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Adsorption of Phenol from Aqueous Solutions on Ca/SBA-15 material

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Abstract

Ordered hexagonal mesoporous Ca/SBA-15 with a large pore diameter was synthesized by an impregnation method using an aqueous solution of calcium chloride as a precursor.The catalytic activity of Ca/SBA-15 was evaluated for the adsorption of phenol in presence of formic acid atambient temperature and under atmospheric pressure.The phenol adsorption capacities of the adsorbents showed that formic acid pretreatmentsignificantly enhanced the surface properties, consequently the adsorption capacities of theadsorbents**.**

1. Introduction

Industrial waste producesmolecules that may pollute water, soil and underground water due to the harmful impacts on ecosystems and humans.Phenolic compounds are common contaminants in water effluents ofmany industries such as oil refineries, plastics, and leather and paint industries. Phenolic compounds are considered as pollutants because they are toxic and hazardous to living organisms, even at very low concentrations. Despite their useful applications as pesticides, lubricants and solvents, phenolic compounds are common poisonous, industrial and persistent pollutants. Several methods such as adsorption[1,2] chemical oxidation [3], coagulation flocculation [4] have been already used to remove phenol containing derivatives from wastewater. Amongall these methods, adsorption appears to be the most effective. The process that is the most widely used for wastewater treatment relates to adsorption methods. In fact, during the last few years, a great variety of materials such as activated carbons[5–8], silica [9,10], polymeric resins [11,12], fly ash [13], clays including kaolinite [14], and zeolites [15-19], has been explored in detail for the removal of phenolic pollutants from wastewater.

Mesoporous materials have a very high adsorption capacity and interesting catalytic propertiesfor organic molecules and they are, therefore, widely employed in wastewater.As such the use of mesoporous materials seems to be promisingbecause of their larger pore volume and diameter, their high surface area, and their regular channel type structures [20,21].Mesoporous silicas with a narrow pore size distribution haveattracted increasing attention as a novel material for separations, adsorption and reactions involving large molecules. Several types of mesoporous silica havebeen developed [22,23].

In the present work, SBA-15 supports are synthesized and their performances for Ca loading was tested in phenol adsorption in presence of formic acid to improve the surface properties of the adsorbents.

2. Experimental

2.1. Preparation of SBA-15 Mesoporous Material

The SBA-15 mesoporous material was prepared using classical synthesis conditions [24].

The particularity of this manipulation compared to the classical synthesis is that the preparation was performedin abeaker without an autoclave. In theseconditions,we obtainedawhite solid that underwent a hydrothermal treatment at 140°C in order to achieve large pore size and limited microporosity.

The solution was further maintained at room temperature for 24 h under stirring for aging. The SBA-15 support was calcined under air at 550°C for 4 h (temperature increase rate= 1°C/min).

2.2. Ca Supported on SBA-15 Material

Various quantities of calciumwereimpregnatedon SBA-15 using $CaCl_2$, 2 H_2Oas precursor salt introduced in a beaker containing 25 mL of distilled watermixed withSBA-15, under vigorous stirring for 15 minutes. HCl and NaOH solutions were respectively used to adjust the pH of the solution. The resultingsuspension was kept under stirring at room temperature for 24h. The solid was separated by filtration anddried at 100°C for one nightand calcined at 400°C in astove for 24 h.

2.3.Phenol Adsorption

Batch adsorption tests were carried out using the bottle pointed method as follows.

Two beakers containing 50 mL of aqueous phenol solutionwere used, 1 g of Ca/SBA-15 catalyst was introduced into each beaker. Formic acid was added to one of the beakers. After 3 h stirring, the mixturesare filtered through a funnel and the concentrations of phenolin solution were determinedusing UV/VIS spectrophotometry at510 nm.

2.4. Catalysts Characterization

Elemental analysis (calcium and titanium) was performed using atomic adsorption fromPerkin. Samples were dissolved in HCl solution under heating before analysis.Specific surface area, pore size distribution and pore volume were obtained from N_2-sorption isotherms collected on a TRISTAR instrument from Micromeritics. Samples were degassed for 1 night at 300°C until the residual pressure in the analysis cell was below 0.15 mbar. The specific surface area was determined from the linear part of the BET plot. The mesopore size distribution was determined by the non local density functional theory (NLDFT)method and calculated using the Autosorb-1 1.52 software. The kernel selected was N_2BETon silica assuming cylindrical pore geometryand the equilibrium based on the desorption branch. Pore volumewasdetermined on the isotherms at P/P_0= 0.97.

3. Results and Discussion

3.1. Textural Analysis

Figures 1 and 2 present the results of the characterization of the SBA-15 sample by N_2-sorption. The synthesized material exhibited a type IV isotherm according to IUPAC classification,with an apparent hysteresis loop indicative of the existence of defined uniform cylindrical mesopores in the frameworks of relatively larger dimensions.Moreover, the pronounced steep in adsorption branche of the isotherm favors a narrow pore size distribution, which is typical of the well-ordered mesoporous structures.

Figure 1: N_2 adsorption–desorption curves at 77 K for SBA-15material

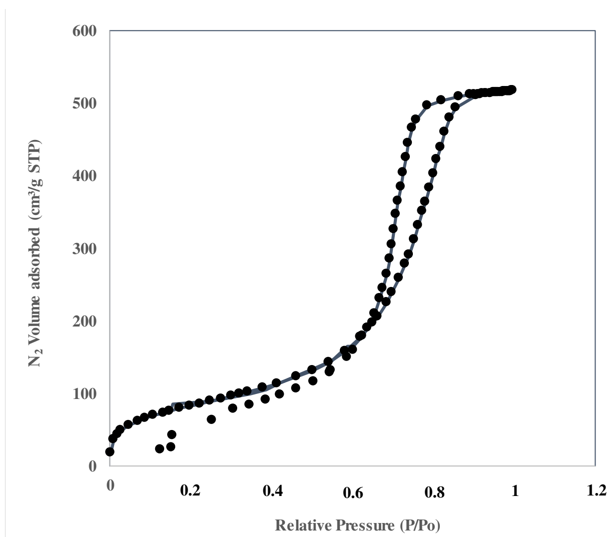
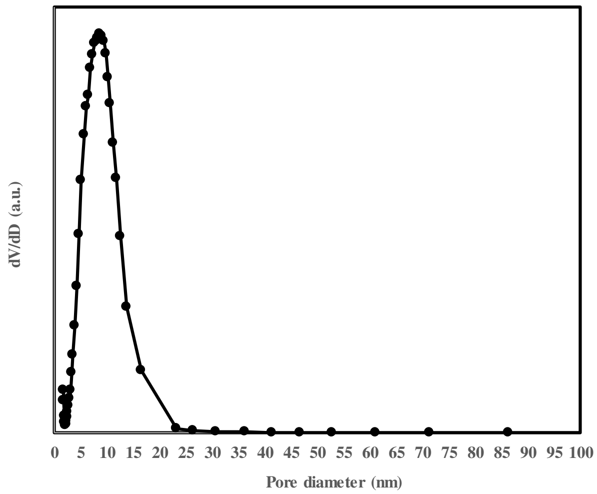


Figure 2: NLDFT pore size distribution obtained over the SBA-15material

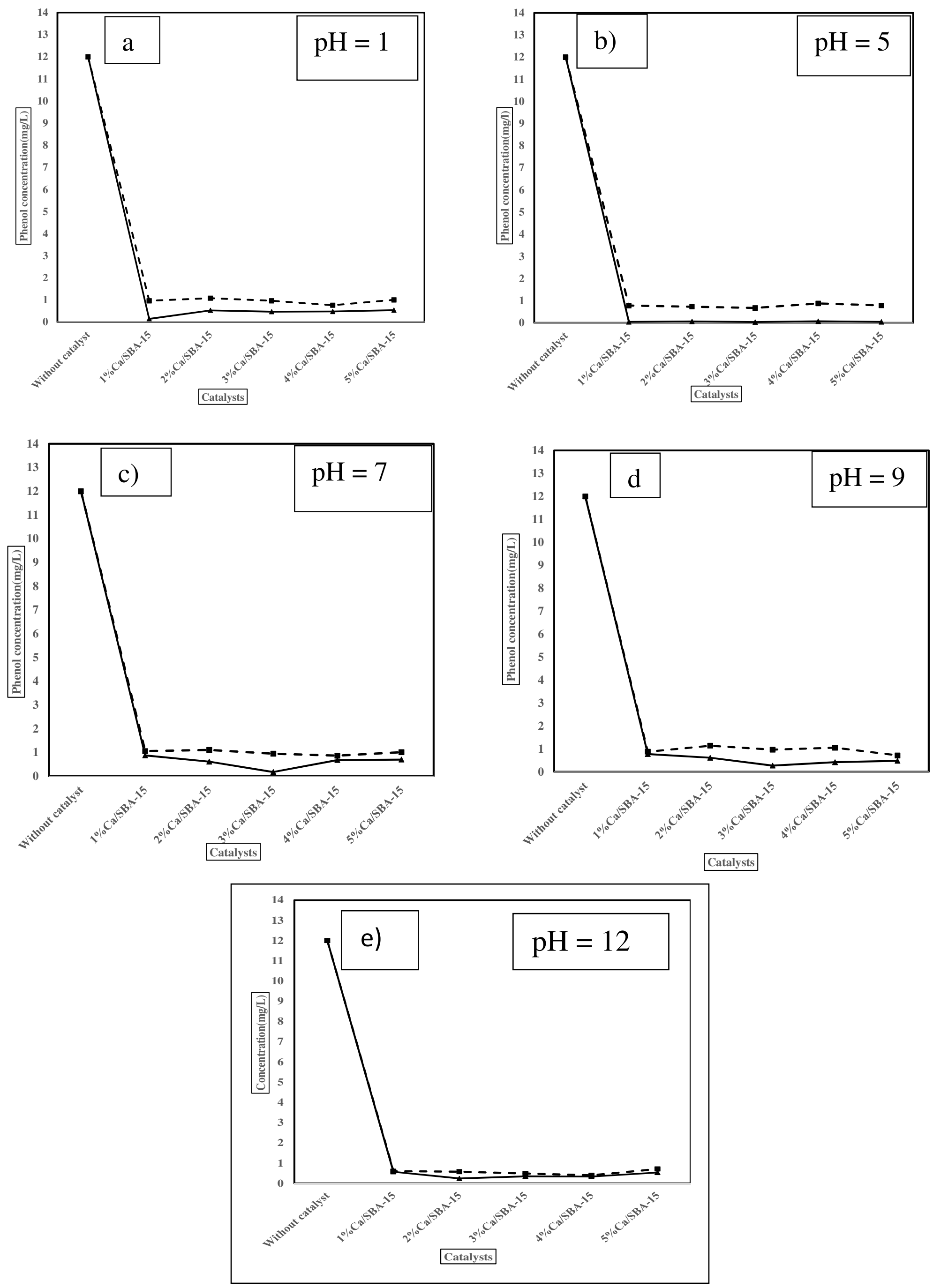


The pore-size distribution for the pure SBA-15 (Figure 2) determined by using BJH adsorption leads to an average pore diameter equal to 7.7 nm. BJH desorption cumulative volume indicates a pore volume of 1.1 cm^3/g. The BET surface area was measured to be $309 m^2/g$.The pore volume and the pore diameter are in good agreementwith the results reported in the literature [25-27].

3.2. Phenol Adsorption over Ca/SBA-15 Materials

The study of phenol adsorption over x%Ca/SBA-15 materials (x in wt% is the value of Ca content introduced in solution) in presence or without formic acid shows that the adsorption capacities are very high (Figures 3 a-e).

Figure 3: Adsorption of phenol over Ca/SBA-15 materials without formic acid (■) and in presenceof formic acid (▲) at different pH: (a) pH = 1, (b) pH = 5, (c) pH = 7, (d) pH = 9, (e) pH = 12.



The study was performed with formic acid during the adsorption of phenol (Figures 3). A difference in adsorption capacities according to the x%Ca/SBA-15 material and the pH was observed indicating generally ahigher efficiency when formic acid is present.

This means that the proportion of ionizedformic acid and ionized phenolwas more important.

Considering that the molecular size of phenol is about 7 Å, the observed adsorption behavior reveals that in adsorbents with a mean micropore width of less than 7.7nm, a fraction of the small pores do not take part in the adsorption process. This is because those pores arenot accessible to the phenol molecules.

4. Conclusion

Our study of adsorption of phenol over Ca supported on mesoporous SBA-15showed that there is an appreciable catalytic performance in terms of phenol adsorption and better activities can be obtained by formic acid loading. It is shown that formic acid enhanced the adsorption characteristics of the adsorbents.

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