Pollution generation, migration and in situ control measures on coal refuses

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Abstract

Coal refuse, from coal mining and coal processing, is the most important source of coal mine. Based on the analysis of the contamination and its migration, some countermeasures on the control of contamination in situ were put forward. The research showed that: the contamination of coal refuse was mainly due to the existence of a large number of pollutants, open-air dumps, oxidation, acidification and spontaneous combustion, and its pollution migration mainly through wind and water. In-situ control of coal refuse pollution is the most effective means because coal refuses piled up and the presence of large quantities of pollutants. Slurry ejection, adding some inhibiting materials to inhibit oxidation (selective bactericide, reducing bacteria, alkaline material) and surface coverage are appropriate control measures in situ. The integration of multi-layer coverage to inhibit contamination and ecological restoration could be the ultimate direction for pollution control of coal refuse piles.

Keywords: coal refuse; contamination; migration; control in situ

1. Introduction

Coal is the predominant energy resource, which accounts for 74% in energy consumption structure in China\textsuperscript{[1]}. Unfortunately a large amount of coal-mining waste are dumped and piled every year, it has contaminated the soil, water, and plant, human around coal mine\textsuperscript{[2]}. According to the survey, coal refuse usually accounts for 10-30\% of coal outputs and more than 3.5 billion tons of coal refuse has been produced. The utilization of coal refuse is less than 30\% in China, so a large number of coal refuse is still piled on the ground.

Coal refuse contains a lot of carbon, which is the source of CO\textsubscript{2} emissions. Open-air coal refuse dumps are easily weathered with the role of wind and rain, and the weathered particles are also easily eroded by wind and rain, which results in air and soil. Coal refuse containing pyrite, sulfur, coal, etc., can easily cause spontaneous combustion and the release of a large number of toxic and harmful gases, such as SO\textsubscript{2}, CO and H\textsubscript{2}S. With the rainfall leaching and flushing, heavy metals in coal refuse could be released into surface water and groundwater, resulting in heavy metal
contamination of water bodies. As the result of large slope, spontaneous combustion, rainfall leaching, the collapse, landslides or mud-rock flow and other geological disasters will happen, resulting in serious economic losses and casualties. Therefore, coal refuse pile has led to serious social problems and environmental problems, and it has become the primary issue of environment in mining areas.

2. Causes of the pollution from coal refuse piles

The pollution from coal refuse piles is mainly due to the existence of pollutants, pile on ground, oxidation and acidification, and spontaneous combustion.

2.1. Large quantities of pollutants in coal refuse

Coal refuse is from coal mining and washing process, which includes the coal-bearing rocks of roof and floor. The main mineral composition of coal refuse includes kaolinite, quartz, as well as other components such as feldspar, calcite, siderite, pyrite and illite. Coal refuse contains a large number of chemical components, such as SiO₂ accounting for 40-60%, Al₂O₃ for 15-30%, CaO for 6%, F₂O₃ for 2-8%, MgO for <3%. Coal refuse has a fixed carbon content of 10%-30% and the heat can reach 3.35×10³-6.28×10³kJ·kg⁻¹. There are so many toxic elements in coal refuse, such as lead, cadmium, mercury, Arsenic, Chromium and sulfur. Trace elements in coal gangue is generally believed that the value is higher than coal, and As, Hg, Cd, Pb, Cu, Zn concentrations much higher than the crustal abundance values. Trace elements could migrate to the surrounding environment, and its extraction leached amount is increased with the decrease of pH, and its pollution is becoming smaller with the increasing of distance. However, some studies have shown that coal refuse in Anhui, Shandong will not cause heavy metals pollution [3-4]. Some scholars have also explored the issue of organic pollutants in coal refuses through the monitoring of 16 kinds PAHs of USEPA priority pollutants. In short, the existence of a large number of pollutants in coal refuse is the underlying causes of environmental pollution [5].

2.2. The dump of coal refuses on ground

Directly piled coal refuses on ground usually changes the regional landscape. Coal refuse is mostly gray-black, it has become a marker of coal mine area. Bare and black coal refuses has resulted in the serious impact on natural landscape of the mining area. The pollutants in open-air coal refuse piles directly exposed to the wind and rain can easily migrate and pollute the surrounding soil, water bodies and groundwater. Coal refuse is easy to be weathered and oxidized, it could cause spontaneous combustion and explosion. Therefore, the exposed coal refuse piles are the main external causes of pollution.

2.3. The oxidation of sulfur in coal refuses

The oxidation is the important cause of pollution. Studies showed that the acidity of coal refuse is most serious pollution and most difficult to control [6-11]. The spontaneous combustion of coal refuse, explosions, landslides and pollution of soil and water are directly related to the acidity of coal refuse. The acidity is mainly due to sulfur [8-12], which could generate acid mine drainage (AMD) as a result of oxidation, the pH value of AMD up to between 2.0-3.5 [13]. In acidic conditions, the heavy metal activation and pollution of the activity has been exacerbated. The process of sulfide oxidation also produced a lot of heat, can easily lead to spontaneous combustion of coal refuse, pollute the environment and trigger explosions and environmental disasters. Therefore, the oxidation of coal refuse is the main incentives of acidic pollution, heavy metal activation, spontaneous combustion, and explosion.

Releases of AMD have low pH, high specific conductivity, high concentrations of iron, aluminum, and manganese, and low concentrations of toxic heavy metals. Chemical, biological and physical factors are important for determining the rate of acid generation; physical factors, particularly the permeability of coal refuse piles, are particularly important. Piles with high permeability have high oxygen ingress, which contributes to higher chemical reaction rates, hence, higher temperatures and increased oxygen ingress through convection. Most testing of Acidithiobacillus ferrooxidans (eg. Thiobacillus ferroxidans) has involved oxidation of pyrite (FeS₂); however, the bacterium may accelerate oxidation of sulfides. It could be 10⁶ times of nomal chemical reaction. Therefore,
restraining the oxidation of pyrite is the key for pollution control.

2.4. Spontaneous combustion of coal refuse

The spontaneous combustion of coal refuse arising from oxidation to a certain extent, at the same time, a large number of greenhouse gases and toxic gas could be emitted, which is the main reason of acid rain and the explosion disaster. The Oxidation of pyrite release the heat and build-up in the coal refuse, which could increase the rate of oxidation. As the open-air piles, the surface of pyrite full contacts with the air, and form the "down slope-style", which makes easy to air penetration and provide the oxygen to internal pyrite oxidation. According to statistics, the annual spontaneous combustion of coal refuse, combustion emissions to the atmosphere with CO10.8 g, SO2 6.5 g, H2S and NO2 2 g a day per square meter. Soluble sulfate and sulfuric acid not only affects the coal refuse, also result in the acidification of surrounding water and soil environment and soil acidification, so it is difficulty for vegetation to survive.

3. The analysis of pollution migration

As shown in Fig.1, the pollution of coal refuse is mainly through the role of wind and water. The role of wind is to make the surface of fine-grained waste rock directly to the vicinity of the atmosphere and soil. And the role of water, firstly through the surface runoff, pollutants and acidic water will be moved to the vicinity of the soil and water, secondly through the water infiltrate into the underground, which contaminate the groundwater.

Therefore, the degree of pollution is often associated with the distance and the main wind direction.

4. In-situ control measures for coal refuse pollution

As the coal refuse is one of solid waste pile, the existence of a large number of pollutants, which is the source of contamination. Many methods are used to remove pollutants, such as removal carbon, desulfurization, and removal heavy metals requiring ex situ processing, which needs high costs. So pollution in-situ control is one of the most effective ways.

Based on the analysis of the causes of pollution, it could be concluded that exposure, oxidation and spontaneous combustion of coal refuse are the main external causes of pollution. Therefore, in situ control of pollution of coal refuse focus on external factors, the core is appropriate cover technology.

4.1. In-situ extinguish fire of coal refuse piles

At the spontaneous combustion serious site (including a serious area of potential spontaneous combustion), the use of in situ grouting is an effective way. Their technical key is to design a reasonable location and depth of grouting, reasonable grouting pipe installation, application of a reasonable proportion of materials (lime, fly ash, and
loess). The process is as follows: 1) through the observation of surface temperature and delineation of the scope of grouting; 2) grouting drilling is still being adopted in high-temperature region at the top of the plane; 3) as for slopes can not be drilling rig project, grouting will be carried out by the means of artificial drilling or coal drilling hole.

4.2. In situ control measures to inhibit oxidation of sulfur

4.2.1. Bactericides used to inhibit catalytic oxidation

The oxidation of pyrite (FeS₂) in coal mine wastes is the source of resulting some environmental problems and disasters such as acid mine drainage (AMD), spontaneous combustion, deflagration and landslip. The oxidation of pyrite could be accelerated greatly by *thiobacillus ferroxidans* (10⁶ times of normal chemical reaction), which have a pivotal role in generating AMD and spontaneous combustion. Therefore, the bactericide technology can effectively inhibit the activity of *thiobacillus ferroxidans* and control environmental problems[14]. The mechanism of oxidation of pyrite and environmental pollution and the function of bactericide technology in inhibiting oxidation was discussed by Kleimann[15]. Some research results showed that anion surfactant, food preservative, certain organic acid were effective bactericide of *thiobacillus ferroxidans*. Bactericide technology for coal wastes treatments is a new field in China. Learning form foreign countries could promote the development of this field in China.

The author has done some researches on bactericide [16]. The research showed that sodium lauryl sulphate and is effective bactericide which can inhibit the activity of *Thiobacillus ferroxidans*. The inhibition is 80.32% with sodium lauryl sulphate concentration 30mg/L, and that of sodium benzoate is 74.36% with of the concentration 40mg/L. Bactericide technology for coal wastes treatments is a new field in China, which could control the pollution and promote the re-vegetation of coal mine wastes.

4.2.2. SRB used to inhibit the oxidation of sulfur

Sulfate-reducing bacteria (SRB) were derived from the coal refuse or the surrounding soil, and used it to control the pollution heavy metals. the research showed that SRB is the effective to control sulfate and heavy metals. the degree of sulfur control is above 90%, which could prevent the oxidation of pyrite and AMD occurrence.

4.2.3. Alkaline materials used to inhibit the oxidation of sulfur

The method is mainly by adding to the phosphate or alkaline substances to prevent or inhibit the oxidation of sulfides. After adding phosphate into coal refuse, phosphate and Fe³⁺ generated precipitation (FePO₄), reduced the activity of Fe³⁺, thereby reducing the oxidation rate of Fe³⁺ on pyrite. The alkaline substance includes NaOH, limestone, lime, Na₂CO₃, fly ash, etc [17]. Study found that an alkaline substance formed after adding the alkaline environment is not conducive to the oxidation of pyrite, as the acidic environment is the most appropriate *Thiobacillus ferroxidans*. This method is often used in combination with the organic material as the cover of coal refuse piles.

4.2.4. Surface cover of coal refuse piles

1) Sludge or fly ash cover. The oxidation of pyrite can be inhibited by organic waste materials such as compost, manures, sewage sludge peat, hay, straw, sawdust. The effective technique using an oxygen cover is the placement of organic waste materials on the mine waste. First, the bacterial activity which completes decomposition of the compost is using up oxygen, preventing its movement down through the tailings, and the changes in particle size prevent water from draining through the compost so that no source of oxygen is available. A second mechanism of inhibition may allow the sludge to complex Fe and eliminate it from oxidizing more pyrite, or adsorb/complex Al and other metal ions, thereby reducing hydrolysis and pH decreases [18].

The main advantages over other types of covers are related with their low hydraulic permeability, high cation exchange capacity and high alkalinity. In addition, the establishment of organic covers, which is considered as a low cost solution for the prevention of acid mine drainage generation.
Fly ash was often used to control the acidic and heavy metals pollution as a surface cover, because fly ash is solid waste with alkaline property and strong absorption ability. Our research showed that fly ash was very effective to control acidic and heavy metal pollution, and sludge cover and Inoculation of SRB were good method to control heavy metal and acidic pollution.

2) Multi-layer coverage. Soil cover is used as air and water barrier to prevent the erosion of coal refuse, help re-vegetation and control the pollution. However, many research and practice also found that the direct cover of soil is not a good way to control the pollution of coal refuse. such as one burning coal refuse piles in Yangquan, Shanxi province were covered with soil directly, there was still spontaneous combustion characteristics of smoke and acidification, vegetation can not survive or low survival rate. Therefore, the multi-coverage will be more effective method. We made the following multi-layer covering the structure of the soil profile reconstruction; firstly, the isolation layer including clay minerals, alkaline materials, sludge, selectively bactericide and sulfate-reducing bacteria was paved to inhibit the oxidation of sulfur and control pollutant, secondly, the middle layer was laid with clay materials to provide the environment of plant growth. Covering method should be taken into account the implementation of rolling process. Pollution control and ecological restoration was achieved by multi-coverage method.

The goal of long-term effectiveness could be achieved after the establishment of vegetation ecosystems by the multi-layer coverage method, which results in reducing surface runoff, wind erosion, dust pollution, acidification and oxidation of sulfur.

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