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B. Sanbundit & D. Chatsiriwech

Chulalongkorn University

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Chemisorption of Hydrogen Cyanide from Pyrolysis of Tobacco Leaves with Chloramine-T

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ABSTRACT

Hydrogen cyanide (HCN) was produced by flameless combustion of dried stripped tobacco leaves, and was adsorbed directly by chloramine-T, which penetrated on a filter pad. Both amounts of breakthrough HCN and HCN on the filter pad were measured. The concentration of HCN was produced steadily at 0.7-0.8 mg HCN per dm³ of smoke. At least 60% of HCN produced was adsorbed on the filter pad. For the given 245 cm³ of smoke with the flow rate of 17.5 cm³ per second, the amount of HCN produced was reduced to below 0.05 mg by adding at least 20 mg of chloramine-T. Direct adsorption of HCN on chloramine-T was chemisorption. Almost 90% of the amount of HCN produced was adsorbed irreversibly on filter pad containing 45 mg of chloramine-T.

Keywords: chemisorption; hydrogen cyanide; tobacco leaves; chloramine-T.

Author α ο: Department of Chemical Engineering, Faculty of Engineering, Chulalongkorn University, Bangkok 10330, Thailand.

I. INTRODUCTION

An increase in population required more both energy and food. Various manufacturers looked for appropriate alternative waste materials; such as wood, bark, bagasse, rice husk and agricultural straw, to produce energy instead of conventional fossil fuels in order to reduce costs. Meanwhile, cultivators still burnt the remaining after cultivation for replantation, as usual, for minimising costs. A large number of toxic gases could be released by combustion of these biomass (Sung-Chul Yi et al. 2005, J. Bekkering et al. 2010, Shuai Yuan et al. 2011, Qiangqiang Ren et

al. 2011, Zi-Wei Zhanga et al. 2011 for instance). As various biomass had nitrogen as a constituent, besides carbon and hydrogen, nitrogen compounds, such as H₂NCO, HCN, NO_x, N₂O, and NH₃, could be generated by combustion (Alexandra 2004, Siegler 1976, Hua Lu Karlsson 2004).

Hydrogen cyanide, HCN, could be produced by the combustion of various synthetic materials containing nitrogen constituent, such as polyamide (nylon), 1,3,5-triazine-2,4,6-triamine (melamine) and polyacrylonitrile. In addition, it could be produced by the pyrolysis of biomass containing amino acids, heterocyclic compounds of nitrogen or dicarboxylic acids (Johnson and Kang 1971, Tso TC et al, 1982, Qiangqiang Ren et al. 2011, Karl-Martin Hansson 2004, W.S. Rickert and P.B. Stockwell 1979). Even though the amount of hydrogen cyanide, HCN, from biomass pyrolysis was small, it was sufficiently harmful to public health. It was so toxic asphyxiants that it inhibited the metabolism process. In the circumstances, cyanide ions would block oxidative respiration and might cause cancer (Bhattacharya and Flora 2009, World Health Organisation (WHO) 2004).

Many substances, such as carbon, activated carbon, zeolites, catalysts, nanotube and architectural materials, were studied on the adsorption of hydrogen cyanide. Oliver et al. (2005) revealed that copper containing synthetic activated carbons produced from porous sulfonated styrene / divinylbenzene resins could adsorb hydrogen cyanide gas effectively without the formation of (CN)₂. Rajakovic et al. (1995) reported that the carbon materials impregnated with metal organic compounds, especially copper (II) - tartrate, could adsorb hydrogen cyanide.

Furthermore, the impregnated organic compounds could adsorb more efficiently than inorganic compounds. While R.R. Kotdawala *et al.* (2008) exposed that the polar compounds, containing carboxyl, hydroxyl or carbonyl groups, had high efficiency for adsorbing hydrogen cyanide, depending on their induced dipole and charge-dipole interactions. Peter Branton *et al.* (2009) also reported that the efficiency of the adsorption of hydrogen cyanide depended on physisorption or chemisorption. In the World Wars, hydrogen cyanide, HCN, was used as a constituent of chemical weapons, then chloramine-T (N-chloro-4 methylbenzenesulfonamide, $C_7H_7ClNO_2S.Na.3H_2O$) and derivatives were used to protect poison gas. In addition, chloramine-T reacted readily with mustard gas to yield a harmless crystalline sulfonamide (Yasukazu Ura, and Gozyo Sakata 2003). In alkalinity solution, chloramine-T reacted with HCN to produce cyanogen chloride.

This study was carried out to investigate the effect of amount of chloramine-T (N-chloro-4-methylbenzene sulfonamide) on reducing the concentration of hydrogen cyanide, HCN, in the combustion gas by direct adsorption. According to the previous researches on the pyrolysis of tobacco leaves, hydrogen cyanide, HCN, could be generated directly by the pyrolysis of tobacco dried leaves, which was used for this study. The influence of both amount of chloramine-T and volume of pyrolysis smoke on the adsorption of hydrogen cyanide were investigated.

II. MATERIALS AND METHODS

2.1 Preparation of tobacco leaves for producing HCN

Dried Virginia tobacco leaves, which were supplied by the Tobacco Authority of Thailand, were used as raw materials for producing hydrogen cyanide, HCN, at the Research and Development Department of Tobacco Authority of Thailand. They were cut into fine strips. A gram of stripes of dried leaves was packed in a paper cylinder with a diameter of 8 mm. and length of 84 mm. All packs of dried leaves were kept in a container at 22°C with 60% relative humidity in

order to control moisture contents of dried leaves. Meanwhile, the glass fibre filter pads, with 2 mm thick and 44 mm diameter, were dipped into the solutions of chloramine-T (N-chloro-4-methylbenzene sulfonamide, $CH_3C_6H_4SO_2 NCl.Na.3H_2O$) for removal of hydrogen cyanide from smoke by adsorption. The concentration of these solutions was varied from 2.8 to 28 g/dm³ for investigating the effects of the amount of chloramine-T on the hydrogen cyanide removal efficiency by adsorption. The filter pads were dried at 100°C for 3 hours. They were put in the adsorption experimental set, consisting of hydrogen cyanide generation with flameless combustion, adsorption on filter pad, collecting the breakthrough hydrogen cyanide in the 0.625 M sodium hydroxide solution, and a piston vacuum pump, respectively, as shown in Fig.1.

2.2 HCN Adsorption Experiment

Each pack of dried leaves was combusted flamelessly at one end of the pack, while the combustion gas or smoke was pumped by the piston vacuum pump through the filter pad containing chloramine-T, as well as without chloramine-T as a reference, with the flow rate of 17.5 cm³/s, as shown in Fig. 1. The breakthrough hydrogen cyanide was collected in the 4⁰ cm³ of 0.625 M sodium hydroxide solution for analysing the breakthrough amount of hydrogen cyanide.

Each condition of adsorption experiment was repeated at least 4 times. The adsorption of hydrogen cyanide in smoke was also carried out with the filter pads containing proline (HNC₄H₇COOH) under the same conditions for comparison purpose. In addition, various compounds on the filter pad after adsorption was analysed by GC-MS (Agilent 6890 GC with 5973 MSD and 7638 series. injector, HP- INNOWAX capillary column).

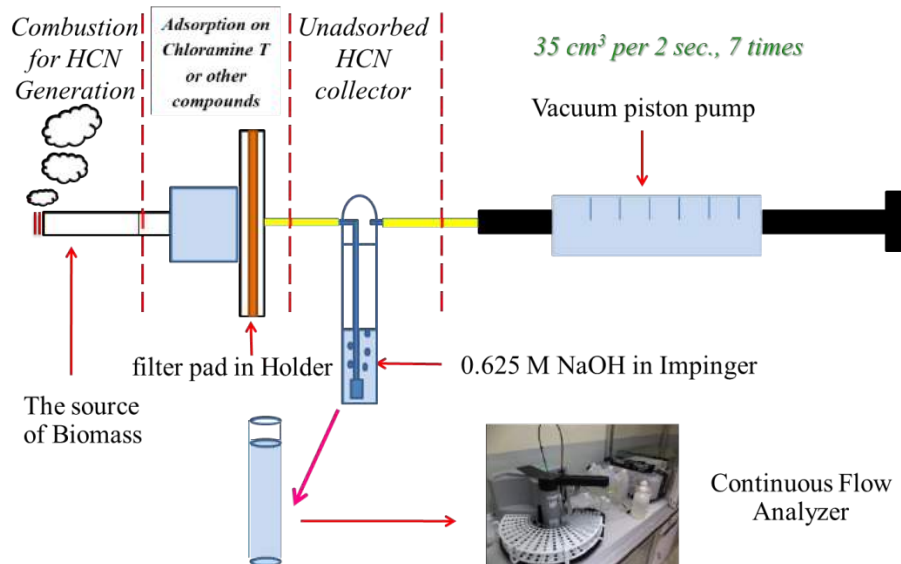


Fig. 1: The Experimental Set for Adsorption of Hydrogen Cyanide From Combustion of Dried Stripes of Tobacco Leaves.

2.3 Adsorption Performance Evaluation

The sodium hydroxide solution, which collected the breakthrough hydrogen cyanide, was analysed by the continuous flow analyser, Skalar, with the detection limits of CN- of 0.02 mg/dm³.

In addition, the amount of hydrogen cyanide adsorbed on the pad was also analysed in the same manner as the breakthrough hydrogen cyanide above, by dissolving the adsorbed hydrogen cyanide on the pad in the 40 cm³ of 0.625 M sodium hydroxide solution. Without

chloramine-T on the filter pad, both breakthrough amount and adsorbed amount of hydrogen cyanide obtained were taken into account as the total amount of hydrogen cyanide in the smoke. The adsorption efficiency was calculated by the ratio of the amount of adsorbed hydrogen cyanide to the total amount of hydrogen cyanide in smoke, and by the difference of the breakthrough amount of hydrogen cyanide from the total amount of hydrogen cyanide in the smoke as defined in the Eq. (1) and Eq. (2) respectively.

$$\text{Adsorption Efficiency} = \frac{\text{Amount of HCN adsorbed} \times 100}{\text{Amount of total HCN in smoke}} \quad (1)$$

$$\text{Adsorption Efficiency} = \frac{(\text{Amount of total HCN in smoke} - \text{Amount of unadsorbed HCN}) \times 100}{\text{Amount of total HCN in smoke}} \quad (2)$$

III. RESULTS AND DISCUSSION

3.1 The Adsorption of HCN on Chloramine-T

For consecutive flameless combustion of 6 packs of dried tobacco leaves, the total amount of hydrogen cyanide in smoke increased proportionally to the cumulative amount of smoke flowing through the filter pad, as shown in Fig.2. Without chloramine-T on the filter pad, the cumulative amount of hydrogen cyanide adsorbed on the filter pad increased proportionally to the

cumulative amount of smoke in the same manner as the variation of total amount of hydrogen cyanide produced. However, the amount of hydrogen cyanide adsorbed on the filter pad was slightly above a half of the total amount. With 10 mg chloramine-T on the filter pad, the cumulative amount of hydrogen cyanide adsorbed increased proportionally to the cumulative amount of smoke but was slightly less than that on the filter pad without chloramine-T.

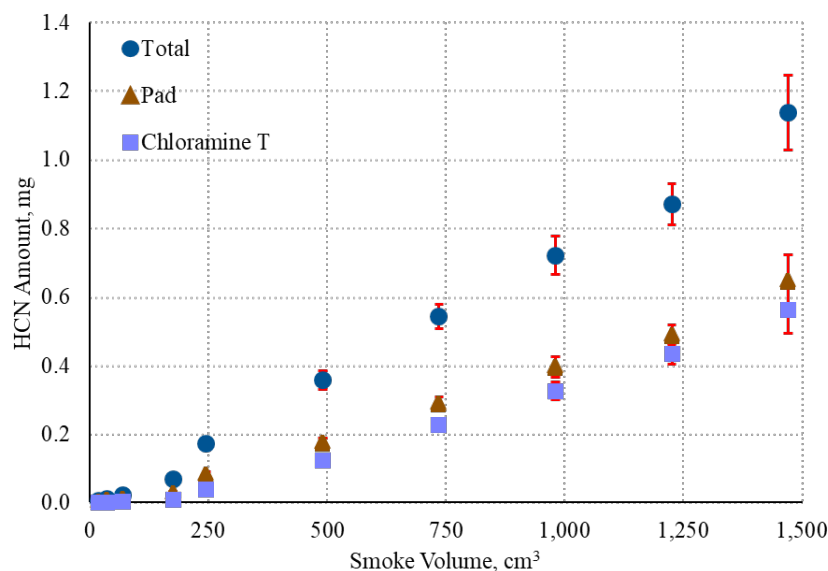


Fig. 2: The Comparison of the Cumulative Amount of Breakthrough HCN with the Cumulative Total Amount of HCN in smoke with flow rate of 17.5 cm³/s.

The concentration of total hydrogen cyanide in smoke was about 0.3-0.4 mg/dm³ for the first pack of dried tobacco leaves, which could produce 245 cm³ of smoke. It became about 0.7-0.8 mg/dm³ for the other 5 packs of dried tobacco leaves, as shown in Fig.3. Without chloramine-T on the filter pad, the concentration of breakthrough hydrogen cyanide in the smoke increased from 0.1 mg/dm³ to just above 0.2 mg/dm³ for the first pack of dried tobacco leaves. Then, it became about 0.3-0.4 mg/dm³ for the rest 5 packs of tobacco leaves. With 10 mg of chloramine T on the filter pad, the concentration of breakthrough hydrogen cyanide in the smoke varied in the same manner as the results of without chloramine-T on the filter pad. However, the concentration of breakthrough hydrogen cyanide was slightly less than the case without chloramine-T.

The adsorption efficiencies of the filter pad without chloramine-T, evaluated by both the direct measured amount adsorbed on the pad, Eq. (1), and the mass balance of the breakthrough amount, Eq.

(2), yielded the same values. Hence, it dropped from just above 60% to above 40% from the first pack of dried tobacco leaves, as shown in Fig.4. Afterwards, it gradually increased to just below

60% for the remaining 5 packs of dried tobacco leaves. With 10 mg of chloramine T on the filter pad, the adsorption efficiency, by the amount adsorbed on the pad, Eq. (1), increased from below 5% to about 50% for all 6 packs of dried tobacco leaves. On the contrary, the adsorption efficiency, by the breakthrough amount, Eq. (2), dropped drastically from above 90% to 70% for the first pack of dried tobacco leaves. Then, it gradually dropped to about 60% for the remaining 5 packs of dried tobacco leaves, in the same manner as that without chloramine-T.

With 10 mg of chloramine-T on the filter pad, only relative small amount of hydrogen cyanide could be adsorbed directly on chloramine-T in comparison with the total amount of hydrogen cyanide produced by consecutive combustion of 6 packs of dried tobacco leaves. Therefore, both the amount of breakthrough hydrogen cyanide and the concentration of breakthrough hydrogen cyanide were quite similar to the results of the adsorption on the filter pad without chloramine-T. The results agreed quite well with the adsorption efficiency, evaluated by the mass balance of the breakthrough amount, Eq.

(2), which was slightly greater than the case without chloramine-T.

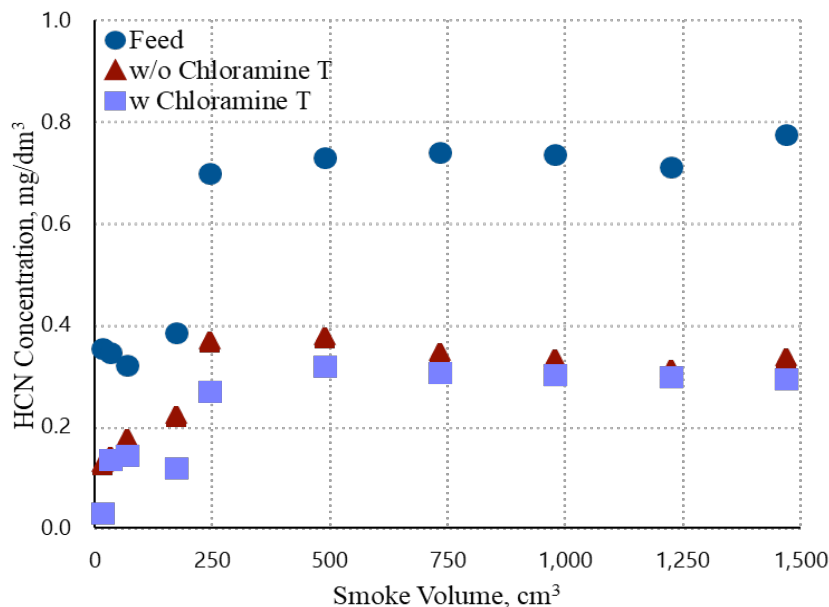


Fig. 3: The comparison of the concentration of breakthrough HCN the filter pad containing 10 mg chloramine-T, as well as without chloramine-T, with the flow rate of 17.5 cm³/s.

3.2 The Effect of Amount of Chloramine-T

As the great variations of hydrogen cyanide concentration and adsorption efficiency were founded for the first pack of dried tobacco leaves, which could produced 245 cm³ of smoke for this study, the effect of chloramine-T amount on the variation of adsorption efficiency was investigated further. The amount of breakthrough hydrogen cyanide dropped from above 0.07 mg to above 0.02 mg as increase in the amount chloramine-T on the filter pad from 3 mg to 45 mg, as shown in Fig.5 (a). While the amount of hydrogen cyanide adsorbed on the filter pad with chloramine-T dropped from about 0.06 mg to just above zero. While, the difference of the amount of breakthrough hydrogen cyanide from the total amount of hydrogen cyanide increased from 0.07-0.08 mg to above 0.14 mg with an increase in the amount of chloramine-T. The contradictory of the reduction of breakthrough amount of HCN and the reduction of amount of HCN adsorbed on the pad was investigated.

On comparison with physical adsorption of HCN on the filter pad containing proline (Sanbudit and Chatsiriwech, 2019), the amount of breakthrough hydrogen cyanide dropped in the same manner

as the results of chloramine-T with the increase in the amount of proline, as shown in Fig.5 (b). While the amount of adsorbed hydrogen cyanide on the filter pad with proline, as well as the difference of the breakthrough amount from the total amount, increased in the same manner as the difference between the total amount hydrogen cyanide and the breakthrough amount of hydrogen cyanide for chloramine-T in Fig.5 (a). The results suggested that HCN was adsorbed on both chloramine-T and filter pad. While HCN, which was adsorbed on chloramine-T with chemical bonds instead of physical one, could not be recovered by dissolving in the sodium hydroxide solution above. Only HCN, adsorbed on the filter pad, could be analysed by dissolving in the sodium hydroxide solution. With the increase in the amount chloramine-T on the filter pad, the breakthrough amount of HCN was decreased in the same manner as that with Proline, under which the adsorption was physisorption. In addition, only a small fraction of HCN was adsorbed on the filter pad with 45 mg chloramine-T.

Consequently, the suitable amount of chloramine-T for chemisorption of HCN in 245 cm³ of smoke was about 45 mg.

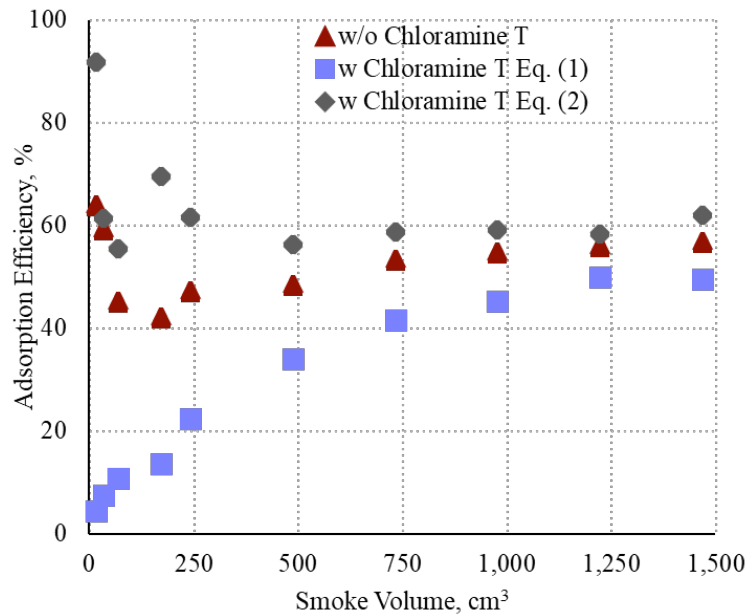


Fig. 4: The adsorption efficiencies calculated by Eq. (1) and Eq. (2) for 10 mg of chloramine-T on the filter pad in comparison with the pad without chloramine-T at the flow rate of 17.5 cm³/s

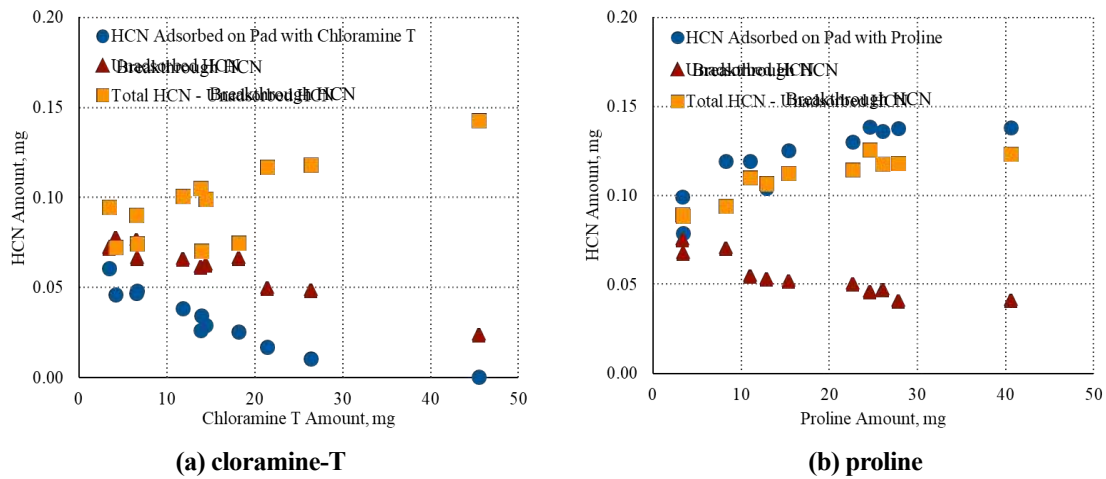


Fig. 5: The amounts of HCN on the pad with chloramine-T (a) and with proline (b), in comparison with the breakthrough amounts and the difference of the breakthrough amount from the total amount of HCN for 245 cm³ of smoke with the flow rate of 17.5 cm³/s.!

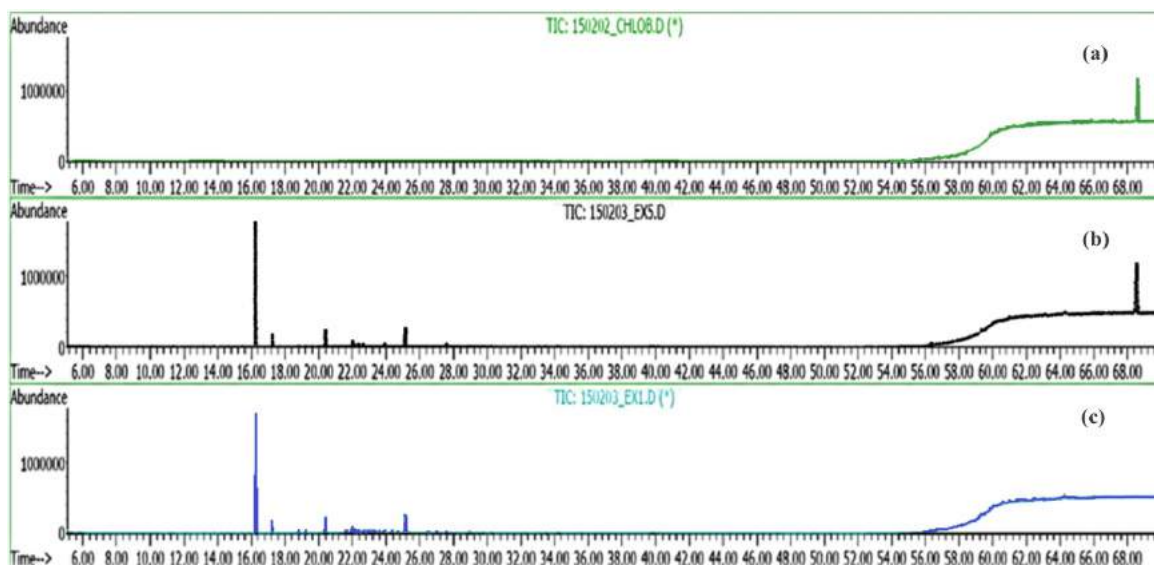


Fig. 6: Chromatograms of compounds on filter pads with chloramine-T before adsorption (a), with chloramine-T after adsorption (b), and without chloramine-T (c).

Additional investigation of chloramine-T on the filter pad after adsorption in comparison with before adsorption was carried out with GC/MS. Before adsorption, only the peak of chloramine-T at 69 min. was found on the chromatogram, as shown in Fig. 6 (a). While Fig. 6 (c) illustrated the chromatograms of some compounds on the filter pad without chloramine-T after adsorption, hence, no peaks revealed at 69 min. Based on the Wiley275 library, the peak at 16.2 min. was identified as nicotine. After adsorption, the chromatograms of similar compounds found on the filter pad containing chloramine-T were similar to those found on the filter pad without

chloramine-T, except the peak at 69 min., as shown in Fig.6 (b). The results revealed that chloramine-T still remained on the filter pad after adsorbing HCN from smoke. For adsorption performance evaluation, the adsorption efficiency was evaluated by the mass balance of the breakthrough amount, Eq. (2), instead. The adsorption efficiency could be improved from 50% to 70% by adding 20-25 mg chloramine-T on the filter pad, as shown in Fig.7. For 245 cm³ of smoke, over 80% adsorption efficiency could be achieved by adding 45 mg of chloramine-T on the filter pad.

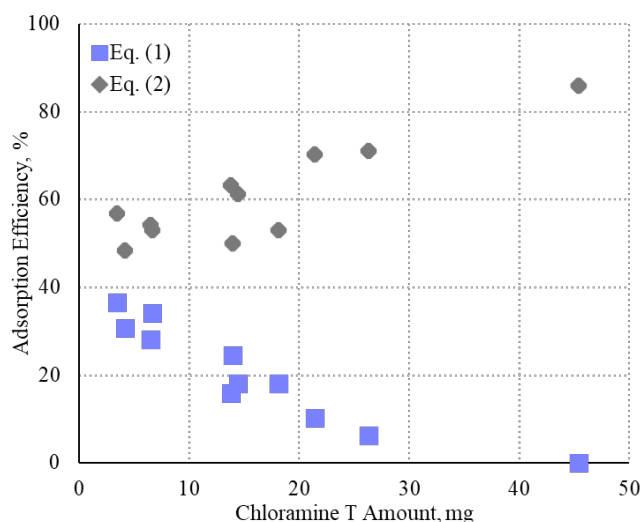


Fig. 7: The effect of the amount of chloramine-T on the adsorption efficiency, evaluated by the adsorbed amounts, Eq. (1), and the breakthrough amounts, Eq. (2), with the flow rate of 17.5 cm³/s.

IV. CONCLUSIONS

The adsorption of hydrogen cyanide on chloramine-T was chemisorption. For the hydrogen cyanide concentration of 0.7-0.8 mg/dm³ and the flow rate of 17.5 cm³/s, the adsorption efficiency could be improved from 50% to over 80% by adding chloramine-T up to 45 mg. While, the concentration of breakthrough hydrogen cyanide could be reduced to 0.3 mg/dm³.

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