Study on Occurrence Characteristics and Natural Degradation of Polycyclic Aromatic Hydrocarbons in Mined-out Area of Northern Shaanxi Coal Mine

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1 Shaanxi Provincial Land Engineering Construction Group Co., Ltd., Xi'an 710075, China
2 Shaanxi Key Laboratory of Land Consolidation, Xi'an 710061, China
3 Key Laboratory of Degraded and Unused Land Consolidation Engineering, the Ministry of Natural Resources, Xi'an 710075, China
4 Shaanxi Provincial Land Consolidation Engineering Technology Research Center, Xi'an 710075, China
5 Institute of Land Engineering and Technology, Shaanxi Provincial Land Engineering Construction Group Co., Ltd., Xi'an 710021, China

Corresponding author e-mail: lunan8836@126.com

Abstract. The rapid economic development is accompanied by the rapid consumption of energy. Coal plays an important role in China's energy production. With the continuous exploitation of coal resources, the mined-out area of coal mines has continued to increase. PAHs are common organic pollutants in coal mines. With the gradual deepening of the research on PAHs, the natural degradation of indigenous dominant bacteria has become the best choice. The application of microbial treatment to repair PAHs-contaminated soil, and the research on the relationship between PAHs and degrading microorganisms are great significance to the bioremediation of PAHs pollution.

1. Formation and Development of the Goaf in North Shaanxi Coal Mine

Coal is an important basic energy source in China, and it holds an extremely important strategic position in the national economy. "Rich coal, poor oil and low gas" is a characteristic of China's energy, which fundamentally determines that for a long period of time, the dominant role of coal in China's energy production and consumption is difficult to change [1-2]. Shaanxi is a major coal mining province in China, and the northern Shaanxi mining area is currently the main coal mining area in Shaanxi [3]. Yushen Mining Area in northern Shaanxi is the essence of Jurassic coalfield in Yushenfu, and it is one of the most important coal production bases in China [4]. The northern Shaanxi coal mining area is located at the junction of the Maowusu Desert and the Loess Plateau in northern Shaanxi, and its native geology and ecological environment are extremely vulnerable. Over the years, due to reasons such as over-intensity mining and inadequate restoration and treatment, large-scale goafs have been caused, leading to a series of geological environmental problems such as surface subsidence and geological disasters in mining areas [5].
2. Typical pollutants present in coal mining areas-PAHs
Polycyclic aromatic hydrocarbons are a persistent environmental pollutant commonly found in water, air, soil and plants [6], with the characteristics of genotoxicity and carcinogenic potential [7]. Polycyclic aromatic hydrocarbons (PAHs) constitute a class of harmful organic chemicals, including three or more linear fused rings arranged in corners and clusters, which are mainly generated from the incomplete combustion process of organic substances (such as coal, petroleum, etc.). Polycyclic aromatic hydrocarbons exist in the ambient air in the gaseous phase and act as adsorbents for aerosols [8]. After atmospheric chemical processes, PAHs can also be converted into more toxic derivatives. The main source of PAH in soil and water is atmospheric deposition. Polycyclic aromatic hydrocarbons are highly fat-soluble, easily absorbed from mammals' intestines, and rapidly distributed in a variety of cellular tissues, causing great damage to human health and the ecological environment [9]. Many studies have shown that PAHs are polluted to varying degrees in surface soils, coal gangue yards, surface water, mine water, and other sediments in coal mines, and are mainly derived from coal combustion and coal residues. And the proportion of middle molecular weight aromatics above four rings is higher [10].

3. Study on Natural Degradation of PAHs

3.1. Related Bacterial Research
The hydrophobicity and lipophilicity of PAH lead to its easy accumulation and accumulation in soil. The soil environment eventually becomes an important carrier of PAH and has strong migration. Bioremediation has attracted the attention of researchers due to its safety and economy in removing environmental pollutants such as PAH. Studies by Adel-Shafy and Mansour [8] show that some Gram-positive and negative Bacteria, fungi and algae that can break down PAH have been isolated and identified. Cerniglia [7] detailedly explained the biochemical principles and catabolic pathways of microbial degradation of PAH. The relationship between the chemical structure of PAH in the ecosystem and the rate of biodegradation is summarized, indicating that these conditions are critical to optimize the bioremediation process. Kuppusamy et al. [11] summarized the repair methods including bio-enhanced technology and bio-stimulation technology for the treatment of PAH-contaminated soil. The combination of physical and chemical technology and biotechnology has also been widely used, but biocatalytic assisted repair technology is still in the development stage. Davie-Martin [12] pointed out that the use of bioremediation not only can degrade PAH into less toxic compounds, but also can perform in situ repair, effectively reducing engineering costs. Compost treatment can reduce the PAH in the soil by about 70%, but different environmental conditions have different effects on the results. Alegbeleye et al. [13] concluded that both short-term and long-term PAH pollution will affect soil microbial community structure, destroy microbial diversity and species uniformity, and soil properties significantly affect microbial enzyme activity. The microorganisms capable of degrading PAH include bacteria, fungi, algae, etc., different environmental conditions such as soil pH, temperature and humidity, redox potential, and salinity. The content, structure, and microbial composition and activity of soil PAHs have a significant effect on the degradation rate of PAHs. In the natural state, microorganisms, as decomposers, mainly use their own enzymes to catalyze PAHs into small molecule organics. Due to the difference in electron acceptors, they eventually decompose them into CO₂, H₂O, and CH₄ etc. [14]. For example, aerobic bacteria epoxidize benzene to form dihydriodiol epoxide under the action of dioxygenase, form catechol through dehydrogenase, and further metabolize to form carbon dioxide and water [15]. Numerous studies have isolated microorganisms, such as bacteria, fungi, and algae, that can transform, degrade, and use PAHs as carbon and energy sources. Pseudomonas, Micrococcus, Rhodococcus, Arthrobacter, Bacillus, Corynebacterium can effectively degrade PAHs.

3.2. Study on natural degradation conditions
Under anoxic or anaerobic conditions, anaerobic bacteria use, for example, sulfate, nitrate, iron,
manganese, and carbon dioxide as electron acceptors to eventually generate carbon dioxide and methane. Some anaerobic bacteria use organic pollutants as electron acceptors. Studies have found that fungi, especially white rot fungi such as Pleurotus spp., Tobacoo and Pleurotus, are all aerobic bacteria capable of degrading PAHs [16]. Mycobacteria can mineralize PAHs such as fluoranthracene, pyrene and benzo (a) pyrene. Bacillus, Pseudomonas and mixed cultures of Pseudomonas and Flavobacterium can effectively degrade B(a)P. Anthracene can be completely mineralized by Bacillus protoplasts API S 272, Bacillus subtilis SBS 526, Bacillus licheniformis, Klebsiella, Bacillus, Nocardia, etc. Jin et al. [17] screened a Pseudomonas fluorescens from farmland soil. After 30 days of culture, the degradation rates of phenanthrene, anthracene, and benzophenanthrene in contaminated soil were all higher than 50%. Liu et al. [18] combined PAHs degradation strain Rhizobium petrolearium SL-1 with phytoremediation of contaminated soil, and the degradation effect was significant.

Some research results show that the optimal pH for soil PAHs degradation is neutral, but due to the nature of the contaminated soil and the specificity of the contaminated site. Temperature determines the degree and rate of degradation of PAHs by microorganisms in the natural environment because the temperature directly affects the chemical composition, physiological characteristics and diversity of PAH compounds [19]. In addition, temperature has significant constraints on adsorption equilibrium and kinetics. And high salt content will dehydrate microbial cells and denature enzymes, which will reduce the metabolic rate of microorganisms [20]. Other scholars have proposed technologies such as mixed cell culture, biosurfactant washing, transgenic methods, and nano-remediation to overcome the limitations of existing soil pollutants and microbial-related technologies in processing high molecular weight PAH, and ultimately ensure the long-term PAH-contaminated soil Repair [11].

4. Scientific issues and significance of this project

Scholars at home and abroad have studied the source, distribution, migration and transformation of PAHs in the environment of coal mining areas. At the same time, some scholars have done preliminary research on the screening, classification and optimization of some environmental conditions of PAHs-contaminated soil microorganisms. The following two aspects still need to be further improved:

1) With the formulation and improvement of relevant standards for water and soil restoration by the state, the management and control of polluted soil and water in typical areas is gradually being carried out. However, for the mined-out area of coal mines, the research focused on the stability of soil structure, environmental problems such as desertification caused by pollution of water resources and changes in water levels. Not enough attention has been paid to the problem of organic pollution in soil, especially the study of the occurrence, migration, transformation and degradation of PAHs pollutants that are widely present in coal mining areas. Therefore, it is urgent to carry out related research.

2) The biodegradation of PAHs in coal mines at home and abroad is mainly concentrated in water bodies (surface water, mine water, shallow groundwater, etc.). Due to the variety of soil types, different properties, and many influencing factors, the impact on the final concentration of pollutants. There were few studies on soil ecosystems, especially for the complex geological environment of the mined-out area of coal mines. The extraction of dominant microorganisms, optimization of degradation conditions, and the mechanism of degradation and conversion of PAHs need to be further studied.

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