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# REGENERATION OF HIGH SILICA ZEOLITES VIA ADVANCED OXIDATION PROCESSES—A PRELIMINARY STUDY ABOUT ADSORBENT REACTIVITY TOWARD OZONE

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Aiming at regenerating adsorbents, the reactivity toward ozone of two high silica zeolites (HSZ), a dealuminated faujasite Y (Fau Y) and a silicalite (Sil Z), was investigated. In case of Fau Y, no physical adsorption occurred but a total degradation of ozone. This phenomenon was attributed to an active sites-role of silanol groups (SiOH), mainly developed by the dealumination step. In contrast, the ozone adsorption in the smaller channels of Sil Z was important (about 17 mmol g<sup>-1</sup>), but when a certain local concentration of ozone was reached, the ozone degradation became significant and increased up to 100%. The ozonated zeolites were regenerated in an oven. Whereas the regeneration was total at 773K, a treatment at 500K seemed insufficient since a co-adsorption phenomenon was observed with suspected nitrous oxides (NO<sub>2</sub>) as a product of ambient nitrogen oxidation. Whereas the Fau Y was not affected at all by ozonation, the adsorption capacities of Sil Z zeolite were enhanced, and it exhibited a higher selectivity for polar compound. This was attributed to a surface modification, by highly reactive species generated during molecular ozone decomposition.

*Keywords:* high silica zeolites; adsorption; regeneration; advanced oxidation processes (AOP); ozone; catalytic decomposition.

## INTRODUCTION

In the treatment of industrial polluted air streams, adsorption on a porous media, such as activated carbon or hydrophobic zeolites, is commonly adopted (Le Cloirec, 1998). Instead of being landfilled and replaced, activated carbon is mainly regenerated via pressure or thermal desorption, aiming at restoring the adsorption activity with as little damage as possible to the original pore structure. Advanced oxidation processes (AOP) were investigated as an alternative regeneration method for activated carbon (Masuda *et al.*, 2001), highlighting its scavenger effect toward ozone (Dusenbury and Canon, 1996), but also in case of zeolites (Leichsenring *et al.*, 1996a; Zlotopol'sky and Smolenskaya, 1996), notably when coked in catalytic petrochemistry (Querini, 2000; Mariey *et al.*, 1996).

In parallel, AOP are investigated as direct technologies for water or gas streams treatment (Shen and Ku, 1999) and, interestingly, adsorbents can provide a good environment for oxidation reaction (Vidic *et al.*, 1997), as shown in previous studies of zeolite regeneration by photocatalysis (Monneyron *et al.*, 2003a) or V-UV radiation combining the effect of photolysis and the creation of ozone from ambient oxygen (Monneyron *et al.*, 2003b).

To use ozonation as a regeneration technique for organic compounds adsorbed on high silica zeolites (HSZ), it is

essential to establish the interaction between ozone and the adsorbent. Moreover, as shown by Chiang *et al.* (2002) for activated carbon, any modification of adsorption properties of the solid is to be determined.

## METHODS AND MATERIALS

### Adsorbents

Two commercial HSZ from TOSOH Corp. were used in this work, a dealuminated faujasite Y (Fichtner-Schmittler *et al.*, 1990) and a silicalite (Olson *et al.*, 1980). The main characteristics of adsorbents are given in Table 1. Prior to experimentation, the zeolites were heated at 230°C (or 500°C when specifically mentioned) overnight, and kept in a desiccator to prevent ambient humidity to interfere with experiments.

### Ozonation Experimental Setup

A commercial ozone generator from air (model 5LO from Trailgaz) was used here, and since the ozone production is function of air pressure ( $p = 0.7$  bar), flow ( $F = 0.25$  m<sup>3</sup> h<sup>-1</sup>), and power applied,  $P$  (W), the ozone inlet concentration was reproduced with an uncertainty of  $\pm 0.5$  g m<sup>-3</sup>. The reactor, regulated in temperature at 298K with an external water flow

Table 1. Main characteristics of zeolites.

|   | HSZ trade name    |               |
|---|-------------------|---------------|
|   | HSZ-390HUD3C      | HSZ-890HOD1C  |
| Type (symbol)   | Faujasite (Fau Y) | ZSM-5 (Sil Z) |
| SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> (mol mol <sup>-1</sup> ) | 360               | 1880          |
| Na <sub>2</sub> O (wt%)   | 0.02              | <0.01         |
| Clay binder content (%)   | 25                | 20            |
| Bulk density (kg l <sup>-1</sup> )  | 0.42              | 0.66          |
| Pellet diameter (mm)  | 3.0               | 1.5           |

circulating in a double glass wall, was 20 cm high and 8 cm in internal diameter. At the inlet, outlet and each 5 cm, an intermediate gaseous sample could be taken and ozone concentration determined with a BMT 964 Ozone Analyser. At these five points, the temperature was followed at the same time, as can be seen in Figure 1. Whereas the inlet and outlet enabled the gas temperature to be followed, the inner-reactor measurements reported the solid pellets surface temperatures.

### Adsorption Equilibria Measurements

The investigation of ozonation influence on adsorption capacity of HSZ toward model VOCs was performed using batch experiments via a standard volumetric method described elsewhere (Monneyron *et al.*, 2003c).

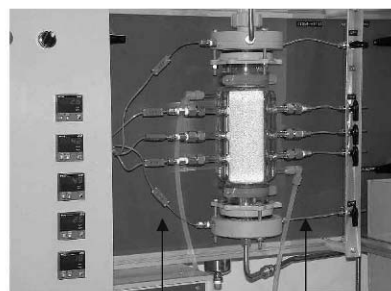
In the same way, the selectivity of adsorbent ( $\eta_{1/2}$ ), calculated using equation (1), where  $X$  and  $Y$  are respectively the adsorbed and gas phase molar fraction, was determined in competitive adsorption experiments (Monneyron *et al.*, 2003c).

$$\eta_{1/2} = \frac{X_1 \cdot Y_2}{X_2 \cdot Y_1} \quad (1)$$

## RESULTS AND ANALYSIS

### Ozonated Air Flow on HSZ Fixed Bed

In this set of experiments, the inlet concentrations ( $C_{IN}$ ) of ozone were equal to 9.5, 18.5, and 26.5 g m<sup>-3</sup> respectively for  $P=50$ , 100 and 150 W. The mass of adsorbent was adjusted to practical requirements. The results obtained on the activated Fau Y exhibited two different parts, the first being an instantaneous breakthrough of ozone up



Temperature measurements Gas phase sampler set up

Figure 1. Adsorption/ozonation experimental setup.

to  $C_{OUT} = 65\% \cdot C_{IN}$ , immediately followed by a decrease of the concentration, as shown in Figure 2. After reaching a minimum, the ozone concentration increased to  $C_{OUT} = C_{IN}$ .

This evolution does not involved any physical adsorption phenomenon, but it has rather to be seen as an evolution of ozone decomposition on the Fau Y surface. There have been very few studies on ozone decomposition mechanisms but, as reported by Tsyganenko *et al.* (Mariey *et al.*, 1997; Tsyganenko *et al.*, 2001), the molecular ozone exhibits some Lewis basic characteristics, notably on silanol groups (SiOH), that were reported by Dhandapani and Oyama (1997) to be well-known catalytic sites for ozone decomposition in indoor air treatment patents. The supplier of the commercial Fau Y used in this study kept undisclosed the chemical nature of the clay binder together with details of dealumination technique. But whatever conditions may have been applied to obtain the high Si/Al ratio, a significant number of SiOH groups were formed, typically 10/100 framework atoms (Meininghaus and Prins, 2000; Kawai and Tsutsumi, 1998), and are suspected to be responsible for the initiation of the decomposition complex mechanism of ozone on Fau Y.

After the regeneration step, the Fau Y had the same global behavior toward gaseous ozone, except that the decomposition was total from the very beginning of the experiment. The sampling of temperature at different points of the adsorbent bed (5, 10 and 15 cm) enabled us to explain the two different behaviors observed until 400 min. Whereas during the second use of Fau Y sample, the full decomposition of ozone was taking place almost completely at the beginning of the bed, shifting to the higher parts with time, in the first experiment the reactions took place in the whole bed and much more slowly, i.e. with a smaller heat emission notably visible at the 5 cm- $\Delta T$  measurement. It is therefore suspected that the contact with ozone increased the number of decomposition active sites by modifying the surface. The ozone decomposition slowly ended, the previously mentioned sites being deactivated with time.

The observed behaviour of Sil Z toward ozonation was very dissimilar from Fau Y. As shown in Figure 3 for the ( $P=50$  W)-experiment, a classical adsorption breakthrough curve was obtained. Moreover, it is clear that physical adsorption of ozone took place on Sil Z since the breakthrough time decreased, and its slope was higher as the ozone concentration increased.

In our case, the maximum outlet concentration was dependent on the inlet concentration ( $C_{OUT,max}/C_{IN} = 100, 75$  and 20%, respectively, for  $P=50, 100$  and 150 W), meaning that some ozone was also decomposed. Whereas for the lowest concentration, the outlet concentration was stable at ( $C_{OUT} = C_{IN}$ ), on the second part of the other curves, the ozone concentration decreased to a steady state of 100% decomposition.

The temperature profiles, shown in Figure 4, obtained during these experiments are another indicator of the two different phenomena happening consecutively. Indeed in the two experiments where ozone decomposition occurred, for  $P=100$  and 150 W, the temperature elevations were quite different from the  $P=50$  W where only adsorption occurred. Indeed, the second temperature increase of  $P=100$  and  $P=150$  W experiments did not exhibit the same behaviour, the respective slopes being 1.8 and 1.3 K m<sup>3</sup> g<sub>O<sub>3</sub></sub><sup>-1</sup> for adsorption and decomposition phases.

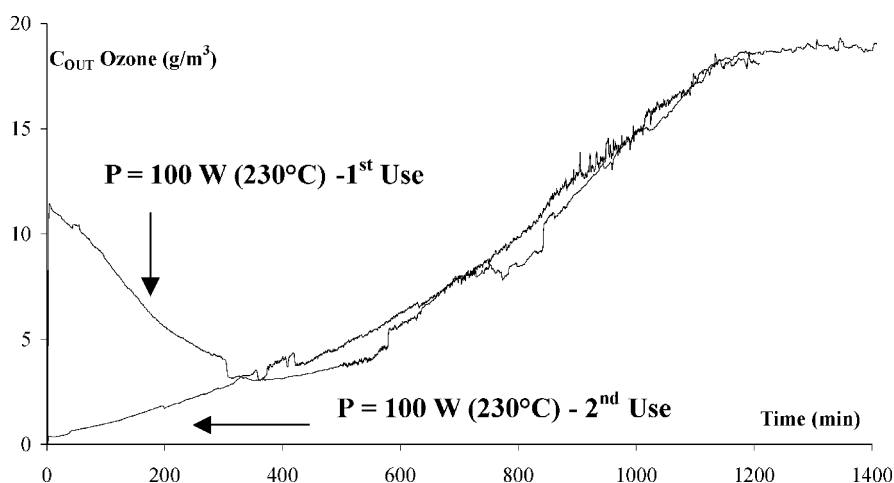


Figure 2. Ozonated air flow on Fau Y (400g-20cm).

As a quick approximation of the adsorption capacity of Sil Z toward ozone, the linear part of the breakthrough was used to estimate  $t_{1/2}$  when ( $C_{OUT} = C_{IN}/2$ ), directly or by extrapolation, assuming that at the beginning of the

breakthrough no ozone decomposition occurred. The values, obtained for  $P = 50$  and  $100$  W, are quite consistent with each other ( $16$  and  $17.5$   $\text{mmol g}_{\text{SilZ}}^{-1}$ ), whereas for  $P = 150$  W, the adsorbed quantity seems overestimated ( $24$   $\text{mmol g}_{\text{SilZ}}^{-1}$ ). This is not surprising since, in the last case, the linear part of the curve is very short ( $C_{OUT,max} C_{IN} = 20\%$ ) so that the ozone decomposition could not be neglected. This surprising difference between the two HSZ was reported also in the aqueous phase by Sakoda *et al.* (2001), leading to a very different aqueous ozone enrichment factor (1:100). In the same way, Izumi *et al.* (2001), investigated the gaseous ozone enrichment on HSZ at low temperatures (down to  $213$  K) where no ozone decomposition occurred, but the difference in adsorption capacities of both zeolites was much less important. These results tend to show that the decomposition of ozone requires a minimum local concentration, the kinetic of decomposition depending notably on the ozone concentration, supplied by its physisorption in the framework channels of Sil Z. This decomposition is therefore much slower than on Fau Y, which is consistent with the

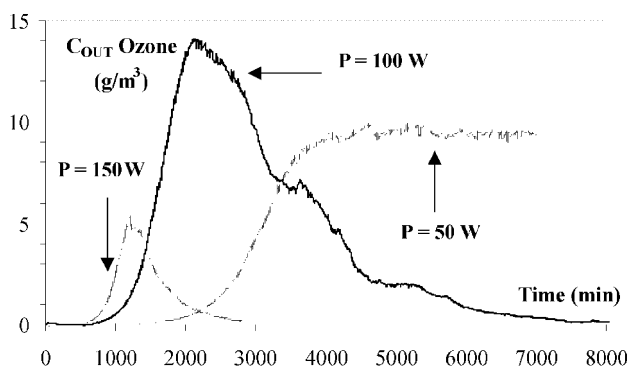


Figure 3. Ozonated air flow on Sil Z (200g-6cm).

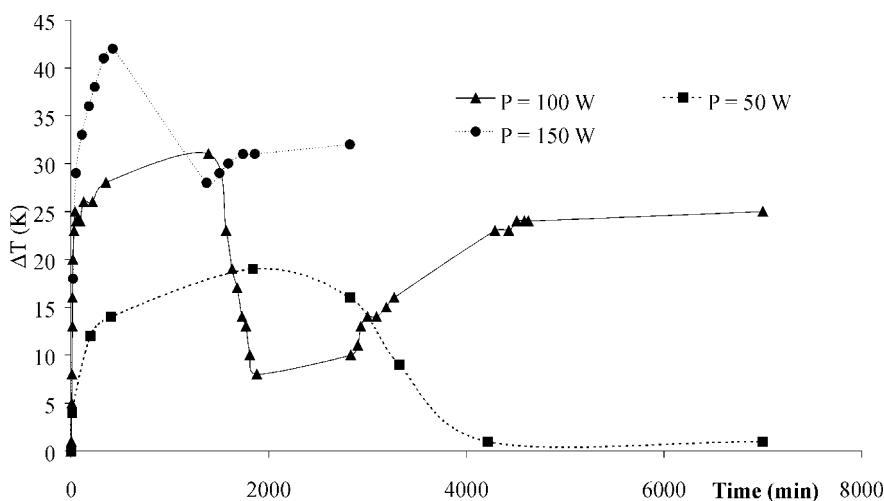


Figure 4. Temperature profile during ozone treatment of Sil Z.

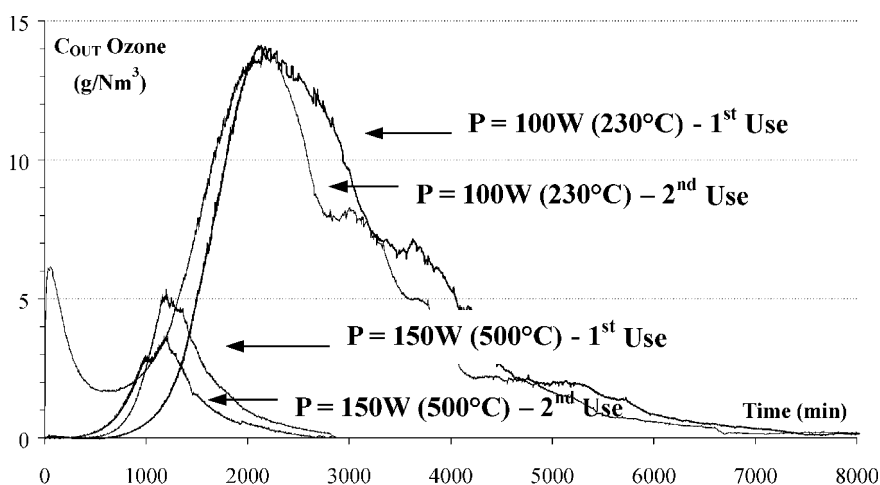


Figure 5. Oven temperature influence on Sil Z regeneration after ozone treatment.

hypothesis of a determining role of silanol groups as active sites, the Sil Z having undergone no dealumination treatment. It may be supposed that in this case the decomposition was initiated by the hydroxyls groups of the clay binder (Alpay *et al.*, 1996) or by local heterogeneity of the zeolite framework. Then, the ozone decomposition involving very reactive species (Dhandapani and Oyama, 1997), some complex radical chain reactions would have led to a slow spreading into the whole bed, down to a total decomposition of the inlet ozone.

#### Regeneration of Sil Z After Ozonation

As shown in Figure 5 by the two experiments conducted for  $P = 150\text{ W}$  after a  $500^\circ\text{C}$  overnight heat treatment, the regeneration of the Sil Z sample was total, whereas in a ( $P = 100\text{ W}$ )-set of experiments, when the oven temperature was generally fixed to  $230^\circ\text{C}$  only, the regeneration was insufficient as an initial partial breakthrough of ozone occurred. However, in both cases, the ozone global physical adsorption capacity decreased, indicating a modification of

zeolite internal surface. Considering these results and as mentioned in the introduction of this paper, it was absolutely necessary to check the adsorption properties of the oxidised zeolites.

#### Adsorption Properties of Ozonated HSZ

Adsorption equilibria measurements showed that the contact of ozonated air with Fau Y (3000 min) had no effect at all on its adsorption capacities in single component isotherms, and on its selectivity toward VOC binary mixtures. On the contrary, the Sil Z zeolite was significantly affected by the ozone treatment (9000 min), as shown by Figures 6 and 7.

Indeed, the adsorption capacity of Sil Z toward both components increased after ozonation, which is to be linked with the hypothesis of a surface modification. This augmentation was much greater and earlier, in terms of minimum concentration, for TOL (+25%, from  $C_e = 0.001\text{ mmol l}^{-1}$ ) than for DIO (+8%, from  $C_e = 0.007\text{ mmol l}^{-1}$ ). This could be attributed to the difference of their micropore fillings (52 and 87%, respectively, for TOL and DIO; Monneyron

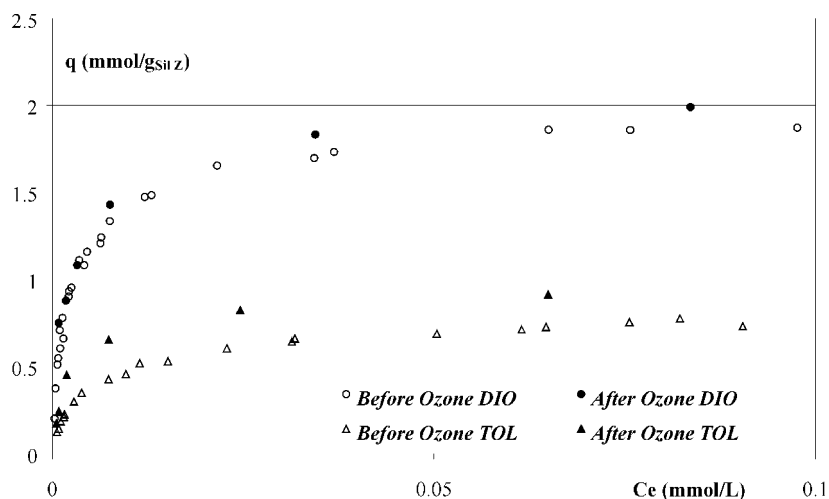


Figure 6. Ozone treatment influence on adsorption capacity of Sil Z toward 1,4-dioxan (DIO) or toluene (TOL).

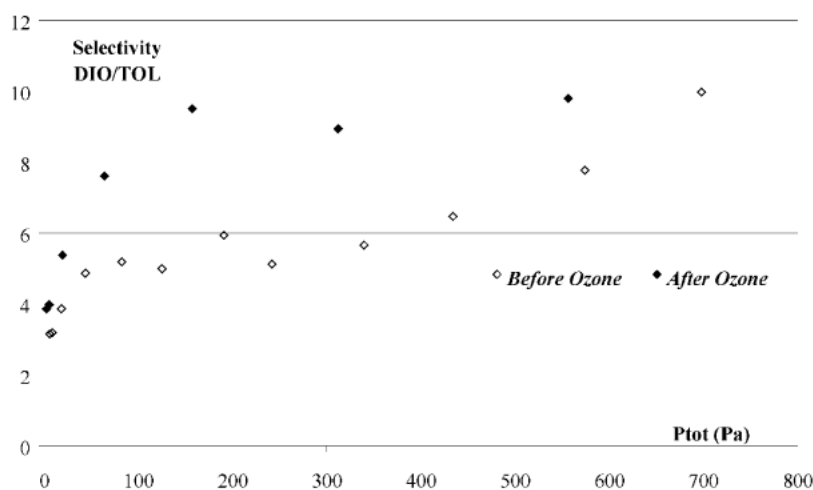


Figure 7. Ozone treatment influence on Sil Z selectivity in a DIO/TOL 50:50 initial molar mixture.

*et al.*, 2003c). Consecutively to the oxidation phase, some strong specific chemisorption bonds were induced, presented by Rochester and Stachan (1996) in the case of DIO, and  $\pi$ -bonds being notable with the aromatic cycle of TOL.

However, as expected, in a competitive adsorption equilibria determination, the Sil Z showed a much higher selectivity for the polar component (DIO), as shown in Figure 7.

## DISCUSSION AND CONCLUSIONS

Regarding the initial breakthrough observed for the second use of Sil Z after a low temperature regeneration (230°C), this significant difference with the first experiment was attributed to a competitive adsorption of ozone with another compound. Indeed, a similar behaviour was observed as a ( $P = 150$  W)-experiment was carried out on a non-activated Sil Z, which had not undergone the preliminary heat treatment prior to any experiment, showing an immediate breakthrough of ozone, up to 92% of  $C_{IN}$ , before a slow decrease began. In this case, the co-adsorbate of ozone was the water molecules previously adsorbed from ambient air humidity (50% RH). Moreover, during the ozone treatment of Sil Z at  $P = 100$  W, a diluted brown vapour, characteristic of nitrous oxides ( $NO_2$ ), was observed in the reactor during the final total ozone decomposition. In the same way, after the ( $P = 50$  W)-experiment, a heavy  $NO_x$  smoke was coming out of the zeolites when introduced into the oven regulated at 230°C. These gaseous emissions were therefore attributed to the reaction between the ambient air nitrogen and the highly reactive species generated consecutively to the intense decomposition of the large amount of adsorbed ozone, leading probably to  $NO_x$  adsorbed molecules partially desorbed during the 230°C overnight regeneration.

It is also worth mentioning that, during this ozone decomposition, radicals are formed and, as reported by Leichsenring *et al.* (1996b), about ozonation of de-aluminated Y-zeolites loaded with polychlorinated dioxin and furan, the active oxidative species were hydroxyl radicals ( $OH\cdot$ ) formed on the zeolite surface, notably when the effluent humidity was increased. Single adsorbed-component TOL oxidation by ozone was performed successfully on both zeolites. Further

analysis of these results and of binary selective oxidation will be presented in a future paper.

## NOMENCLATURE

|           |  |
|-----------|--|
| Ce        | equilibrium concentration in the gas phase, $mmol\ l^{-1}$ |
| $C_{IN}$  | ozone inlet concentration, $g\ m^{-3}$                     |
| $C_{OUT}$ | ozone outlet concentration, $g\ m^{-3}$                    |
| F         | ozonated air flow, $m^3\ h^{-1}$                           |
| p         | pressure, bar  |
| $P_{tot}$ | total pressure in the gas phase, Pa                        |
| P         | ozone generator operating power, W                         |
| q         | VOC adsorbed quantity, $mmol\ g^{-1}$                      |
| X         | adsorbed phase molar fraction                              |
| Y         | gas phase molar fraction                                   |

### Greek symbols

|              |  |
|--------------|--|
| $\Delta T$   | temperature uptake in the reactor at a given height: difference between initial and during ozone loading temperatures, K |
| $\eta_{1/2}$ | selectivity of adsorbent for component 1 vs. component 2   |

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