THE TEMPERATURE EFFECTS OF THE ADSORPTION OF PHOSGENE ON MILITARY FILTERS

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Introduction

Activated carbon is a good adsorbing material. But depending on the adsorbate, the adsorption can be a physi- or a chemisorption. Organic vapours tend to be physisorbed. A weak reversible interaction between the molecules and the surface takes place. Anorganic vapours are mostly chemisorbed. A chemical bond is formed between the surface and the molecule. Another chemical reaction is possible too: decomposition of the vapour. Physisorption takes place in the pores, in the same way chemisorption occurs at the active sites of the carbon or at the impregnated (metal) complexes.
When a gas, e.g. phosgene, is chemisorbed [1,2], heat will be released because of the exothermic reaction. This results in the warming up of the filter bed. Here, prudence is called for due to the low self combustion temperature of activated carbon, which is sometimes even under 200°C.
The aim of this work is to study the influence of the different parameters on the temperature during the adsorption. These results can be used to draw a conclusion about the safety of the chemisorption process.

Experimental

In breakthrough time measurements, different parameters have an effect on the result: inlet concentration [3], linear velocity of the air flow [4] and (pre)-humidification [5]. The influence of these parameters to test their influence on the temperature has been investigated.
Nine self-made filters are tested with contaminated air (containing phosgene). The filters are filled with a carbon bed of 2 cm; the activated carbon that has been used is Chemviron ASC-TEDA (12x30). This activated carbon is impregnated with Cr, Cu and TEDA (triethylene diamine)-complexes and has a self-combustion temperature of 200-220°C. The temperature is measured with a thermocouple in the centre of the filter at half the height of the carbon bed (See fig. 2).
The values that are used in the tests are:

- inlet concentration (ppm): 5000, 8500; 13 000; 21 000
- flow rate (cm/s): 1.96; 3.93; 9.95 and 19.89
- prehumidification of the filter – humidification of the filter: 0-0 %RH; 0-70 %RH; 70-70 %RH.
- Air flow temperature: 20-23°C

The temperature has been measured during the test until breakthrough.

Results and Discussion

Results

The test parameters and the results are summarized in the following table (See table 1):

↓ variable parameter	C_{in} (ppm)	v (cm/s)	RH (%)	T_{max} (°C)	t_b (min)	Nr
inlet conc. (C_{in})	5232	3.93	0-70	85	23	1
	8600	3.93	0-70	84	23	2
	13066	3.93	0-70	115	15	3
air flow (v)	12700	1.96	0-70	103	37	4
	13066	3.93	0-70	115	15	3
	13407	9.95	0-70	140	18	5
	13038	19.89	0-70	151	6	6
humi-dity (RH)	21177	1.96	0-0	93	23	7
	21282	1.96	0-70	133	22	8
	20386	1.96	70-70	64	24	9

Table 1: Results from the tests.

The temperature evolution shows the same curve for all the tests during the chemisorption of phosgene at the carbon surface (See fig. 1).

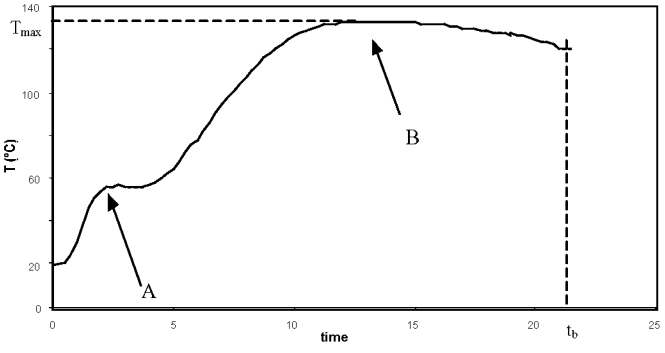


Fig. 1: Temperature evolution of a filter during the test

For every test, the curve forms a plateau at ± 50°C (Fig. 1 point A) at the beginning of the test. This is due to the heat originated from the heat front at the beginning of the carbon bed (See fig. 2).

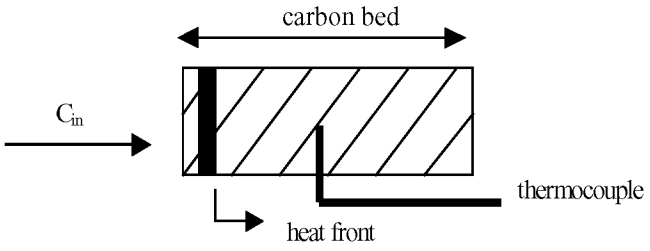


Fig. 2: Heat front at the beginning of the test

Diffusion of the reaction heat warms up the whole filter. The heat front moves through the filter bed. The temperature (after the plateau) rises when the heat front approaches the thermocouple until it reaches its maximum temperature (Fig. 1, point B), when the heat front has reached the thermocouple. After this maximum, the filter cools down because of the air flow.

Discussion

- The influence of the inlet concentration:

  From the data (See table 1: test 1, 3 and 4, 8; also see fig. 3) can be concluded that a higher inlet concentration results in a higher maximum temperature. This shows that the reaction between the active sites and the phosgene molecules occurs immediately when they get in contact with each other. A higher concentration means a higher molecule rate through the carbon bed. As a consequence, more molecules will be adsorbed and more reaction heat will be released to warm up the filter.

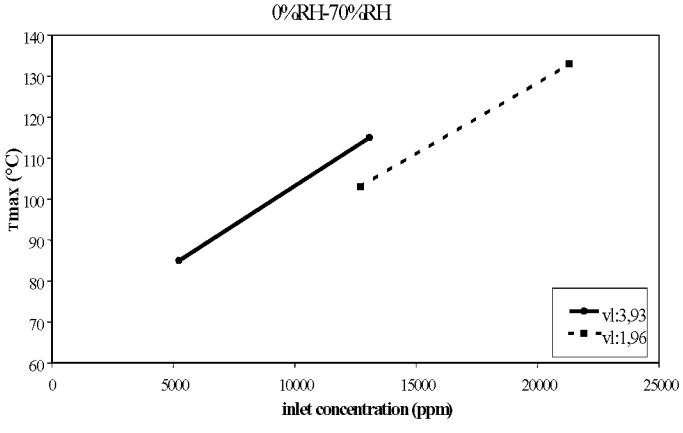


Fig. 3: Influence of the inlet concentration at T_{max}.

- The influence of the humidity:

  Humidity can play an important role for the capacity of the filter. For organic vapours, breakthrough time shortens when the activated carbon is preloaded with humid air [5]. On the contrary, a comparison of the breakthrough times of phosgene for humid and dry air, shows no significant difference (See fig. 4) [2].

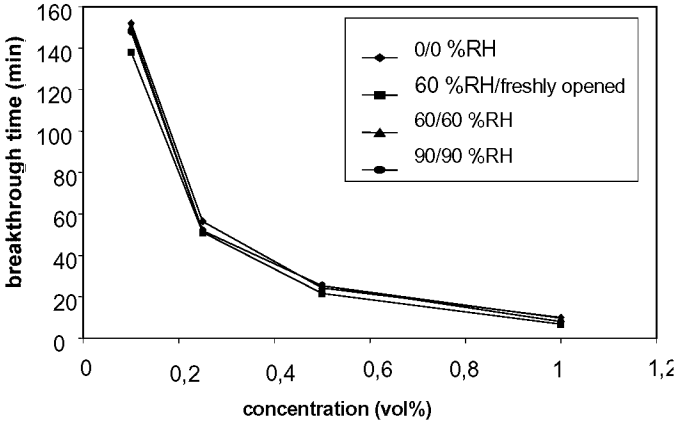
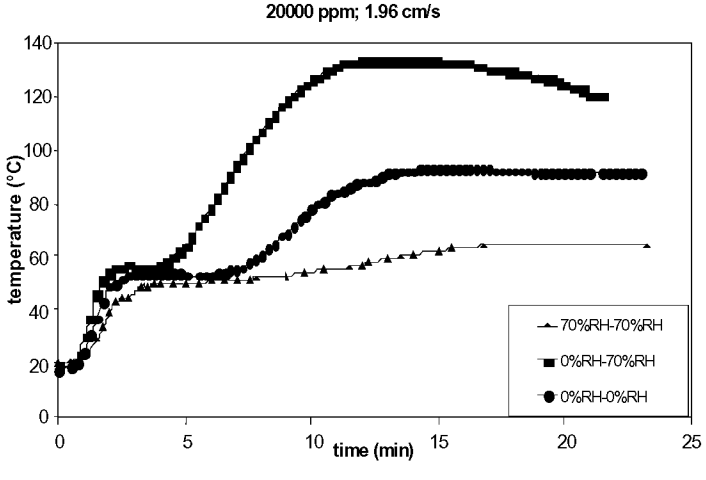


Fig. 4: Influence of air stream humidity/pre-wetting of the filter on the breakthrough curve.

But for the influence of the humidity on the temperature, the results (See fig. 5, See also table 1; test 8-9) show that T_{max} with prehumidification is significantly smaller than without. It is even smaller than T_{max} of test 4 (See table 1) with nearly half the inlet concentration. A possible explanation is the loss



of weight of the filter after the test. The reaction heat is used to evaporate the pre-adsorbed water. Less heat is left over to warm up the air stream.
Fig. 5: Influence of the prehumidification/humidification of the filter.

The maximum temperature of a filter test with no prehumidification and with dry air lies between the T_{max} of the filter with prehumdification and the filter without prehumidification (both are measured with humid air) (See table 1: tests 7-9; see also fig. 5). But, as yet, no plausible explanation has been found.

- <u>The influence of the linear velocity of the air flow</u>

Comparing tests with a different linear velocity shows that a higher linear velocity results in a shorter breakthrough time. When looking at the graphic (See fig. 6; see also tests 3,4 and 6), the patern is similar for all the linear velocities. But for a higher linear velocity, the trend that can be seen is: a shorter plateau, i.e. a steeper raise to T_{max}, after which the carbon bed will cool down faster .

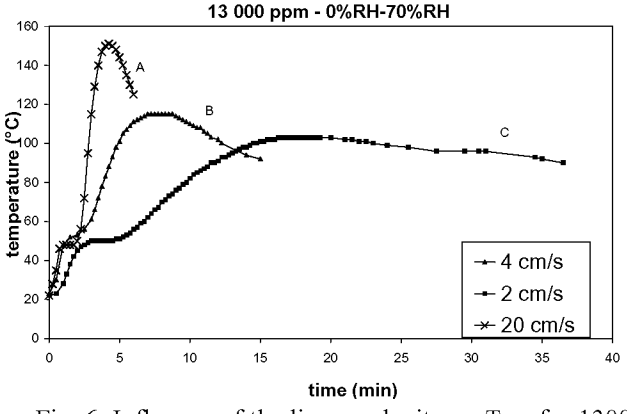


Fig. 6: Influence of the linear velocity on T_{max} for 13000 ppm.

The same contaminated air volume is blown through the filter at a higher rate, which fastens the whole process. The total heat during the test is the same, which results in a higher T_{max}.

When the maximum temperature is plotted against the linear velocity (See fig. 7; see alos tests 3 → 6), the curve tends to converge to a constant temperature for very high linear velocities.

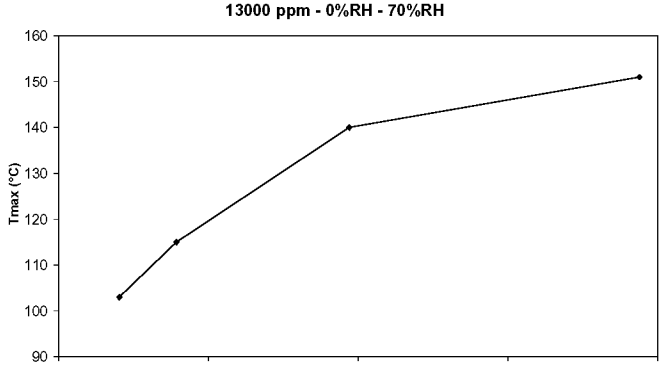


Fig. 7: Raise of temperature as a function of the linear velocity.

Conclusions

Three important parameters in measuring breakthrough time have been examined in order to investigate their effect on the heating of the activated carbon during the chemisorption of phosgene.

- The change of inlet concentration results in a change in maximum temperature. But this influence on T_{max} is not as substantial as the influence of respectively humidity and airflow.
- When dry air is used, T_{max} is lower than with humid air. But, when the filter is pre-wetted, the heat is used to evaporate the water, which results in a T_{max} that is even lower than that of the test with dry air.
- Linear velocity has a substantial effect on the temperature. However at very high values of the air flow, T_{max} seems to converge to a constant value.

As a remark can be said that when the activated carbon, Chemviron ASC-TEDA, was used, the temperature during the test stayed low enough with a sufficient safety margin to exclude self-combustion of the filter.

References

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