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Original Research Paper

Research on Removing Hydrogen Sulphide Based on Different Types of Activated Carbon

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ABSTRACT

The removal of hydrogen sulphide by using the activated carbon method is introduced in this paper. The desulphurization efficiency of desulphurizer is researched for different kinds of activated carbon under laboratory conditions. It focuses on the research of the removal efficiency of hydrogen sulphide by granular activated carbon, renewable granular activated carbon, granular activated carbon containing manganese dioxide and powdered activated carbon. The best type of activated carbon removing hydrogen sulphide has been obtained on this basis. The results of the studies show that the processing efficiency of granular activated carbon is far better than that of regeneration. The more the number of regeneration, the worse the desulphurization efficiency. The best activated carbon of desulphurization efficiency is the powdered activated carbon in the experiment. After the addition of manganese dioxide in granular activated carbon, the desulphurization efficiency could not be improved. It is clear that manganese dioxide does not play any role.

INTRODUCTION

Hydrogen sulphide is a kind of colourless, highly toxic, acidic gas. It has a strong odour of rotten eggs and a very small amount of hydrogen sulphide may bring harm to people. In humid conditions, it also corrodes concrete and steel (Shuang Zhang 2013). Many industrial productions are accompanied by hydrogen sulphide emissions, such as oil refining, food handling, coking plant, chemical fibre plant, paper mill, effluent disposal and so on (Lei Yan 2012). Besides, sulphur dioxide is produced after the combustion of hydrogen sulphide. It will cause serious pollution to the atmospheric environment. For that reason, concentration is limited strictly in China and some foreign countries on hydrogen sulphide in different environments (Bingnan Ren & Ping Ning 2009), and it must be conducted desulphidation before chemical tail gas emission. So whether it is from the environment or the production. Hydrogen sulphide must be removed from the exhaust gas (Zhaohua Li 2002). At present, there are many methods for disposing hydrogen sulphide gas at home and abroad. They are divided into dry and wet methods. The method of wet desulphurization is a mature method. However, its disadvantages are obvious, such as large desulphurization equipment, low efficiency, high cost and desulphurization load, the existence of mass transfer resistance, problems of sulphur recovery, sulphur blockage in tower, etc. By contrast, advantages of the dry desulphurization technology are simple process and equipment, convenient operation, low cost, small load, high degree of purification for inorganic and organic sulphur (Wei Xie 2006). There are many frequently-used methods of dry desulphurization, including ferric oxide method, zinc oxide method, activated carbon method and so on (Jihong Zhou 2006). This paper focuses on the research of removal efficiency for removing hydrogen sulphide by using granular activated carbon, renewable granular activated carbon, granular activated carbon containing manganese dioxide and powdered activated carbon.

MATERIALS AND METHODS

The hydrogen sulphide flow chart used in the study is shown in Fig. 1.

The activated carbon method: The activated carbon has been commonly used as a solid desulphurizing agent and the desulphurization process is generally in the range of 5~60°C (Jiazhong Zhang & Ping Ning 2004). Compared with other industrial desulphurization agents, it is widely used, especially for treating gas contain low concentration hydrogen sulphide (Xueqian Wang & Ping Ning 2001). This is because there are many advantages for the activated carbon, including simple operation, high sulphur capacity and desulphurization efficiency, etc. Activated carbon desulphurization relies mainly on the active groups on the surface of the activated carbon to catalyse the reaction of sulphide and oxygen (Jinyi Sun & Xiping Lin 2002). The chemical reaction is as follows:

$$2H_2S + O_2 \rightarrow 2S + 2H_2O$$

There are many different kinds of desulphurization

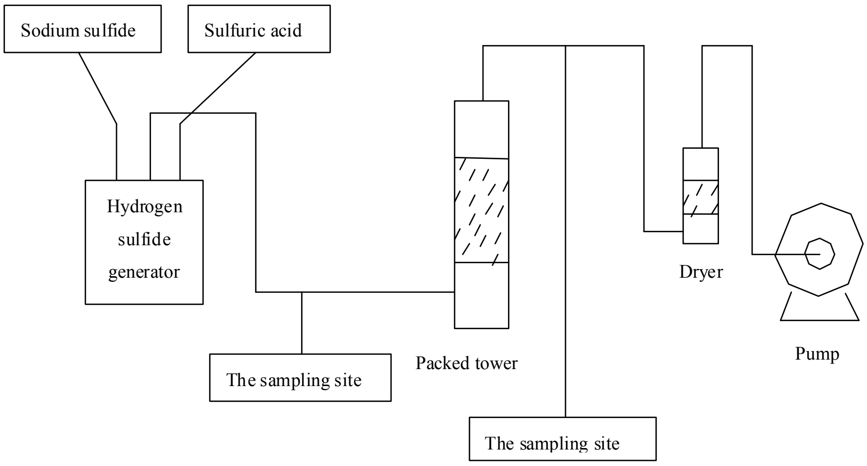

Fig. 1: Hydrogen sulphide absorption flow chart.

mechanisms discussion about activated carbon method and they are different from each other. After a summary of the scholars (Yonghou Xiao 2006), the rational process is the solution and adsorption reaction mechanism. The mechanism has four steps. Firstly, the moisture adsorbed by the activated carbon in the air forms a water film on the surface. Secondly, H_2S and O_2 diffuse into the pores of activated carbon and dissolve in the water film, then H_2S dissociation produces ions HS^-. Thirdly, the adsorption and activation of O_2 conduct under the effect of activated carbon. Besides, O_2 reacts with HS^-. Lastly, the active oxygen generated by the O-O bond rupture also reacts with HS^-, the reaction product S deposits gradually in holes of activated carbon.

The absorption of hydrogen sulphide: The 100mL zinc acetate is added to the four absorption bottles respectively. The tubualted bottle with sodium sulphide and sulphate is opened. Then the absorption bottle is connected to the proper position in the gas path. When the reaction is stable, gas flow is regulated to enter the bottle. We stop to aerate and take the absorption bottle out after turning the contents milky white in the bottle. At a certain time interval, we put up other absorption bottles, and then record flow meter readings, temperature and atmospheric pressure.

The determination of hydrogen sulphide concentration: Hydrogen sulphide is absorbed even by zinc acetate to form zinc sulphide, which reacts with iodine. Excess iodine is titrated with sodium thiosulphate. The reaction is as follows.

$$H_2S + Zn(CH_3COO)_2 = ZnS + 2CH_3COOH$$

$$ZnS + I_2 + 2HCl = ZnCl_2 + 2HI + S$$

$$I_2 + 2Na_2S_2O_3 = Na_2S_4O_6 + 2NaI$$

The calculation formula of the hydrogen sulphide content is:

$$H_2S\ (mg/m^3) = (V_1\text{-}V_2) * C * 17.04 * 1000 / (V_0 * f) \quad ...(1)$$

In this formula, V_2 is the volume of standard solution of sodium thiosulphate needed for titration in the blank experiment. V_1 is the volume of standard solution of sodium thiosulphate in the experiment. C is the concentration of standard solution of sodium thiosulphate. The mmol weight of hydrogen sulphide is 17.04. f is the coefficient, which the sample gas volume convert into standard volume and f = (273/273 + temperature°C)*(pressure mm Hg*0.1333/760*0.1333).

THE RESULTS AND ANALYSIS

The comparison of desulphurization efficiency between granular activated carbon and renewable granular activated carbon: Through the above experimental devices, we used granular activated carbon, and granular activated carbon of the first time and second time regeneration for removing hydrogen sulphide. The desulphurization efficiency is given in Tables 1, 2 and 3 respectively.

According to the desulphurization efficiency for the above three types of activated carbon, we get the curve chart of desulphurization efficiency with time change respectively (Fig. 2).

The gas flow of the outlet 2 is much higher than 300 mL/min. But the efficiency calculated in accordance with the 300 mL/min was lower than the actual. By the data in the Table 3, it can be inferred that the concentration of hydrogen sulphide does not reach the absorption saturation in the outlet 2, and the efficiency calculated is still in the rising phase of the desulphurization efficiency curve.

As can be seen from Fig. 2, although the efficiency is

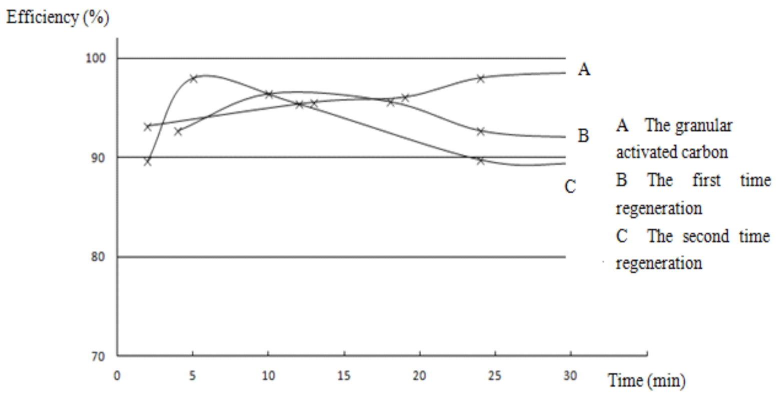

Fig. 2: The comparison curve chart of desulphurization efficiency with time change.

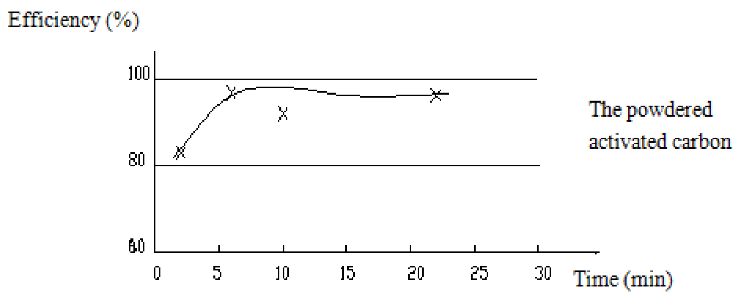

Fig. 3: The desulphurization efficiency of the powdered activated carbon with time change.

similar for the same outlet in the three groups, the saturation time of granular activated carbon is much higher than the other two groups. And with the increase of the number of regenerations, the saturation time is shorter, but the ability for removing hydrogen sulphide is weaker. Therefore, the best type for removing hydrogen sulphide is granular activated carbon in the three groups.

The comparison of desulphurization efficiency between granular activated carbon containing manganese dioxide and powdered activated carbon: ***The desulphurization efficiency of powdered activated carbon***: The desulphurization efficiency of powdered activated carbon for removing hydrogen sulphide is given in Table 4.

According to the above desulphurization efficiency of powdered activated carbon, we get the curve chart of desulphurization efficiency with time change as shown in Fig. 3.

The gas flow of the outlet 3 is not accurate. If the gas flow is 300 mL/min, the desulphurization efficiency is 92.12%. But the data are much higher than 300 mL/min, so the desulphurization efficiency is more than 92.12%. As can be seen from Fig. 3 and Table 4, we can infer that the concentration of hydrogen sulphide does not reach the absorption saturation in the outlet 3, the efficiency calculated is still in the rising phase of the desulphurization efficiency curve. Gradually the efficiency curve begins to decline steadily after this time.

The desulphurization efficiency of granular activated carbon containing manganese dioxide: The desulphurization efficiency of the granular activated carbon containing manganese dioxide for removing hydrogen sulphide is given in Table 5.

According to the above desulphurization efficiency of granular activated carbon containing manganese dioxide, we get the curve chart of desulphurization efficiency with time change as shown in Fig. 4. As per the above two types of activated carbon desulphurization efficiency, we get the curve chart of desulphurization efficiency with time change respectively as shown in Fig. 5.

As can be seen from Fig. 5, the processing efficiency of

Table 1: The desulphurization efficiency of granular activated carbon.

Number	Sample time (min)	Gas flow (mL/min)	Sample volume (mL)	Concentration (mg/m^3)	Efficiency (%)	The time interval (min)
The inlet	0.5	300	150	6216.31	0	0
The outlet 1	1	300	300	426.6	93.14	2
The outlet 2	1	300	300	1706.44	72.55	13
The outlet 3	1	300	300	243.78	96.08	19
The outlet 4	1	300	300	121.89	98.04	24

Note: (1) The temperature is 23.5°C and the atmospheric pressure is 102.6Kpa, so f=0.932. (f=19 in the blank experiment).
(2) The gas flow of the outlet 2 is much larger than 300 mL/min. But the efficiency calculated in accordance with the 300 mL/min in Table 1, so it is lower than the actual.

Table 2: The desulphurization efficiency of the first time regeneration.

Number	Sample time (min)	Gas flow (mL/min)	Sample volume (mL)	Concentration (mg/m^3)	Efficiency (%)	The time interval (min)
The inlet	1	300	300	1996.17	0	0
The outlet 1	1	300	300	146.26	92.67	4
The outlet 2	1	300	300	72.59	96.36	10
The outlet 3	1	300	600	90.73	95.45	18
The outlet 4	1	300	300	848.35	57.5	24

Note: The temperature is 21.5°C and the atmospheric pressure is 102.6Kpa, so f=0.939. (f=19.9 in the blank experiment).

Table 3: The desulphurization efficiency of the second time regeneration.

Number	Sample time (min)	Gas flow (mL/min)	Sample volume (mL)	Concentration (mg/m^3)	Efficiency (%)	The time interval (min)
The inlet	0.5	300	150	6216.31	0	0
The outlet 1	1	300	300	546.74	89.66	2
The outlet 2	1	300	300	121.5	98.04	5
The outlet 3	1	300	300	242.99	95.4	12
The outlet 4	1	300	300	546.74	89.66	24

Note: The temperature is 22.5°C and the atmospheric pressure is 102.6Kpa, so f=0.935. (f=18.2 in the blank experiment).

Table 4: The desulphurization efficiency of powdered activated carbon.

Number	Sample time (min)	Gas flow (mL/min)	Sample volume (mL)	Concentration (mg/m^3)	Efficiency (%)	The time interval (min)
The inlet	0.42	300	125	10823.69	0	0
The outlet 1	1	300	300	1828.33	83.11	2
The outlet 2	1	300	300	89.68	96.87	6
The outlet 3	1.5	300	450	853.22	92.12	10
The outlet 4	0.42	300	125	356.25	96.71	23

Note: (1) The gas flow of the outlet 3 is much larger than 300mL/min. The data are not accurate in this group.
(2) The temperature is 23.5°C and the atmospheric pressure is 102.6Kpa, so f=0.932. (f=19 in the blank experiment).

Table 5: The desulphurization efficiency of granular activated carbon containing manganese dioxide.

Number	Sample time (min)	Gas flow (mL/min)	Sample volume (mL)	Concentration (mg/m^3)	Efficiency (%)	The time interval (min)
The inlet	0.42	300	125	1149.31	0	0
The outlet 1	1	300	300	272.2	76.32	4
The outlet 2	1	300	300	146.26	87.27	10
The outlet 3	1.5	300	450	241.96	78.95	18
The outlet 4	0.42	300	125	302.45	73.68	24

Note: The temperature is 21.5°C and the atmospheric pressure is 102.6Kpa, so f=0.939. (f=19.9 in the blank experiment).

powdered activated carbon is far better than that of granular activated carbon containing manganese dioxide. This is because the advantages of the powdered activated carbon are beneficial to full contact for hydrogen sulphide gas and activated carbon, thereby the amount of removal and the desulphurization efficiency can be improved. The benefits include small particles and spacing, high resistance and for a long time the hydrogen sulphide passes through the powdered activated carbon.

The comparison of desulphurization efficiency between granular activated carbon and powdered activated carbon: According to the above analysis, we get the comparison curve chart of desulphurization efficiency between granular activated carbon and powdered activated carbon with time change as shown in Fig. 6.

Fig. 6 shows that the efficiency is similar for them, but the deviation has occurred in both charts, such as the efficiency of the outlet 3 in the circle and the outlet 2 in the box.

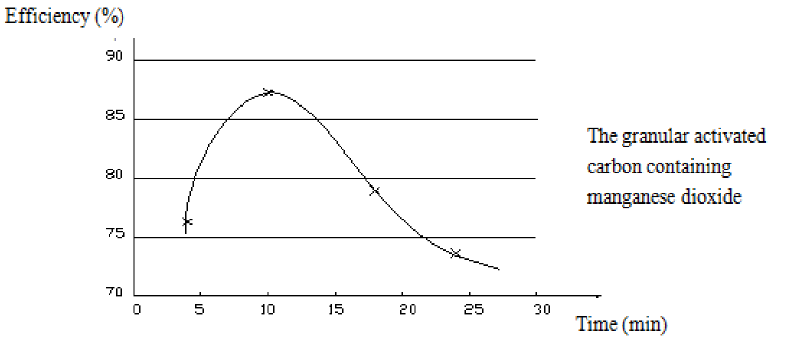

Fig. 4: The curve chart of desulphurization efficiency of the granular activated carbon containing manganese dioxide with time change.

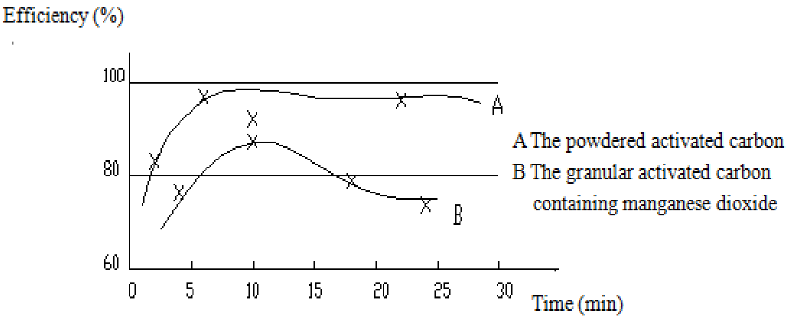

Fig. 5: The comparison curve chart of desulphurization efficiency with time change.

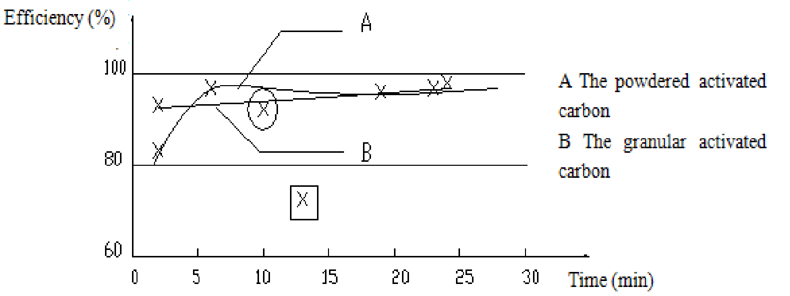

Fig. 6: The comparison curve chart of desulphurization efficiency with time change.

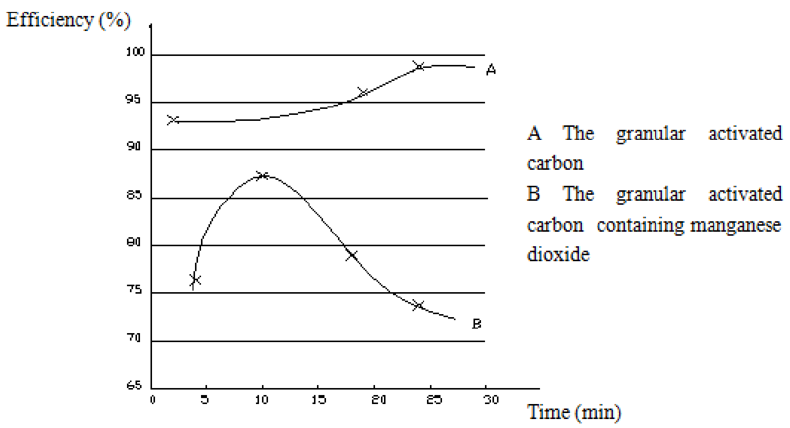

Fig. 7: The comparison curve chart of desulfurization efficiency with time change.

Therefore, the processing efficiency of powdered activated carbon is far better than that of granular activated carbon. This is because the advantages of the powdered activated carbon are beneficial to full contact for hydrogen sulphide gas and activated carbon, so the amount of removal and the desulphurization efficiency can be improved.

The comparison of desulphurization efficiency between granular activated carbon containing manganese dioxide and granular activated carbon: According to the above analysis, we get the comparison curve chart of desulphurization efficiency between granular activated carbon containing manganese dioxide and granular activated carbon with time change as shown in Fig. 7.

As can be seen from Fig. 7, the processing efficiency of granular activated carbon is far better than that of granular activated carbon containing manganese dioxide. It shows that manganese dioxide does not play any role.

CONCLUSIONS AND SUGGESTIONS

Conclusions

1. The processing efficiency of granular activated carbon is far better than that of regeneration. The more the number of regeneration, the worse the desulphurizatio efficiency.

2.The best activated carbon for desulphurization efficiency is the powdered activated carbon in the experiment.

3.After the addition of manganese dioxide in granular activated carbon, the desulphurization efficiency cannot be improved. It is clear that manganese dioxide does not play any role.

Suggestions

1. In order to adjust the flow in different situations and avoid reverse absorption phenomena, the large-scale flowmeter is selected.

2. The new plastic pipe is advised to be used. This is because the new pipe is soft, not easy

to break and leak.

3. After finishing each set of experiments, it is necessary to use iodine solution to soak the inlet of absorption bottle for a while. It makes the zinc sulphide to dissolve in iodine and maintains the smooth gas flow.

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