ACTIVATED CARBON HONEYCOMBS FOR CHEMICAL REACTIONS APPLICATIONS

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Introduction

Monolithic activated carbon honeycombs with various pore size distributions suitable for chemical and petrochemical applications have been fabricated using a synthetic polymeric carbon source and Corning's proprietary cellular honeycomb extrusion technology. The results reported here include Corning's capability in tailoring pore size distribution of the activated carbon honeycomb, their property and examples illustrate their superior performance as a catalyst support in selected chemical reaction applications.

Experimental

Activated carbon honeycomb fabrication: Activated carbon honeycombs were fabricated using high carbon yield polymeric resin with low viscosity. The principle of the process has been described in a previous paper by Gadkaree [1]. The main steps include dipping a porous ceramic honeycomb in a resin, drying and curing, carbonizing in nitrogen and activating in carbon dioxide or steam.

Analysis: The honeycombs are analyzed by nitrogen adsorption at 77K using Quantachrome Autosorb 1 for pore size distribution. The *t*-method micropore volume and BJH desorption mesopore and macropore volumes are used to obtain the cumulative percent pores in the three pore size regions.

Carbon catalyst manufacture: Activated carbon supported honeycomb catalyst was produced by incipient wetness impregnation method, one of the most popular commercial approaches to make carbon-supported catalysts. About 1wt% of Pt based on carbon was loaded onto the carbon honeycomb. The samples were dried at 120°C overnight and then calcined at 400°C for 2 hours in inert gas. The catalyst was reduced at 400°C in hydrogen for 2 hours.

Reaction: Toluene hydrogenation was selected to evaluate catalyst performance because there is a well-established relationship between catalytic activity and catalyst properties. The reaction was carried out at 100°C and 50 mL/min H2 and 0.01 mL/min liquid toluene. The space velocity is 864 (1/hr). For commercial catalysts, the equal amount of Pt in catalysts was charged into the reactor. To make sure the same space velocity, the obtained samples were diluted with cordierite granules.

Results and Discussion

Physical Property

Ceramic honeycombs coated with polymer resin were subjected to various carbonization and activation conditions to modify pore size and its distribution. Widely different pore size distributions have been obtained through tailoring the processing conditions. Figure 1(a) and 1(b) illustrate the pore properties measured by nitrogen adsorption at 77K. In the mesorpore range of the BJH desorption branch, pores centered at around 38, 50, 75 and 100 angstroms with broad distribution have been obtained, as is shown in Figure 1(a). Figure 1(b) shows the capability of engineering the bi-modal and tri-modal distributions in the mesopore range. Table 1 shows the physical property of Corning's honeycomb carbon catalyst supports. Surface area of these carbon catalysts ranges from 700 to 1000 m2/g and pore volume ranges from 0.6 to as large as 1.55 cc/g carbon. At least 50 vol% of the pores are in the mesopore range. The fraction of meso-pores can be increased to about 80 to 90 vol%.

Catalytic Performance

The catalytic activity (turnover frequency: s^{-1}) of the carbon honeycomb supported Pt catalysts in gas-phase toluene hydrogenation is shown in Figure 2 with comparison to commercial beaded catalysts of Pt/C and Pt/ Al_2O_3. It is clear that Corning's carbon honeycomb supports catalyzed with Pt perform at least an order magnitude better than commercial Pt/C and Pt/Al_2O_3 catalysts in term of metal use efficiency. Furthermore, four carbon honeycombs formed at different conditions (as listed in Table 1) to possess different pore size distribution have significant impact on their corresponding catalytic activity as shown in Figure 2.

Conclusions

Capabilities of tailoring pore size and its distribution of the polymer based honeycomb catalyst support have been demonstrated. This is very important in using as support in chemical processing applications. The model reaction of hydrogenation reaction of toluene to methylcyclohexane indicates that the best Corning's honeycomb carbon catalyst support catalyzed with Pt has at least 10 times higher turn-over frequency than the three commercial carbon catalysts tested. The data also indicate that the pore size distribution is a dominating factor in optimizing the hydrogenation activity of the Corning catalysts.

References:

Gadkaree, K.P., ***Carbon***, 1998; 36 (7-8):981-989.

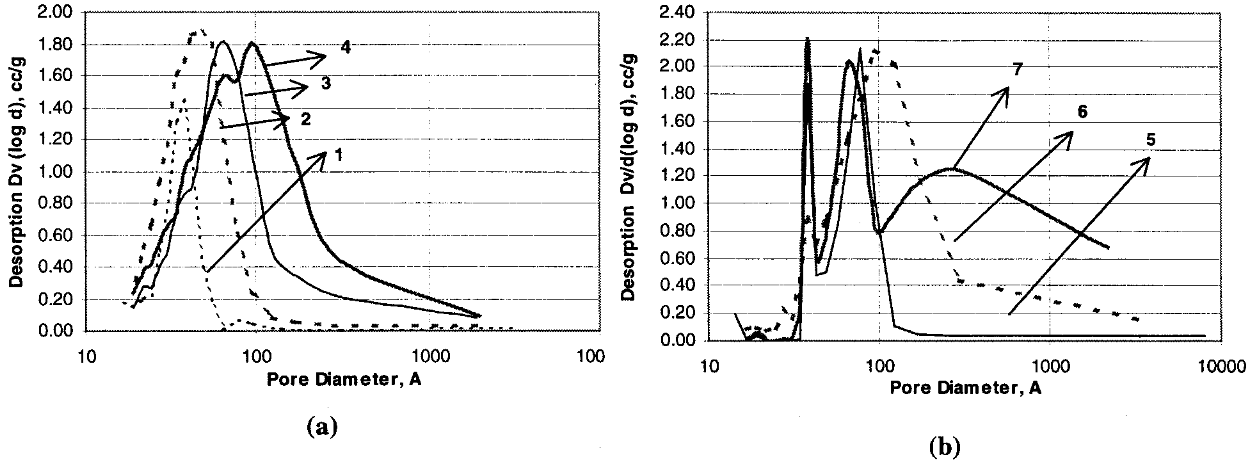

Figure 1. Pore Size Capabilities of Corning's Activated Carbon Honeycomb Catalyst Supports

Table 1. Physical Properties of Corning's Activated Carbon Honeycomb Catalyst Supports

sample	S.A., m2/g	Pore volume			total pore volume	frac. micro	frac. meso	frac. macro
		t-micro	meso	macro	cc/g			
1	698	0.27	0.32	0.02	0.61	0.45	0.52	0.04
2	1041	0.29	0.81	0.03	1.12	0.26	0.72	0.02
3	863	0.17	0.91	0.07	1.16	0.15	0.79	0.06
4	828	0.04	1.30	0.08	1.41	0.03	0.92	0.06
5	939	0.32	0.59	0.03	0.95	0.34	0.62	0.03
6	922	0.09	1.28	0.17	1.55	0.06	0.83	0.11
7	853	0.01	1.35	0.61	1.97	0.01	0.68	0.31

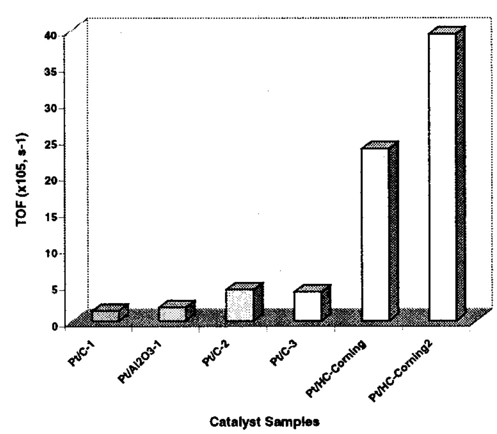

Figure 2. Turn-over Frequency of Corning's Catalyzed Carbon Honeycomb Support and Three Commercial Beaded Catalysts in Toluene Hydrogenation Reaction.

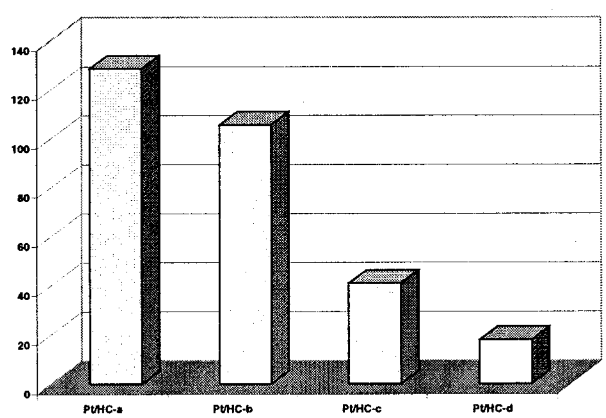

Figure 3. Effect of Pore Size Distribution on the Turn-over Frequency of Corning's Catalyzed Carbon Honeycomb Supports in Toluene Hydrogenation Reaction.