# Removal of H<sub>2</sub>S from Exhaust Gas by Use of Alkaline Activated Carbon

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**Abstract.** The purpose of this research was to select an activated carbon and alkaline solution blend that generated the best  $H_2S$  adsorption on alkaline-activated carbon.  $RB_2$  (activated carbon) impregnated with NaOH solution was shown to have the optimum  $H_2S$  removal efficiency. The optimum NaOH concentration was 50 mg per gram of carbon.  $H_2S$  adsorption via  $RB_2$ -NaOH $_{50}$  was five times that of a corresponding fresh-activated carbon. The adsorption equivalent of  $H_2S$  is nearly 1 (mol- $H_2S$ /mol-AOH), therefore,  $H_2S + AOH \rightarrow AHS + H_2O$  was the major reaction. The  $H_2S$  adsorption isotherm corresponded to the Freundlich isotherm.

**Keywords:** hydrogen sulfide, adsorption capacity, alkaline activated carbon, length of unused bed, adsorption wave

# Introduction

Hydrogen sulfide, an important industrial chemical, can be an asset or a liability. On the negative side, it presents an obstacle to the exploitation of petroleum and geothermal reservoirs, and its toxicity and offensive odor make it environmentally objectionable. The odor threshold value is only several ppb (Vigneron et al., 1994; Orszulik, 1997). Therefore,  $H_2S$  can be a malodorous air pollutant in the community (Windholz, 1976).

There are various plants that emit hydrogen sulfide, such as petrochemical plants, food-processing plants, semiconductor plants, sewage treatment plants, or pharmaceutical plants (Ikeda et al., 1988). Generally,

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the effluent gases from such plants are treated by washing followed by neutralization with chemicals, adsorption onto activated carbon, condensation, masking, or direct or catalytic combustion (Ikeda et al., 1988). Among these treatment methods, the washing method and/or the adsorption method have been used most frequently, and many kinds of impregnated activated carbons have been developed with superior deodorizing performance and used (Ikeda et al., 1988; Tsai et al., 1992; Tanada et al., 1981).

Many factors can affect the adsorption capacity of activated carbon; these include the specific surface area, pore size distribution, pore volume, and surface functional groups (Chiang et al., 1999). Generally, the adsorption capacity increases with the specific surface area due to the great number of adsorption sites (Gregg and Sing, 1982; Cheremisinoff and Ellerbursch, 1978).

Basically, there are two different preparation processes for activated carbon: physical and chemical activation. Physical activation involves carbonization of a carbonaceous precursor followed by activation of the resulting char in the presence of activating agents such as CO<sub>2</sub> or steam. Chemical activation, on the other hand, is a single step method (Mattson and Mark, 1971; Kinoshita, 1988; Bansal et al., 1990). The chemical agents used in the chemical process are normally alkali and alkaline earth metals or acids such as KOH, K<sub>2</sub>CO<sub>3</sub>, NaOH, Na<sub>2</sub>CO<sub>3</sub>, AlCl<sub>3</sub>, ZnCl<sub>2</sub>, MgCl<sub>2</sub> and H<sub>3</sub>PO<sub>4</sub> (Ikeda et al., 1988; Tsai et al., 1992; Verheyen et al., 1995; Ahmadpour and Do, 1996; Molina-Sabio et al., 1996; Deng et al., 1997).

Impregnated activated carbon removes gaseous pollutants via an irreversible reaction between the additives and pollutants. Hydrogen sulfide can be effectively captured by impregnated activated carbon and converted to sulfur when the operation temperature is between 24–170°C (Coskun and Tellefson, 1980).

A KI solution impregnated on activated carbon can act as a promoter and enhance the reaction rate (Klein and Henning, 1984). There are several impregnation solutions that may increase the adsorption capacity of  $\rm H_2S$  such as methulol-melamine-urea, n-butylamine, and urea. The adsorption capacity of  $\rm H_2S$  on impregnated activated carbon may be increased up to 1.5 times the value of raw activated carbon (Tanada et al., 1981). Thutsui and Tanada (1987) have used N-containing activated carbon to adsorb hydrogen sulfide and dimethyl sulfide.

Serre and coworkers investigated the adsorption of mercury on activated carbon. Their results indicated Hg removal increased with a decrease in particle size of the activated carbon at a given feed rate (Serre et al., 2001). In general, when the particle size increases, both the bulk mass transfer coefficient and the specific external surface area per gram of adsorbent decrease.

Freundlich's equation is used in chemisorption because it more frequently corresponds to the experimental results over a wider range of variables than the Langmuir equation. The Freundlich equation is an experimental isotherm that is displayed as follows (Gregg and Sing, 1982; Adamson, 1982):

$$q_e = KC_f^n \tag{1}$$

where  $C_f$  denotes equilibrium concentration,  $q_e$  represents the adsorption capacity at an equilibrium concentration and K is a constant where the value depends on

the temperature, specific surface area of adsorbent and other factors. Moreover, n denotes a constant, which depends on the temperature.

This research explored the feasibility of odor removal using alkaline impregnated activated carbon adsorption. Adsorption of  $H_2S$  to alkaline impregnated activated carbons was investigated. The relationship between adsorption capacity and additives chemical was studied. In addition, the particle size of the adsorbent is discussed in this study. Furthermore, the removal efficiency of alkaline impregnated activated carbons was also compared to the corresponding fresh activated carbon. Finally, the adsorption reactions of  $H_2S$  with activated carbon were investigated.

# **Materials and Methods**

Preparation of Impregnated Activated Carbons

Norit  $RB_2$  (size: 2 mm) and  $RB_4$  (size: 4 mm) carbons, made from peat, were used in this study. All solutions were prepared from chemicals provided by Merck Chemicals Company, Germany. A pH meter (model 440, Corning, USA) was used for pH measurement. Strong acid (1 N HCl) and base (1 N KOH, NaOH, Na<sub>2</sub>CO<sub>3</sub> and (K<sub>2</sub>CO<sub>3</sub>) were used for alkaline addition analysis. Unless otherwise noted, strong acid (0.1 M HClO<sub>4</sub>) and strong base (0.2 M NaOH) were used for pH adjustments.

Fifty grams of activated carbon were put into a stainless tube ( $\varnothing$ : 15 mm and length: 35 cm) with Nitrogen at 140°C for six hours to dewater and activate the carbons. Furthermore, the carbons were immersed in 1 N of NaOH, KOH, K2CO3 or Na2CO3 solution and stirred for 30 min. The immersed activated carbons were kept in a vacumm oven (1–0.1 mmHg at room temperature) for 20 min and then moved into a dryer (glass container with silica gel) for 190 min. The immersed carbons were filtered and dried in oven at 130°C for 60 hours. The activated carbons (RB2 or RB4), immersed in NaOH, KOH, K2CO3 and Na2CO3 solutions, are presented as RB(2or4)-NaOH, RB(2or4)-KOH, RB(2or4)-K2CO3 and RB(2or4)-Na2CO3, respectively.

#### Alkali Content on Carbon

The alkali content (NaOH, KOH, K<sub>2</sub>CO<sub>3</sub>, and Na<sub>2</sub>CO<sub>3</sub>) on each impregnated activated carbon was determined by titration. The carbon was placed in a vacuum oven

 $(10^{-2}-10^{-3}$  mmHg,  $105^{\circ}$ C) for 24 hours. 100 mL of 1 N hydrochloric acid (HCl) solution were added to the impregnated carbon and the slurry was shaken for 24 hours at 25°C. The supernatant was then titrated with a 1 N NaOH solution, and the content of alkali on carbon was calculated.

# Physicochemical Characteristics Analysis

The physical characteristics of activated carbon, including specific surface area, micropore area, total pore volume, micropore volume, and pore diameter were measured with liquid N<sub>2</sub> adsorption. A Micropore Analyzer (ASAP 2010, Micromeritrics Inc., USA) at 77 K was used.

Surface composition, namely hydrogen and oxygen, was analyzed with an Element Analyzer (Heraeus CHN-O Rapid Element Analyzer, USA). Five samples were analyzed in duplicate. Acetanilide was used as the standard. Another Element Analyzer (Tacussel Coulomax 78) used sulfanilic acid and 1-chloro-2, 4-dintrobenzene as standards to analyze sulfur and chlorine.

Samples for SEM/EDX (Hitachi-S-2500 Scanning Electronic Microscope with an Energy Dispersive X-ray Spectrometer KEVEX Level 4) analysis were coated with gold film to enhance the electric conductivity during the SEM operation. Elemental microprobe and distribution mapping techniques were used to analyze the sodium distribution on the alkali carbon surface.

### Adsorption Process

The adsorption column was 20 cm in length and 28 mm in diameter. The bottom of the adsorption column was packed with 10 cm of glass-beads. The glass-beads were used to assure the adsorbates were mixed well. Activated carbon was packed into the adsorption column over the glass beads. Cylinders of H<sub>2</sub>S (concentration: 8000 ppmv, Scott Gas company, USA) were blended with nitrogen gas. The flow rate was controlled by a mass flow meter (Sierra series 9000, USA) and ranged between 140 and 10,000 mL/min. The schematic diagram of the adsorption equipment and flow direction is shown as Fig. 1. The H<sub>2</sub>S concentration ranged from 20 to 8000 ppmv. The H<sub>2</sub>S concentration was analyzed by a Gas Chromatograph (Varian 3400, USA) with a Flame Photometry Detector and chromatographic column (G.S.Q.: 30 m, Ø: 0.53 mm). The injector, column, and detector temperatures were 200, 150, and 200°C, respectively. The retention time of the H<sub>2</sub>S was 1.68 min. The H<sub>2</sub>S adsorption capacity on activated carbon in a gas mixture was analyzed by the GC/FPD and calculated by column adsorption kinetic curves. Quality control was also conducted to ensure experimental data performance. Two methods were used to measure the adsorption capacity at equilibrium condition. The first method measured the weight of the adsorption column with a balance that stabilized at less than 0.5 mg. The other method used the gas chromatograph to monitor the effluent concentration of H<sub>2</sub>S in the column adsorption system. The amount

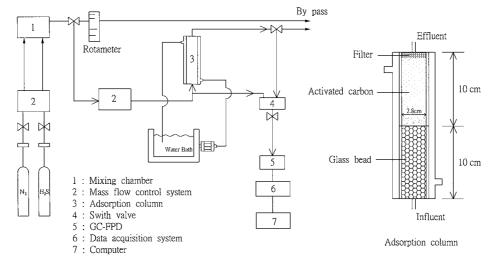


Figure 1. Schematic diagram of adsorption system.

of  $\rm H_2S$  adsorbed on the activated carbon was measured until the ratio of C(effluent concentration)/Co(influent concentration) was approximately 0.95. When the two methods were compared, the difference in adsorption capacity was less than 0.5 mg/g.

A Gas Chromatograph (GC: HP 5890 SERIES II, FPD, USA) with a chromatographic column (DB-624 fused silica capillary: 60 m, ID: 0.25 mm), and a Mass Spectrometer (MS: HP 5972 MASS SPECTROMETER, USA) were used to analyze the desorbed gas. Quality control was also conducted to ensure experimental data performance.

# Length of Unused Bed and Adsorption Wave

High (8,000 ppmv) and low (250 ppmv) influent  $H_2S$  concentrations were prepared for the column adsorption. Results were used to select the optimum adsorbent and impregnated alkaline solution for  $H_2S$  adsorption.

The length of the unused bed (LUB) was calculated as follows (McCabe et al., 1993; Weber, 1972):

$$LUB = \left(1 - \frac{q}{q_0}\right) \cdot L = \left(1 - \frac{t'}{t_0}\right) \cdot L \tag{2}$$

$$t_0 = \int_0^\infty \left(1 - \frac{C}{C_0}\right) dt \tag{2a}$$

$$t' = \int_0^\infty \left(1 - \frac{C}{C_0}\right) dt \tag{2b}$$

where

L: length of adsorption bed,

 $q_0$ : mass of adsorbate per mass of adsorbent at equilibrium,

q: mass of adsorbate per mass of the adsorbent,

 $C_0$ : influent adsorbate concentration,

C: outlet adsorbate concentration when time is t

 $t_0$ : time of adsorbent at equilibrium,

t': integrated time.

The length of the adsorption wave (Lz) was calculated as follows (McCabe et al., 1986; Weber, 1972):

$$L_z = L \cdot \frac{(t_e - t_b)}{(t_e - t_f)} \tag{3}$$

$$t_{\rm f} = (1 - F) \times (t_{\rm e} - t_{\rm b})$$
 (3a)

$$F = \int_0^l \left(1 - \frac{C}{C_0}\right) d\left[\frac{(t - t_b)}{(t_e - t_b)}\right]$$
 (3b)

where

 $t_e$ : equilibrium time when C reaches  $0.9C_0$ ,

 $t_b$ : breakthrough time when C reaches  $0.1C_0$ 

t: time

 $t_{\rm f}$ : formation time,

F: fraction of total utilization.

#### **Results and Discussion**

This research investigated the adsorption characteristics of  $H_2S$  on alkaline activated carbon. KOH,  $K_2CO_3$ , NaOH and  $Na_2CO_3$  were selected as the alkaline reagents. Those were impregnated to enhance the adsorption capacity of the activated carbon. The typical column adsorption kinetic curves for 4 mm and 2 mm pellets are shown as Figs. 2 and 3. Resulted indicated the time of breakthrough point of 4 mm pellets was earlier than that of 2 mm pellets.

Length of Unused Bed (LUB) and Adsorption Wave (Lz)

Tables 1 and 2 show the length of the unused bed (LUB) and  $H_2S$  adsorption wave (Lz) on the activated carbons. There were two influent conditions for  $H_2S$ : 1) 8,000 ppm and 140 mL/min, and 2) 250 ppm and 10,000 mL/min. The length of the unused bed for the RB<sub>4</sub> alkaline activated carbons was between 0.68 and 2.98 cm and that of the RB<sub>2</sub> alkaline activated carbons

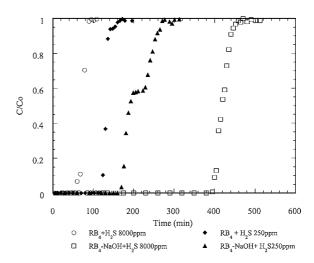


Figure 2. Typical column adsorption kinetic curves for 4 mm pellets.

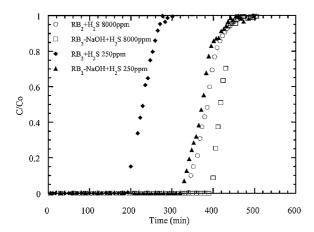


Figure 3. Typical column adsorption kinetic curves for 2 mm pellets.

between 0.63 and 2.46 cm. Generally, the LUB of the impregnated alkaline activated carbon was less than the activated carbon because the alkaline reacted with the H<sub>2</sub>S and enhanced the adsorption rate. In addition, the lower influent concentration and greater influent flow rate (250 ppm and 10,000 mL/min) generated the larger LUB. The lower concentration and greater flow rate reduced the adsorption site- H<sub>2</sub>S reaction (lower retention time in the adsorption bed). The LUB sequence at 8000 ppm and 140 mL/min was: RB<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub> >  $RB_2 > RB_4 > RB_4-Na_2CO_3 > RB_4-KOH > RB_2 KOH > RB_4-NaOH > RB_4-K_2CO_3 > RB_2-Na_2CO_3 >$ RB<sub>2</sub>-NaOH. The LUB sequence at 250 ppm and 10,000 mL/min was:  $RB_4 > RB_2$ -NaOH  $> RB_4$ -KOH  $\cong RB_2$ - $K_2CO_3 > RB_2 \cong RB_4-Na_2CO_3 > RB_4-NaOH > RB_2 Na_2CO_3 > RB_4-K_2CO_3 > RB_2-KOH$ .

The length of the  $H_2S$  adsorption wave on the alkaline activated carbon was less than that on activated carbon. The Lz sequence was:  $RB_4 > RB_2 > RB_4$ -Na<sub>2</sub>CO<sub>3</sub>

 $> RB_2-KOH > RB_4-NaOH > RB_2-Na_2CO_3 > RB_4 KOH > RB_4-K_2CO_3 > RB_2-NaOH > RB_2-K_2CO_3$ at 8,000 ppm and 140 mL/min. The Lz sequence at 250 ppm and 10,000 mL/min was:  $RB_4 > RB_2-K_2CO_3$  $> RB_4-Na_2CO_3 > RB_4-KOH > RB_4-NaOH > RB_2$  $> RB_2-Na_2CO_3 > RB_4-K_2CO_3 > RB_2-KOH > RB_2-KOH$ NaOH. In general, the length of the adsorption wave was larger at the lower concentration and higher flow rate (retention time was lower). In addition, the RB2 adsorption wave and length of the unused bed was smaller than that of RB<sub>4</sub>. The RB<sub>2</sub> adsorption column was more completely utilized than the RB<sub>4</sub>. NaOH was selected as the most effective additive alkaline. Results indicated that the smaller adsorbent particle size corresponded to the smaller length of the unused bed. Furthermore, the smaller adsorbent particle sizes enhanced the adsorption rate. Increasing the particle size resulted in a lower specific external surface area and a lower external mass transfer coefficient as well as an increase in the internal mass transfer resistance.

### Physicochemical Characteristics of Surface

Two separate samples were taken from each adsorbent for physical characteristic analysis. In order to assure the quality of the analysis, the two samples were analyzed in duplicate. The surface physical characteristics included BET specific surface area, micropore area, micropore volume, pore size distribution and pore diameter. Table 3 shows the results of the physical characteristics of the analyzed activated carbon samples.

The BET specific surface area of the RB<sub>2</sub> activated carbon and the alkaline activated carbon were 931 and 758 m<sup>2</sup>/g, respectively. The specific surface area of the alkaline activated carbon was 18.6% less that of the RB<sub>2</sub> samples. Apparently NaOH altered the pore

| Table 1. | Adsorption wave and | length of unused | bed of H2S adsorbs | on RB <sub>4</sub> and alkaline RB <sub>4</sub> . |
|----------|---------------------|------------------|--------------------|---------------------------------------------------|
|          |                     |                  |                    |                                                   |

| Concentration | Parameter                     | $RB_4$ | RB <sub>4</sub> -KOH | RB <sub>4</sub> -Na <sub>2</sub> CO <sub>3</sub> | RB <sub>4</sub> -K <sub>2</sub> CO <sub>3</sub> | RB <sub>4</sub> -NaOH |
|---------------|-------------------------------|--------|----------------------|--------------------------------------------------|-------------------------------------------------|-----------------------|
| 8000 ppm      | Adsorption wave (Lz) cm       | 2.48   | 0.92                 | 1.63                                             | 0.90                                            | 0.96                  |
| 140 mL/min    | Length of unused bed (LUB) cm | 1.11   | 0.90                 | 1.07                                             | 0.68                                            | 0.69                  |
|               | Adsorption capacity (mg/g)    | 4.21   | 23.2                 | 17.4                                             | 29.5                                            | 23.7                  |
| 250 ppm       | Adsorption wave (Lz) cm       | 6.39   | 4.86                 | 5.13                                             | 2.62                                            | 4.65                  |
| 10000 mL/min  | Length of unused bed (LUB) cm | 2.98   | 2.26                 | 2.18                                             | 1.64                                            | 1.81                  |
|               | Adsorption capacity (mg/g)    | 3.79   | 10.0                 | 2.05                                             | 10.8                                            | 6.63                  |

Table 2. Adsorption wave and length of unused bed of H<sub>2</sub>S adsorbs on RB<sub>2</sub> and alkaline RB<sub>2</sub>.

| Parameter                     | $RB_2$                                                                                                                                 | RB <sub>2</sub> -KOH                                                                                                                                            | RB <sub>2</sub> -Na <sub>2</sub> CO <sub>3</sub>                                                                                                                                         | RB <sub>2</sub> -K <sub>2</sub> CO <sub>3</sub>                                                                                                                                                                                                                                                                                             | RB <sub>2</sub> -NaOH                                                                                                                                                                                                                                                                                                                                                                              |
|-------------------------------|----------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Adsorption wave (Lz) cm       | 1.84                                                                                                                                   | 1.29                                                                                                                                                            | 0.96                                                                                                                                                                                     | 0.65                                                                                                                                                                                                                                                                                                                                        | 0.83                                                                                                                                                                                                                                                                                                                                                                                               |
| Length of unused bed (LUB) cm | 1.72                                                                                                                                   | 0.80                                                                                                                                                            | 1.76                                                                                                                                                                                     | 0.65                                                                                                                                                                                                                                                                                                                                        | 0.63                                                                                                                                                                                                                                                                                                                                                                                               |
| Adsorption capacity (mg/g)    | 22.3                                                                                                                                   | 33.5                                                                                                                                                            | 25.5                                                                                                                                                                                     | 22.5                                                                                                                                                                                                                                                                                                                                        | 21.5                                                                                                                                                                                                                                                                                                                                                                                               |
| Adsorption wave (Lz) cm       | 3.31                                                                                                                                   | 2.32                                                                                                                                                            | 3.01                                                                                                                                                                                     | 5.28                                                                                                                                                                                                                                                                                                                                        | 1.28                                                                                                                                                                                                                                                                                                                                                                                               |
| Length of unused bed (LUB) cm | 2.20                                                                                                                                   | 1.01                                                                                                                                                            | 1.73                                                                                                                                                                                     | 2.26                                                                                                                                                                                                                                                                                                                                        | 2.46                                                                                                                                                                                                                                                                                                                                                                                               |
| Adsorption capacity (mg/g)    | 8.25                                                                                                                                   | 12.5                                                                                                                                                            | 14.0                                                                                                                                                                                     | 12.5                                                                                                                                                                                                                                                                                                                                        | 13.5                                                                                                                                                                                                                                                                                                                                                                                               |
|                               | Adsorption wave (Lz) cm Length of unused bed (LUB) cm Adsorption capacity (mg/g) Adsorption wave (Lz) cm Length of unused bed (LUB) cm | Adsorption wave (Lz) cm 1.84 Length of unused bed (LUB) cm 1.72 Adsorption capacity (mg/g) 22.3 Adsorption wave (Lz) cm 3.31 Length of unused bed (LUB) cm 2.20 | Adsorption wave (Lz) cm 1.84 1.29 Length of unused bed (LUB) cm 1.72 0.80 Adsorption capacity (mg/g) 22.3 33.5 Adsorption wave (Lz) cm 3.31 2.32 Length of unused bed (LUB) cm 2.20 1.01 | Adsorption wave (Lz) cm       1.84       1.29       0.96         Length of unused bed (LUB) cm       1.72       0.80       1.76         Adsorption capacity (mg/g)       22.3       33.5       25.5         Adsorption wave (Lz) cm       3.31       2.32       3.01         Length of unused bed (LUB) cm       2.20       1.01       1.73 | Adsorption wave (Lz) cm       1.84       1.29       0.96       0.65         Length of unused bed (LUB) cm       1.72       0.80       1.76       0.65         Adsorption capacity (mg/g)       22.3       33.5       25.5       22.5         Adsorption wave (Lz) cm       3.31       2.32       3.01       5.28         Length of unused bed (LUB) cm       2.20       1.01       1.73       2.26 |

Table 3. Physicochemical characteristics of RB<sub>2</sub> and RB<sub>2</sub>-NaOH. a,b

| Physicochemical characteristics |                                       | $RB_2$ | RB <sub>2</sub> -NaOH | Percent of variation (%) |  |
|---------------------------------|---------------------------------------|--------|-----------------------|--------------------------|--|
| Physical characteristics        | BET surface area (m <sup>2</sup> /g)  | 931    | 758                   | -18.6                    |  |
|                                 | Micropore area (m <sup>2</sup> /g)    | 646    | 529                   | -18.1                    |  |
|                                 | Micropore volume (cm <sup>3</sup> /g) | 0.304  | 0.249                 | -18.1                    |  |
|                                 | Pore diameter (Å)                     | 15.61  | 15.60                 | -0.1                     |  |
| Elemental composition (%)       | N                                     | < 0.01 | < 0.01                | _                        |  |
|                                 | C                                     | 74     | 71                    | -4.1                     |  |
|                                 | Н                                     | 2.9    | 2.3                   | -20.7                    |  |
|                                 | S                                     | 0.5    | 0.3                   | -40                      |  |
|                                 | Cl                                    | < 0.01 | < 0.01                | _                        |  |

<sup>&</sup>lt;sup>a</sup>Sample number is 3 and the measurement of pore diameter is less than 2000 Å.

size and pore size distribution which in turn decreased the specific surface area via precipitation.

The micropore area measured  $646 \text{ m}^2/\text{g}$  and  $529 \text{ m}^2/\text{g}$  for the RB<sub>2</sub> and alkaline activated carbons, respectively. Results indicate that NaOH decreased the micropore surface area 67% of the specific surface area.

Upon NaOH impregnation, the micropore volume also decreased from  $0.304~\rm cm^3/g$  to  $0.249~\rm cm^3/g$  (a decrease of 18.1%) and the pore diameter from  $15.61~\rm to$   $15.57~\rm Å$  (a decrease of 0.3%). There was no obvious difference in pore diameter between the RB<sub>2</sub> and alkaline activated carbons.

Table 3 shows the major surface element concentrations of the  $RB_2$  and alkaline activated carbons. The  $RB_2$  concentrations of nitrogen, carbon, hydrogen and oxygen were <0.01%, 74%, 2.83% and 23%, respectively. The concentrations of N, C, H, and O on the alkaline activated carbon were <0.01%, between 69–75%, between 1.76–2.28% and between 23–29%, respectively.

Figures 4 and 5 shows the SEM photograph and sodium distribution of alkaline activated carbon. Results indicated that the NaOH crystal formed on the surface of activated carbon. This could be observed with the naked eye and a SEM photograph. Furthermore, the EDX analysis indicated that the sodium was evenly distributed on the surface of alkaline activated carbon.

# NaOH Impregnated on Activated Carbon

In order to understand the influence of the impregnation procedure on the quantity of NaOH impregnated on an activated carbon, this investigation varied vacuum and immersion durations. The results are shown as Fig. 6. When the vacuum and stationary duration were extended, the quantity of NaOH impregnated on activated carbons was enhanced. The adsorption capacity of  $\rm H_2S$  on four different impregnated activated carbons is shown as Fig. 7.

<sup>&</sup>lt;sup>b</sup>Percent of variation =  $[(RB_2-NaOH-RB_2)/RB_2] \times 100\%$ .

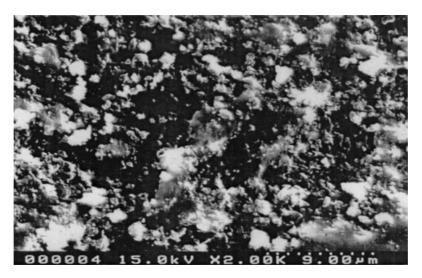


Figure 4. SEM photomicrograph of RB2-NaOH.

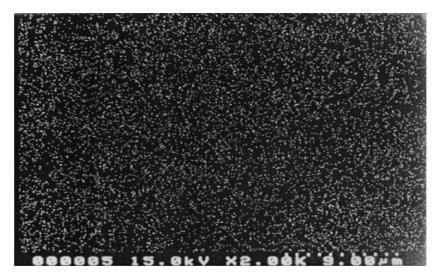


Figure 5. SEM/EDX photograph of RB2-NaOH.

When the  $H_2S$  influent was 0.92 mg/min (250 ppmv, 3.1 L/min, at 25°C), the adsorption capacity of  $RB_2$ -NaOH<sub>50</sub> (50 mg of NaOH impregnated on 1 g of activated carbon) was 43.5 mg/g and that of the other adsorbents were  $RB_2$ -NaOH<sub>65</sub> (33.3 mg/g),  $RB_2$ -NaOH<sub>70</sub> (29.3 mg/g), and  $RB_2$ -NaOH<sub>23</sub> (19.4 mg/g).

Comparison of the adsorption capacity of NaOH impregnated activated carbons to H<sub>2</sub>S adsorption, indicated that RB<sub>2</sub>-NaOH<sub>50</sub> had the largest adsorption capacity of the four NaOH impregnated activated carbons. Results showed that 50 mg of NaOH

impregnated on 1 g of activated carbon was optimum. NaOH could completely adsorb on the active sites of the activated carbon. When the NaOH concentration was greater than 50 mg-NaOH/g-activated carbon, the pores of activated carbon were blocked. The drying procedure generated NaOH crystal and led to an incomplete reaction with  $\rm H_2S$  and reduced the adsorption capacity. When the concentration of NaOH impregnated on the activated carbon was smaller than 50 mg-NaOH/g-activated carbon, not all of the activated carbon active sites completely reacted with the NaOH.

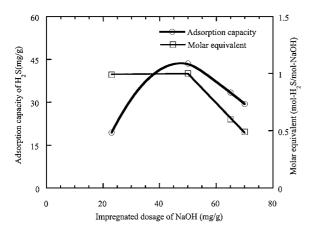


Figure 6. Adsorption capacity of H2S on various RB2-NaOH.

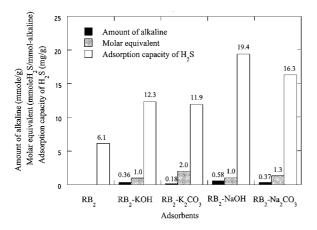


Figure 7. Adsorption capacity of four kinds of alkaline impregnated activated carbons.

# Adsorption Reaction

In order to explore the reaction of alkaline activated carbons and odor gases, thermodesorption was used to desorb the adsorbates. GC/FPD and GC/MS were used to analyze the components of the desorption gas.  $H_2S$  and  $CS_2$  were analyzed in the desorption gas from the alkaline activated carbons by GC/MS. Results indicated that  $H_2S$  adsorption on the alkaline activated carbon was both physical sorption and chemisorption.

The quantity and sequence of alkaline impregnation on  $RB_2$  activated carbon  $(RB_2)$  were NaOH  $(0.58 \text{ mmol/g}) > Na_2CO_3$  (0.37 mmol/g) > KOH  $(0.36 \text{ mmol/g}) > K_2CO_3$  (0.18 mmol/g). As the NaOH molecular size is smaller than that of the others, it is easier to transport into the pores of activated carbon. NaOH had the largest impregnated capacity.

The H<sub>2</sub>S inflow loading was 0.87 mg/min and the adsorption molar equivalents were RB<sub>2</sub>-KOH (1.00), RB<sub>2</sub>-K<sub>2</sub>CO<sub>3</sub> (1.95), RB<sub>2</sub>-NaOH (0.99), and RB<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub> (1.29 mol-H<sub>2</sub>S/mol-alkaline) (Fig. 7). The results denote that when H<sub>2</sub>S adsorbed on RB<sub>2</sub>-KOH and RB<sub>2</sub>-NaOH, one molecule of H<sub>2</sub>S could react with one molecule KOH or NaOH. When the H<sub>2</sub>S adsorbed on RB<sub>2</sub>-K<sub>2</sub>CO<sub>3</sub> and RB<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub>, the reactions included chemical and physical adsorption. Activated carbon cannot be completely occupied by RB<sub>2</sub>-K<sub>2</sub>CO<sub>3</sub> and RB<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub>, therefore chemical and physical adsorption both exist on RB<sub>2</sub>-K<sub>2</sub>CO<sub>3</sub> and RB<sub>2</sub>-Na<sub>2</sub>CO<sub>3</sub>.

The reactions of  $H_2S$  on non-alkaline activated carbon could be proposed as follows: the adsorption of  $H_2S$ , the catalytic oxidation of  $H_2S$  and the reaction of  $H_2S$  with alkaline ash content on activated carbon.

The reactions of  $H_2S$  and four kinds of alkaline are as follows:

### 1. NaOH or KOH

H<sub>2</sub>S is a diproton acid that reacts with a hydroxide group as follows:

$$H_2S + AOH \rightarrow AHS + H_2O$$
 (4)

or

$$H_2S + 2AOH \rightarrow A_2S + 2H_2O$$
 (4a)

where A is denoted as K or Na.

One mole of AOH reacts with one mole  $H_2S$  in reaction (4). If the reaction is performed as reaction (4a), one mole of AOH will react with 0.5 mole of  $H_2S$ . Since the adsorption equivalent of  $H_2S$  is nearly 1 (mol- $H_2S$ /mol-AOH), reaction (4) is the major reaction.

# 2. Na<sub>2</sub>CO<sub>3</sub> or K<sub>2</sub>CO<sub>3</sub>

$$H_2S + A_2CO_3 \rightarrow AHS + AHCO_3$$
 (5)

$$H_2S + A_2CO_3 \rightarrow A_2S + H_2CO_3 \qquad (5a)$$

Since the coefficient of reactions (5) and (5a) are identical, one cannot determine the predominant reaction.

 $H_2S$  was desorbed at 130°C and 18 mL/min. The sequence of desorption efficiency for the five activated carbons was  $RB_2~(90.12\%) > RB_2\text{-}K_2\text{CO}_3~(21.54\%) > RB_2\text{-}Na_2\text{CO}_3~(9.12\%) > RB_2\text{-}KOH~(1.18\%) > RB_2\text{-}NaOH~(0.77\%).}$ 

The desorption temperature could not break the chemical bond that was formed by chemisorption. Therefore, the recovery efficiency of thermal desorption can be presented as the percent of  $H_2S$  physically adsorbed on the activated carbon.

When the percentage of thermal desorption and molar equivalent of alkaline activated carbon are denoted,  $RB_2$ - $K_2CO_3$  had the higher molar equivalent (1.95) and the greater percentage (21.54%) of thermal desorption. The adsorption sites were not completely occupied by  $K_2CO_3$  due to physical adsorption.

# Adsorption Capacity

In order to compare the deviations of  $H_2S$  adsorption capacity on activated carbon ( $RB_2$ ) and alkaline activated carbon ( $RB_2$ -NaOH), the research was performed at different concentrations. Figure 8 summarizes the results of the adsorption capacity (at varying concentrations of adsorbates) on the activated carbons. The adsorption isotherm is denoted by the effect of influent concentration on the adsorption capacity of  $RB_2$ -NaOH. The  $H_2S$  adsorption capacity was nearly 20 mg/g on the  $RB_2$ -NaOH. Increased influent loading yielded a greater adsorption capacity. In addition, the adsorption isotherm corresponded to the Freundlich isotherm.

The Fredundlich adsorption equation is favored for adsorption because n < 1, that is the higher the influent concentration the greater the adsorption capacity. The adsorption isotherm is smoother at low concentrations than at high concentrations. When the concentration was increased, the adsorption isotherm peaked.

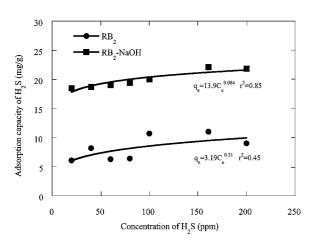


Figure 8. Adsorption capacity of H2S on RB2 and RB2-NaOH.

Increased concentration enabled the adsorbate to flow into the adsorption column. The outlet concentration reached saturation. But as influent concentration increased to greater than 161 ppm, the adsorption capacity lowered. This result shows that the concentration was in excess of the adsorbent loading.

Comparing the two adsorbents,  $RB_2$ -NaOH adsorbed more odor gas than  $RB_2$ . The adsorption capacities of  $RB_2$  varied with the influent concentration. Low concentrations of odor gases were adsorbed on  $RB_2$ , which corresponds to the Freundlich isotherm, but when the concentration was greater than 161 ppm, the adsorption capacity was decreased.

#### **Conclusions**

Experimental results indicate that the concentration of  $H_2S$  can be effectively controlled by alkaline impregnated activated carbons. All the alkaline solutions used in the study were found to significantly increase the adsorption capacity of  $H_2S$  on impregnated activated carbons. NaOH increased the adsorption capacity more than the others. According to the adsorption wave and length of unused bed results,  $RB_2$  was the best adsorbent for  $H_2S$  adsorption. The  $RB_2$ -NaOH fit the adsorption isotherm model better than  $RB_2$ . When the optimum NaOH concentration was 50 mg per gram carbon, the adsorption capacity of  $RB_2$ -NaOH $_{50}$  increased to five times that of corresponding fresh activated carbon.

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