Removal of Solid Impurities from Coal Gasification Wastewater by Sand Filtration

Li Dai¹ and Hualin Wang^{1,*}

¹National Engineering Laboratory for Industrial Wastewater Treatment, East China University of Science and Technology, Shanghai, 200237, PR China

Abstract. Coal gasification wastewater has complex pollutant components and high COD value, and contains a variety of toxic and harmful substances. The treatment of coal gasification wastewater has always been one of the important problems in the development of coal gasification industry in China. In this paper, aiming at the removal of fine particle pollution in coal gasification wastewater, the sand filtration method was used to explore the changes of parameters such as solid content removal, separation accuracy, grade efficiency under the conditions of separating different bed thicknesses. With this separation method, the fine particle pollutants in coal gasification wastewater can be effectively removed by more than 95%, the separation accuracy can reach 0.46 μm . The operation conditions are optimized for further industrial application.

1 Introduction

China's fossil energy structure is more coal and less oil. As the main mineral energy, coal has an important impact on China's industrial economic development[1]. In recent years, the rise of coal chemical industry has further improved the utilization value of coal and reduced China's dependence on petroleum resources. However, with the increase of environmental protection requirements, wastewater from coal gasification has gradually become a bottleneck restricting the development of coal chemical industry[1,2].

Coal gasification wastewater has complex pollutant components and high COD value, and contains a variety of toxic and harmful substances[1,3]. Coal gasification wastewater poses a threat to human health. If these wastewater are not treated effectively, it will lead to crop death and water pollution. At the same time, because some substances in coal gasification wastewater are difficult to be removed by biological means[4], it is environmentally persistent and will cause long-term harm to the ecological environment once discharged[1]. The treatment of coal gasification wastewater has always been one of the important problems in the development of coal gasification industry in China. For coal gasification wastewater treatment, domestic and foreign researchers have carried out a lot of research work[5].

At present, the common remediation methods to deal with coal gasification wastewater contain physical, chemical and biological processes, namely adsorption by activated carbon[6] or clays[7], solvent extraction[8], liquid membrane[9], coagulation[10], bubble[11] wet air oxidation[12], ozonation[13], photocatalytic oxidation[14] and decomposition by Fenton reagents [5,15]. In order to

deal with this kind of complex coal gasification wastewater with strong toxicity and low biodegradability, it often needs to adopt a variety of technologies for combined treatment[16]. Although effective, some of these techniques present a number of disadvantages[5]. Nowadays, the more feasible treatment process is to remove solid and oil, recover phenol and ammonia, biochemical treatment, advanced treatment, membrane treatment[17], evaporation[18] and crystallization[19], etc. However, before phenol ammonia recovery, coal gasification wastewater needs to be pretreated with solid removal. Now that separation process during industrial production accounts for 10-15% of the global energy consumption[20], it's urgent to develop a kind of separation method and process with high efficiency, high accuracy and low energy consumption for the removal of solid impurities in coal gasification wastewater.

Deep bed filtration is usually employed to remove fine and colloidal particulates from liquid suspensions[21]. When a diluted suspension flows through a filtering granular bed, the particles to be removed approach the filter grains (collectors) by various transport mechanisms (direct interception, inertia, sedimentation, molecular diffusion, electrostatic forces, hydrodynamic action) and they can further adhere to the collector surface (by van der Waals forces, electric double layer interactions, hydrogen bonding, mutual adsorption of polymeric species)[22]. Sand[23], anthracite coal, granular activated carbon[6], garnet, natural zeolites, synthetic inorganic sorbents, ion exchange resins, glass and expanded polystyrene beads, are common granular materials used to remove particles from a diluted suspension by deep bed filtration, and

* Corresponding author: wanghl@ecust.edu.cn

rapid sand filtration is widely applied to water treatment[24].

In this study, a sand filtration method based on irregular medium was proposed, and a small test by the method was carried out to separate particle impurities from coal gasification wastewater. Through the indexes of solid content, separation efficiency, separation accuracy and classification efficiency in coal gasification wastewater, the solid content and separation efficiency of quartz sand bed under different bed thicknesses were investigated, and the feasibility of removing solid impurities in coal gasification wastewater by this separation method was evaluated. At the same time, using this method, the separation accuracy of solid impurities in coal gasification wastewater can reach 0.46 μm , which further verifies the feasibility of the separation method. The operating conditions of industrial application of this method were also optimized.

2 Material and methods

2.1. Experimental equipment and setup

The separator adopted in the experiment was made of Organic glass. The dimension of the separator was as shown in Fig. 1 and Table 1, i.e. the inner diameter D of the main part was 70 mm, the length H of the cylinder was 12000 mm, the head height h_1 was 50 mm, the pipe dimension d of the top of the head was 10 mm, and the filling height L of the separator was different. The principal of the method is to use the bed piled up by quartz sand filtration medium (1.5 mm) to form a deep bed filtration.

The experimental process was shown in Figure 1. First, the coal gasification wastewater to be treated was poured into the sink, then the pump was started and the flow rate was adjusted. The coal gasification wastewater was separated by the separation bed, and after it runs stably for 2 minutes, it was sampled and marked at the outlet sampling port. In this experiment, the removal of fine solid impurities in coal gasification wastewater by separation bed was mainly investigated, and the feasibility of the separation process was also investigated. In the bed filtration process, particles were removed during the flow of asuspension through a granular bed (the top of bed, the middle of bed, the bottom of bed). It followed that if particles followed the fluid streamlines; many of them would never touched a grain surface and been removed from the flow. However, transport moved the particles across the streamlines, in such a way that they reached the immediate vicinity of a grain surface. When they arrived there, attachment can occur depended on the nature of the surface-particle interactions.

During the experiment, the flow velocity of coal gasification wastewater was adjusted 10 m/h during the whole experiment process to match a rational ratio of industrial production and the test. After inlet properties of the experimental system held stable for over two minutes, Meanwhile, the effluent samples were taken, and was recorded respectively.

Fig. 1. Flow chart and principle diagram of coal gasification wastewater separation. (1) schematic diagram at top of bed; (2) schematic diagram at middle bed; (3) schematic diagram at bed bottom

Table 1. Structure parameters.

D (mm)	H (mm)	L (m)	h_1 (mm)	d (mm)
70	12000	1.9、3.8、4.75、 5.7、7.6、9.5、11.4	50	10

2.2 Experimental material

The coal gasification wastewater was produced by a coal chemical plant in Yunnan, China. The pH of the coal gasification wastewater was 9.2-9.4 and COD was about 34,000 mg/L.

The particle size test of solid impurities in the coal gasification wastewater was shown in Figure 2 (a) and Fig. 2 (b). The $D_v(50)$ particle size of the solid impurities in the coal gasification wastewater was 7.98 μm and the specific surface area was 1393 m^2/kg , of which 99% was in the waste liquid Fig. 2(c) was a scanning electron microscope of solid particles, from which it can be seen that the solid particles in the coal gasification wastewater were fine particles of different sizes, and the surface of the fine particles was rough, as shown in fig. 2(d), and there were more particles about one micron.

Fig. 2. Particle size distribution and SEM of solid particles in coal gasification wastewater. (a)(b) Particle size distribution ;(c) (d) Scanning electron micrograph

2.3 Experimental procedure and test methods

The particle size distribution was detected by British laser particle size analyzer Mastersizer 3000 (The measuring range is 0.01~3500 μm , the accuracy is better than 1% and the accuracy is better than 5%). The solid particles in coal gasification wastewater were photographed by a high-resolution scanning electron microscope 250. The solid content of impurities in coal gasification wastewater was tested by the national standard drying method.

2.4 Statistical analysis

In the current work, the solid content (N) was calculated as follows:

$$N = \frac{(W_1 - W_0)}{V} \quad (1)$$

where W_1 is the weight of filter membrane, weighing dish and solid particles after drying, mg; W_0 is the weight of filter membrane and weighing dish after drying; V is the volume of filtered coal gasification wastewater sample.

The separation efficiency (η) was calculated as follows:

$$\eta = \frac{(N_{in} - N_{out})}{N_{in}} \times 100\% \quad (2)$$

where N_{in} is the solid content of particles in the inlet coal gasification wastewater, mg/L; N_{out} is the solid content of particles in the sample coal gasification wastewater after separation, mg/L.

The solid content at one particle size (N_i) was calculated as follows:

$$N_i = N_{out} \times f_i \quad (3)$$

where N_{out} is the solid content of particles in the sample coal gasification wastewater after separation, mg/L; f_i is the ratio of the particle size (i) distribution in the outlet coal gasification wastewater sample, %.

The grading efficiency (η_i) was calculated as follows:

$$\eta_i = \frac{(N_0 - N_i)}{N_0} \times 100\% \quad (4)$$

where N_0 is the solid content of particles in the inlet coal gasification wastewater, mg/L; N_i is the solid content of at this particle in the sample liquid after separation, mg/L.

3 Results and discussion

As can be seen from Figure 3, the coal gasification wastewater was treated by sand filtration. With the increase of sand filtration separation bed, the solid content in the separated coal gasification wastewater gradually decreases, and its corresponding separation efficiency also shown an increasing trend[24]. When the inlet solid content was 7783 mg/L, the bed was 1.9 m, the

solid content in the outlet coal gasification wastewater decreases to 4247 mg/L, the separation efficiency was 45%. When the separation bed was 3.8 m, the solid content in the outlet wastewater was 3705 mg/L, the separation efficiency was 52%; When the separation bed was 4.75 m, the solid content in the outlet wastewater was 2547 mg/L, and the separation efficiency was 67%; When the separation bed was 5.7 m, the solid content in outlet wastewater was 832 mg/L, the separation efficiency was 89%. With the increase of separation bed, the solid content at outlet decreased greatly, the separation efficiency increased greatly. When the separation bed was 7.6 m, the outlet solid content was 714 mg/L, the separation efficiency was over 90%. As the separation bed continued to increase to 11.4 m, the outlet solid content decreased to 341 mg/L, the separation efficiency was over 95%.

It can be seen that when the separation bed increased, the removal of solid content in coal gasification wastewater increased obviously[21], but the increase was larger within 5.7 m. The relative increase of separation efficiency was not as great as that of the former when it was 5.7-11.4 m. At the same time, it was proved that the increase of bed has a relatively suitable value.

Fig. 3. The solid content and separation efficiency curve vary with the different beds.

The solid content curves of coal gasification wastewater samples at the inlet and after separation under different particle sizes were shown in Figure 4. It can be concluded that with the increase of separation bed, the solid content of coal gasification wastewater samples at different particle sizes tended to decrease[22], the solid content of particles larger than 100 μm and smaller than 1 μm was closed to zero.

When the separation bed was 1.9 m, the particles below 0.76 μm were completely removed, and with the increase of separation bed, the particles below 0.87 μm were completely removed. Particles larger than 100 μm were also completely removed. When the separation beds were 1.9 m, 3.8 m, 4.75 m, 5.7 m, 7.6 m, 9.5 m and 11.4 m, the solid content of 0.46 μm ~1 μm decreased from 444 mg/L to 86 mg/L, 82 mg/L, 37 mg/L, 48 mg/L and

30 mg/L. The solid content of 1 μm ~10 μm decreased from 4273 mg/L to below 2992 mg/L, and the minimum was 1886 mg/L. The solid content of 10 μm ~40 μm decreased from 2148 mg/L to below 1716 mg/L, and the minimum was 1055 mg/L. The solid content of 40 μm ~100 μm decreased from 364 mg/L to below 281 mg/L, and the minimum was 4.6 mg/L. Particles larger than 100 μm were completely removed, and the solid content decreased from 553 mg/L to 0 mg/L.

Fig. 4. The content corresponding to different particle sizes in coal gasification wastewater after separation of different beds.

The graded separation efficiency as shown in Figure 5, the separation bed height increases, and the graded efficiency increases. For the particles with particle size less than 1 μm and larger than 100 μm , the curve changes of the graded efficiency in the separation bed tend to be consistent. The graded efficiency was more than 99% and tended to 100%. When the particle size distribution was from 1 μm to 100 μm , the graded efficiency gradually increased with the increase of bed thickness. When the separation bed was 1.9 m, and the particle size was 1.5 μm , the graded efficiency was lower than 5%. The separation bed was increased to 3.8 m, the graded efficiency increased from 20% to 32% at the particle size was 10 μm ; The separation bed was 4.75 m, the graded efficiency of particle size distribution from 1 μm to 100 μm was greater than 50%; When the bed was 5.7 m, the grade efficiency of 20 μm ~70 μm was larger than 65% and less than 85%. The grade efficiency of 1 μm ~20 μm and 70 μm ~100 μm was larger than 80%, then less than 90%. When the separation bed was 7.6 m, the grade efficiency of 1 μm ~100 μm was greater than 85% and less than 95%; When the separation bed was 9.5 m, the grade efficiency of 1 μm ~100 μm was greater than 90%, and the maximum grade efficiency was 100%; When the separation bed was 11.4 m, the grade efficiency of 1 μm ~100 μm was more than 92%, and the maximum grade efficiency was 100%.

The grade efficiency of particles smaller than 1 μm was more than 99%. According to the separation principle, the main separation mechanism for particles smaller than 1 μm was diffusion[25]. However, small particles were more easy to adhere to the surface of the

separation medium and deposit due to their electrostatic adsorption[24,26]. For the larger particle size larger than 100 μm , the removal was mainly due to the blocking[22] action of the separation medium. With the separation bed, the path of coal gasification wastewater passing through the separation bed increased, and the possibility of being blocked by the bed also increased[27]. Therefore, with the increase of separation bed, the grade efficiency of particle level tends to increase.

Fig. 5. The graded efficiency curve of solid particles in coal gasification wastewater by different beds.

4 Conclusion

In the present study, inspired by the mechanism of deep filtration and the removal of particulate pollutants from coal gasification wastewater, this study explored the feasibility experiment of removing insoluble particulate impurities from coal gasification wastewater by deep filtration with irregular quartz sand media. The removal performance of granular impurities in coal gasification wastewater by separation beds with different thicknesses was studied. The operation conditions were optimized for further industrial application and was of great significance for industrial application. Results suggested that:

With the sand filtration separation method, the fine particle pollutants in coal gasification wastewater can be effectively removed by more than 95%, and the separation accuracy can reach 0.46 μm .

The particles below 0.76 μm were completely removed, the solid content from 260 mg/L to 0 mg/L. The separation efficiency was 100% and the grade separation efficiency also was 100%; The solid content of larger than 100 μm decreased from 553 mg/L to 0 mg/L. the separation efficiency was 100% and the grade separation efficiency also was 100%.

The solid content of 0.46 μm ~1 μm decreased from 444 mg/L to 30 mg/L. The separation efficiency was greater than 80%, the grade separation efficiency also was greater than 39%; The solid content of 1 μm ~10 μm decreased from 4273 mg/L to below 2992 mg/L, the separation efficiency was greater than 30%, the grade

separation efficiency also was greater than 10%; The solid content of 10 μm ~40 μm decreased from 2148 mg/L to below 1716 mg/L. the separation efficiency was greater than 23%, the grade separation efficiency also was greater than 8%; The solid content of 40 μm ~100 μm decreased from 364 mg/L to below 281 mg/L. the separation efficiency was greater than 22%, the grade separation efficiency also was greater than 14%.

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