributed. Therefore, conducting research on the causes and distribution of H$_2$S in coal, which belongs to terrestrial facies of coal, is particularly important.

The polarity characteristics of H$_2$S determine that the distribution (enrichment) of H$_2$S is controlled by many factors. The control action of factors in the abnormal gathering of H$_2$S in coal mines, such as, the regional structure, the nature of the reservoir cap, groundwater activity, temperature, and the burial depth of the coal seam should be considered synthetically.

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At present, research on the mechanisms causing H_2S is still low. The genesis category of H_2S in the coal mine can be judged comprehensively by geologic and tectonic evolution, thermal evolution history of coal and rock series, characteristics of carbon and sulfur isotope, groundwater chemistry, methane component characteristics, source of sulfate, enrichment state of pyrite in coal and activity characteristics of sulfate-reducing bacteria.

Coal mines that contain H_2S are widely distributed in China. With the increasing depth of coal mining, the threat that H_2S poses to miners' occupational health as well as the safe and efficient exploitation of the mine is increasingly serious. Therefore, it is important to identify the genesis and distribution characteristics of hydrogen sulfide in coal mines as soon as possible, to ensure effective prediction and safety and efficient mining.

6. ACKNOWLEDGEMENTS

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