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Surface chemistry of a viscose-based activated carbon cloth modified by treatment with ammonia and steam

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Abstract

The influence of ammonia treatment at 800 °C on the catalytic activity of a viscose-based activated carbon cloth (ACC) was evaluated for the oxidative retention of H₂S or SO₂ at room temperature. Change in the surface chemistry was observed by X-ray spectroscopy of nitrogen (N1s) and by temperature programmed desorption (TPD). Dynamic adsorption of H₂S or SO₂ in moist air onto a packed bed of activated carbon cloth was monitored by measurement of the breakthrough curves at room temperature. ACC modified by ammonia showed noteworthy enhanced SO₂ and H₂S loading relative to the untreated ACC. Improved SO₂ retention rate could be replicated several times after regeneration by washing at room temperature, in contrast to the case with H₂S. The likely reasons for the behavior of H₂S and SO₂ on the ammonia-treated ACC are discussed with reference to the recent literature.

Keywords: A. Activated carbon, Carbon cloth; B. Surface treatment; D. Catalytic properties, Surface properties

1. Introduction

A number of groups have investigated the surface chemistry of N-containing activated carbons as catalysts or catalyst supports for the low temperature oxidation of pollutants in gas or water media. Stohr et al. [1] have studied the introduction of nitrogen into granulated activated carbon and HOPG graphite powder by ammonia treatment in dry inert atmosphere at 700–900 °C. Nitrogen was chemisorbed in the treatment and XPS showed two N1s signals with binding energies at 401–400 and 399–398 eV. When ammonia treatment was performed at mild temperature, the peak at high binding energy (amine groups) was intense compared with the peak at low binding energy (pyridinic groups). At 700 °C, the volume of micro- and mesopores was unchanged after reaction, while a significant increase of the pore volume in the micropore range and of the apparent surface area was observed in the reaction with ammonia at or above 800 °C. Chemisorbed nitrogen species on the surface of NH₃-activated carbon, which would facilitate the dissociative chemisorption of O₂ as superoxide ions, enhanced the surface oxidation of probe molecules such as H₂ or aqueous H₂SO₃ to H₂O or H₂SO₄, respectively. The catalytic activities were well reproducible after the NH₃-activated carbon ageing in air during long-term storage.

Recently, ammonia treatment of activated carbons at high temperature has been used to introduce a high density of surface nitrogen groups on pitch-based and resin-based activated carbon fibers (ACF) to optimize the oxidative retention of up to 5000 ppmv of SO₂ in moist air at 20–150°C and atmospheric pressure [2–5]. In spite of a high cost for preparation, several important advantages are imparted to ACFs. Their small external diameters and uniform micropore structure, directly open to the outer surface, confer faster adsorption/desorption kinetics and a lower pressure drop as compared to granular activated carbons. Their fibrous shape permits their use in varieties of easy handling. The woven and knitted forms allow effective and rapid electrical thermal conditioning. ACFs are more suitable for the recovery of sulfuric acid at room temperature than granular activated carbon because the diffusion of sulfuric acid is much easier in the pores of the fiber [6–10]. However, in the case of ACF modified with ammonia and steam, a part of SO₂ would remain non-
desorbable by distilled water extraction at ambient temperature [11].

Within the above scope, the present paper focuses on the ammonia modification of a commercial viscose-based activated carbon cloth (ACC) for the oxidative retention of H₂S or SO₂ in moist air at room temperature. It combines XPS and XANES to determine nitrogen and sulfur functionalities, TPD to examine the stability of nitrogen and oxygen surface functional groups and adsorption studies to investigate the performance of the adsorbents for the oxidative retention of SO₂ and H₂S.

2. Experimental

2.1. Samples

The commercial activated carbon cloth used in this study was provided by Actitex (Ref.: WKL20). The sample was selected among a set of Actitex activated carbon cloths for its good mechanical properties and high resistance towards air oxidation at 300 °C. It was prepared from a viscose rayon cloth (precursor) using a classified method which involves carbonization followed by CO₂ activation at 700 °C. For convenience, the commercial Actitex sample was designated by the code ACC.

In this study, ammonia-steam was preferred to pure ammonia because ammonia treatment using an aqueous solution of ammonia is more straightforward. Yet, this experimental option is not optimal because addition of steam increases pore development and thus micropore widening [12]. Ammonia-steam activation was performed by placing a 0.5–1-g sample in a quartz tube under flowing nitrogen (100 ml min⁻¹) at 800 °C during 40 min. After treatment, the sample was cooled down in a stream of N₂. The loss of weight during this process was 13 wt% daf. The obtained sample was designated by the code ACCN.

2.2. Porosity values

The adsorption measurements [13–15] were carried out by a static technique in a high vacuum gravimetric apparatus (McBain balances). Micropore half-width (σ) of treated and untreated ACC was calculated from the experimental isotherms using the Dubinin–Radushkevich (DR) equation.

2.3. TPD –MS

The sample (5–10 mg) was thermal-programmed in a static technique in a high vacuum gravimetric apparatus (McBain balances). Micropore half-width (σ) of treated and untreated ACC was calculated from the experimental isotherms using the Dubinin–Radushkevich (DR) equation.

2.4. XPS

X-ray photoelectron spectra were measured using a Surface Science Instrument SSX 100 spectrometer with a monochromated Al Kα X-ray source (hν = 1486.6 eV), a hemispherical electron analyzer, and a multichannel detector. Carbon fibers were placed in an especially designed support ensuring that no signal was observed from the sample holder. Before each XPS analysis the samples were dried under vacuum at 80 °C. Samples were then outgassed in the pretreatment chamber at 10⁻⁵ Torr prior to transfer to the analysis chamber. The vacuum applied during the measurements was typically less than 5 x 10⁻⁵ Torr and data were accumulated in separate regions. The C1s peak of the “graphitic” carbon (binding energy, BE = 284.6 eV) was taken as reference in calculating BEs and accounting for charging effects. Data processing was achieved with a Winspec software using a nonlinear, least-squares fitting algorithm and a Shirley baseline. High resolution envelopes were fitted using mixed Lorentzian–Gaussian curves. For all elements, atomic concentrations were estimated based on comparisons of integrated peak intensities normalized by the atomic sensitivity factors. BEs were reproducible to within 0.2 eV.

2.5. XANES

Nitrogen K-edge measurements were performed at the Laboratoire pour l’Utilisation du Rayonnement Electromagnétique (LURE, Orsay, France) on the VUV Super-Aco storage ring. They were carried out on the Sacemor system connected to the SA72 beam line equipped with a high-energy TGM monochromator (resolution of about 0.2 eV at the N K edge). The spectra were recorded in total electron-yield detection, by dividing the signal by that normalized from the fluctuations in the X-ray intensity of a copper grid freshly coated with gold, simultaneously recorded. They were recorded at normal X-ray incidence (E parallel to the surface). The samples were ground to a very fine particle size, dispersed in acetone, and applied dropwise to high-purity tungsten plates, and the solvent was allowed to evaporate. All measurements were made at room temperature under ultra high vacuum (10⁻¹⁰ Torr). Calibration of the monochromator at the N1s edge was made setting the N1s⁎ (CN) of polyacrylonitrile (PAN, Aldrich) at 401.4 eV.
Fixed bed experiments

Sulfur removal was carried out in a fixed bed contained in a micro-column (I.D.: 4 mm) surrounded by a tube furnace. The air stream flowing through the fixed bed (inlet at the lower end of the vertical micro-column) contained HS (5000 ppmv) or SO\textsubscript{2} (3000 ppmv) with 75\% water saturation at 25°C. The mass of ACC and the total flow rate was about 20 mg and 35 ml min\textsuperscript{−1}, respectively (contact time in weight of sample per gas flow rate was about 0.4 mg min\textsuperscript{−1} ml\textsuperscript{−1} and average residence time in the bed: 0.1 s). The HS or SO\textsubscript{2} concentrations in the inlet (C) and outlet (C\textsubscript{act}) gases were continuously detected with a flame photometric detector (FPD). After a protocol recommended by Bagreev et al. [16], before each adsorption experiment, the samples were prehumidified for 2 h in flowing air with 75\% humidity at room temperature. Adsorption experiments were carried out at room temperature (25°C) and continued until the HS or SO\textsubscript{2} concentration in the effluent gas (C) equalled that in the feed stream (C\textsubscript{feed}). The first adsorption experiment was performed with the sample as received (without any conditioning). After HS adsorption, regeneration of the bed was carried out by heating for 10 min in flowing N\textsubscript{2} at 500°C. After SO\textsubscript{2} adsorption, regeneration was done by washing in a solution of 28\% ammonia in water at room temperature and drying by pressing and flushing with dry helium. Four subsequent adsorption−regeneration experiments were carried out to determine how completely the capacity of the bed could be restored following the regeneration step. The amount of sulfur retained by the active carbon was computed by integration of the breakthrough curve until the breakthrough point corresponding to a de-SO\textsubscript{2} ratio \((C/C)\) of about 0.005. Retention rate (loading) was expressed in mg of SO\textsubscript{2} or H\textsubscript{2}S per g of dry sample. The relationship between intensity of photon emission from sulfur \((i)\) detected by FPD and the amount of sulfur \((M)\) is given by \(i \times M^n\), where \(n\) is close to 1 with experimental conditions used in this study (high S concentration, low hydrogen flow rate). After desorption under vacuum at 80°C of gases physically adsorbed, the amount of thermal-desorbed SO\textsubscript{2} or H\textsubscript{2}S was determined on the loaded sample for (loading) was expressed in mg of SO\textsubscript{2} or H\textsubscript{2}S per g of dry sample. The relationship between intensity of photon emission from sulfur \((i)\) detected by FPD and the amount of sulfur \((M)\) is given by \(i \times M^n\), where \(n\) is close to 1 with experimental conditions used in this study (high S concentration, low hydrogen flow rate). After desorption under vacuum at 80°C of gases physically adsorbed, the amount of thermal-desorbed SO\textsubscript{2} or H\textsubscript{2}S was determined on the loaded sample for (loading) was expressed in mg of SO\textsubscript{2} or H\textsubscript{2}S per g of dry sample. The relationship between intensity of photon emission from sulfur \((i)\) detected by FPD and the amount of sulfur \((M)\) is given by \(i \times M^n\), where \(n\) is close to 1 with experimental conditions used in this study (high S concentration, low hydrogen flow rate). After desorption under vacuum at 80°C of gases physically adsorbed, the amount of thermal-desorbed SO\textsubscript{2} or H\textsubscript{2}S was determined on the loaded sample

\[
\text{Retention rate} = \frac{i \times M^n}{i \times M_{\text{total}}} = \frac{\text{loading}}{\text{loading} + \text{desorbed}}
\]

Table 1 lists the pore structure parameters. The porous system of the commercial activated cloth (ACC) is mainly composed of a well developed micropore volume, resulting in a BET surface area close to 800 m\textsuperscript{2} g\textsuperscript{−1}. The volume of mesopores is small. Ammonia-steam activation enlarged the pore volume (Gurvich volume over 0.5 cm\textsuperscript{3} g\textsuperscript{−1}) close to 1200 m\textsuperscript{2} g\textsuperscript{−1} and the micro pore width (from 0.91 to 1.24 nm). Burn-off (b.o.) of approximately 13\% obtained after ammonia-steam activation at 800°C during 40 min is low in comparison with burn-off achieved by steam alone (b.o. of 20\% after a 30-min soaking time at 700°C). This low burn-off was explained by Li et al. [5] who showed that ammonia inhibits steam activation. NH and NH radicals formed by NH decomposition would quench the active sites formed by reaction of steam with the carbon matrix.

Table 1

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Volume (V_{\text{m}}) (cm\textsuperscript{3} g\textsuperscript{−1})</th>
<th>Surface area (S) mesopores (m\textsuperscript{2} g\textsuperscript{−1})</th>
<th>Micropore volume (V_b) (2\times) (nm)</th>
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<td>0.000</td>
<td>0.317</td>
<td>0.353</td>
</tr>
<tr>
<td>ACCN</td>
<td>0.000</td>
<td>0.472</td>
<td>0.518</td>
</tr>
</tbody>
</table>

Table 3. Results and discussion

3.1. Pore structure

Table 1 lists the pore structure parameters. The porous system of the commercial activated cloth (ACC) is mainly composed of a well developed micropore volume, resulting in a BET surface area close to 800 m\textsuperscript{2} g\textsuperscript{−1}. The volume of mesopores is small. Ammonia-steam activation enlarged the pore volume (Gurvich volume over 0.5 cm\textsuperscript{3} g\textsuperscript{−1}) close to 1200 m\textsuperscript{2} g\textsuperscript{−1} and the micro pore width (from 0.91 to 1.24 nm). Burn-off (b.o.) of approximately 13\% obtained after ammonia-steam activation at 800°C during 40 min is low in comparison with burn-off achieved by steam alone (b.o. of 20\% after a 30-min soaking time at 700°C). This low burn-off was explained by Li et al. [5] who showed that ammonia inhibits steam activation. NH and NH radicals formed by NH decomposition would quench the active sites formed by reaction of steam with the carbon matrix.

3.2. Surface groups

Aromatic system where one nitrogen atom replaces a carbon atom (e.g. azachrysene or azapyrene). According to Refs. [17–20], the N1s XPS spectra were fitted with four components having mixed Gaussian–Lorentzian peaks with FWHM of 1.8 eV; one pyridinic N peak at 398.3 eV, one pyrrolic N peak at 400.1 eV, quaternary N at 401.3 eV and N-oxide at 403.1 eV. ACCN XPS displays a dominant zone of ‘pyridinic’ nitrogen, as confirmed by XANES measurements (Fig. 2). Similar XPS spectra were reported by several authors: Stohr et al. [1] for peat-based activated carbon treated by ammonia or
HCN at high temperature, Mangun et al. [3,4] for ACFs produced from steam/CO$_2$ activated woven phenolic fibers treated with dry ammonia above 700°C. They could be assigned to 'pyrrolic' N or to 'graphitic' N (N in inner position of polyaromatic clusters with circular catenation, as defined by Pels et al. [18] and Casanovas et al. [19]). XANES work in the literature [21] showed the presence of pyridinic and pyrrolic functionalities after ammonia treatment of carbons, and pyridinic, pyrrolic and pyridone functionalities after high temperature oxidation of ammonia treated carbons. The latter may be comparable to the situation described in this paper where ammonia-steam treatment was used. Similar results have been obtained for ammonia treated oxidized carbons. These data support the conclusions from XPS and TPD experiments described in this paper.

In Fig. 3, TPD N$_2$, which evolves at $\sim$ 1400 K, appears to be the predominant nitrogen gas. After Ikeda and Mackie [22], the initial principal nitrogen-containing product of pyrolysis of pyridine (at 1100–1240 K in a stirred flow reactor) is cyanoacetylene with subsequent elimination of HCN as a secondary product. In the case of carbons, the exact mechanism by which thermal rupture of nitrogen-containing rings occurs has not yet been established. At low heating rate, residence time becomes relatively long, and N$_2$ is supposed to be generated by secondary gas phase reaction of HCN with free radicals and with char-N [23]. TPD N$_2$ profiles presented in Fig. 3 are the same as those obtained by Stanczyk and Boudou [24] for acridine or carbazole cokes. Thermal treatments during ACC preparation or modification would have removed the small temperature shoulder observed at $\sim$ 1050 K with the model compounds. The increase of nitrogen emission and the correlated reduction of hydrogen emission, occurring in two peaks centred at 1200 and 1500 K during ACCN TPD, suggests that N atoms could

<table>
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<th>Table 2</th>
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<tr>
<th>Sample code</th>
<th>XPS N/C ratio</th>
<th>TPD (mmol g$^{-1}$)</th>
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<td>0.29</td>
</tr>
<tr>
<td>ACCN</td>
<td>5.69</td>
<td>0.01</td>
</tr>
</tbody>
</table>

$^a$ Atomic N$_{1s}$/C$_{1s}$ ratio $\leq 100 \leq 0.595$ (instrumental sensitivity factor).

$^b$ CO$_2$ as free carboxylic-type (675 K).

$^c$ CO$_2$ as lactone-type (675 K). $^d$ CO as ketone-type (1,1250 K). $^e$ CO as pyrone-type (1,1250 K).
have replaced CH aromatic groups during attack by N radicals generated from ammonia at high temperature. Table 2 gives the integrated amounts of thermally desorbed CO$_2$ and CO, expressed as mmol per gram of dry sample. H$_2$O was essentially physisorbed. In contrast to the untreated sample (ACC), TPD of the ammonia-steam

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3.3. Breakthrough curves
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The normalized breakthrough curves for H$_2$S and SO$_2$ adsorbed over as-received ACC and the ammonia-modified sample (ACCN) are presented in Fig. 4. Experimental time is expressed as the cumulative amount of H$_2$S or SO$_2$ introduced into the column at constant inlet/outlet pressures, flow rates and inlet concentration ($C_0$: 5000 ppmv for H$_2$S and 3000 ppmv for SO$_2$). This unconventional way to express the time axis enables to read directly the retention rate at the breakthrough point. Compared to Mochida’s setup [6], experimental conditions were simplified by using a vertical reactor and low W/F ratio to avoid any continuous elution of H$_2$SO$_4$ out of the fixed bed during adsorption (this enabled us to better compare H$_2$S and SO$_2$ dynamic adsorption). In these conditions, breakthrough curves level off at $C/C_0 = 1$, where $C$ is the outlet concentration. Activated carbon cloth obtained by ammonia-steam activation (ACCN) exhibits enhanced SO$_2$ and H$_2$S retention. However, H$_2$S and SO$_2$ loading dropped substantially after the first thermal regeneration. This drop has already been reported by Bagreev et al. for H$_2$S adsorbed on granulated activated carbons after regeneration by washing [28] or thermo-oxidative regeneration [29], and by Muniz et al. [2] for SO$_2$ cyclic adsorption–thermal desorption on ammonia treated ACF. The oxidation of the fiber, due to regeneration, could explain the irreversible decrease of the ACF SO$_2$ regeneration ability. The strong increase of H$_2$S loading from the first to the second adsorption steps on the untreated ACC sample could be due to the removal of most acidic groups, which have a negative effect on the dissociation of H$_2$S in the surface water film [30]. In the case of the N undoped
ACC, SO₂ adsorption is insensitive to the presence of surface acidic groups due to the fact that SO₂ is much more soluble in water than H₂S in acidic surface pH range [31].

3.4. Adsorbed sulfur species

The XPS S2p signals from the adsorbed sulfur on ACCN are shown in Fig. 5. By using the energy position of the sulfur 2p signal given by XPS data bank tables of pure reference compounds, it was possible to deconvolute the spectrum from adsorbed sulfur into three doublets (S2p3/2 and S2p1/2) of mixed Gaussian–Lorentzian line-shape having 2:1 relative intensity and FWHM of 1.8 eV and a maximum at approximately:

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Fig. 4. Breakthrough profiles of H₂S (5000 ppmv) or SO₂ (3000 ppmv) for the untreated or ammonia treated activated carbon cloth. (1–4) Means first to fourth adsorption–thermal regeneration cycle.

Fig. 5. Overall (I), N1s (II) and S2p (III) XPS spectra for ACCN after adsorption of H₂S and SO₂.
1. 163.5 eV for elemental sulfur (S\(_n\)) and for organic sulfides. After four consecutive H\(_2\)S adsorption–desorption–thermal regeneration cycles on ACCN, the proportion of residual sulfur extracted by temperature programmed reduction (TPD at 30 K min\(^{-1}\) with a carrier gas made of a mixture of 5% H\(_2\) in He) was about 40% of the total adsorbed sulfur. This proportion reached about 60% for ACC. TPR profiles showed a single H\(_2\)S peak centred at 930–1000 K which could be assigned to polyaryl sulfides or thiophenes [32,33].

2. 167.6 and 168.7 eV for SO\(_3\) and SO\(_4\) species, respectively. In agreement with XPS, TPD did not show any physisorbed SO\(_2\)—as reported by some authors [34,35], indicating that the major part of SO\(_2\) was oxidized to SO\(_3\) or converted to \(\text{H}_2\text{SO}_4\) by hydration of SO\(_3\) after its adsorption. Fig. 5 shows an increase of the 401.2 eV component and an apparent collapse of the pyridine component due to a complexation of SO\(_3\)/SO\(_4\) with N surface groups. This change is more pronounced after SO\(_2\) than after H\(_2\)S adsorption. It is well known that when pyridinic N is involved in adsorption complexes, the N1s core electron can undergo an \(\pm 1\) to \(\pm 1.7\) eV chemical shift, compared to the N1s binding energy for pure pyridine [36–38]. Such an effect of SO\(_2\) on the N1s XPS of ammonia-treated ACF has only been reported by one other group [5]. The authors assign the 401.6-eV peak to a conversion of pyridine nitrogen to quaternary nitrogen. Sulfur trioxide is known to be one of the most reactive inorganic compounds; it is an oxidizing agent and a Lewis acid able to combine with free electron pairs of various atoms—in these the donor strength is variable: it reaches its maximum with nitrogen. Otherwise, with excess water SO\(_3\) combines vigorously to form sulfuric acid—with less than equimolar amounts of water—polysulfuric acids are formed. Comparison of total sulfur loading with the part of complexed sulfur on nitrogen sites—assuming that there is 1 mol per nitrogen site—implies a partitioning of sulfur between the carbon surface and the surface nitrogen sites. In the case of SO\(_2\) adsorption, the proportion of complexed sulfur on N sites was appreciably higher than for H\(_2\)S adsorption. As presented in the next section, study of ACCN activity regeneration showed that sulfur complexation can be reversed by washing/hydrolysis extraction.

3.5. Regeneration

By averaging the retention rate of the second, the third and the fourth successive adsorption–thermal–regeneration cycles, one can calculate that H\(_2\)S retention rate on ACCN was only increased by about 32% with respect to the untreated sample and by about 71% for SO\(_2\). However, in contrast to H\(_2\)S, thermal regeneration produced a quick decrease of the oxidative retention for SO\(_2\) (Fig. 6). After several consecutive SO\(_2\) adsorption–thermal regeneration cycles, TPD of ACCN (Fig. 7) revealed that, contrary to H\(_2\)S, thermal regeneration had not only produced an oxidation of the carbon matrix (introduction of acidic surface groups), but also of the nitrogen aromatic surface groups. When regeneration was performed by washing/hydrolysis in a solution of 28% ammonia in water at room temperature, ACCN activity for the oxidative adsorption SO\(_2\) was almost completely recovered (Fig. 6)—much less for H\(_2\)S adsorption. After four consecutive SO\(_2\) adsorption–washing cycles on ACCN, TPD did not release any residual sulfur and showed that the carbon surface had
been only slightly oxidized. In these conditions, the average SO₂ retention rate increased by about 350% with respect to the untreated sample. Compared to SO₂, the higher thermal regenerability of N-containing activated carbon after H₂S adsorption, is due to a smaller H₂SO₄ generation, and the lower efficiency of the N treatment for H₂S adsorption/oxidation can be explained by the fact that elemental sulfur and a substantial amount of water from H₂S/HS⁻oxidation, as well as 

subsequently formed by oxidation of S to SO₂ and SO₃, remains in the micropores, block the active centres in this region and lower the surface pH [39]. In the case of SO₂, free H₂SO₄ tends to be continuously eluted by moist air flowing through the microporous carbon [6].

4. Conclusions

Ammonia-steam treatment at 800°C of a viscose-based activated carbon cloth produced a development of porosity at the expense of a slight micropore widening (1.24 nm). The treatment selectively transformed the surface map of functional groups by introducing a high density of pyridinic-type nitrogen and by thermally desorbing the largest part of acidic oxygen groups, leaving behind some residual basic oxygen groups which generate CO at high temperature. The results obtained by cyclic adsorption–regeneration on fixed bed show that the ability of the nitrogen-doped ACC to remove SO₂ or H₂S from moist air at room temperature is clearly enhanced by the following factors affecting its porosity parameters and chemical properties:

- Increase of micropore volume (expressed also by a high BET surface area) without a too large widening of micropores—leading to a decrease of the adsorption energy.
- Removal of most surface acidic groups present on the undoped ACC.
- Increase of surface basic and polar sites: pyridinic groups and pyrone-type groups.

The following differences have been observed between H₂S and SO₂ adsorption–regeneration:

- Ammonia treatment augmented more strongly the oxidative retention of SO₂.
- After desorption of the physisorbed sulfur, XPS measurements showed that sulfite/sulfate and perhaps SO₃ form complexes on the nitrogen surface sites.
- Thermal regeneration of the spent ACCN after SO₂ adsorption produced a considerable oxidation of the carbon matrix and of the nitrogen sites, while thermal regeneration after H₂S adsorption produced a little oxidation

- Regeneration by washing with cold aqueous solution of ammonia—to help decomplexation—removed all sulfur left by SO₂ adsorption while insignificant sulfur removal occurred by washing the spent N-doped ACC after H₂S adsorption which essentially produces elemental sulfur and organic sulfur covalently bound to the carbon matrix and a small amount of strongly trapped sulfates. In spite of some sulfur complexation, a better mass transfer of the reaction products out of the micropores during adsorption partly accounts for the higher positive influence of the ammonia treatment on the catalytic oxidation of SO₂ at room temperature.

Acknowledgements

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