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Low-Temperature H₂S Removal from Steam-Containing Gas Mixtures with ZnO for Fuel Cell Application. 2. **Wash-Coated Monolith**

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This work is part of our efforts to explore more-effective ways to remove hydrogen sulfide (H₂S) for fuel cell applications. Various absorbents (ZnO, SnO₂, coprecipitated NiO-MoO₃, supported CuO-ZnO, V_2O_5 -ZnO, and ZnO supported on γ -alumina) were tested for H_2S removal. The absorbents that were wash-coated onto the monolith were compared with particulate traps in the inlet H₂S concentration range of 0.5-8 parts per million by volume (ppmv). The monolith provides much-higher dynamic capacity (the amount of H₂S trapped before breakthrough) under the same conditions. The ZnO-based monolith demonstrated the best performance. An extremely low H₂S outlet concentration (as low as 20 parts per billion, by volume (20 ppbv)) was observed over ZnO-based monolith samples for extended periods of time, under various conditions relevant for the desulfurization of gas products from the autothermal reforming of hydrocarbon fuels for a proton-exchange membrane fuel cell. The capacity of the H2S trap is dependent on the monolith characteristics (active component loading per cubic inch, and the number of cells per square inch) and operating conditions (including inlet H₂S concentration, space velocity, and temperature). Wash-coating of ZnO that was chemically modified by an ammonium carbonate treatment onto a monolith with 400 cells per square inch gave the best H₂S absorbence with higher dynamic capacity.

Introduction

One of the promising ways to increase the efficiency of energy utilization is to use a fuel cell instead of the combustion of hydrocarbon fuels, because the former is intrinsically highly efficient. The greatest concern for low-temperature proton-exchange membrane (PEM) fuel cells that use a hydrocarbon fuel is hydrogen purity. In particular, even traces of sulfur compounds and CO could greatly decrease the outlet power.

In the gaseous products from the autothermal reforming of hydrocarbon fuels, the sulfur compound is hydrogen sulfide (H₂S), which must be removed before the subsequent water-gas shift reaction and before entering into the anode chamber of a fuel cell stack. A particulate ZnO-based H2S trap was developed in our recent work. The particulate absorbent that consisted of modified ZnO was effective in the temperature range of 300-400 °C, but the dynamic capacity (the capacity before breakthrough) for sulfur removal from the steamacross the bed. To solve the aforementioned problem, we have begun to explore monolith-based H₂S traps. Monoliths have several advantages, relative to packed beds. Monoliths usually have greater geometric surface areas and can be operated at only a small fraction of the pressure decrease of the particulate bed.^{2,3} Monoliths can incorporate the added advantage of containing the active component in a thin layer, such that its pores are highly accessible to the reactant flow. Because the thin layers show lower resistance to pore diffusion, wash-coated

containing mixture was low (0.417 g S/100 g ZnO in the case of 3-mm extrudates and 2.8 g S/100 g ZnO in the

case of particles 0.106-0.15 mm in size at 300 °C and

a space velocity (SV) of 2660 h^{-1}). To increase the

dynamic capacity of the trap with ZnO particles whose

utilization was limited by diffusion, their particle size

should be as small as possible. However, a major

concern with small particles is the pressure decrease

monoliths may be more productive at high conversions, relative to particles on a volume basis. Studies in the

1970s showed that a monolith cellular structure was

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more effective than particulates for catalytic converters for automotive applications.

The purpose of the present work is to clarify the difference in the dynamic performance of particulate and wash-coated monolithic absorbents for H₂S removal, and to determine if the use of a wash-coated monolith could provide higher dynamic capacities at low-pressure decreases through the bed for fuel cell (FC) applications.

Experimental Section

Commercially available ZnO samples (HTZ-5, supplied by Haldor-Topsoe) and ZnO samples prepared by Engelhard Corporation were tested in the form of wash-coated monoliths and in the form of 3-mm cylindrical extrudates. The ZnO sample from Haldor-Topsoe was prepared using commercially available ZnO powder, and the extrudates were made without any binder or additives, according to the sample specification. For the Engelhard ZnO samples, commercially available ZnO powder was chemically modified by a pretreatment using ammonium carbonate, calcined at 300 °C, then mixed with a Ca-bentonite binder and extruded. The procedure for the pretreatment and extrudate preparation has been described in our companion paper.1 The bulk chemical composition of the ZnO powder samples (without binder) from both Engelhard and Haldor-Topsoe is ZnO; however, these samples differ in regard to morphology.1 ZnO samples provided by Nanophase Technologies Corporation (denoted as Nanotek) and US Zinc (referenced as AZO-66) were also tested for comparison. SnO₂, $Zn(NO_3)_2 \cdot 6H_2O$, γ -Al₂O₃ (95 m²/g), Cu(NO₃)₂·2.5H₂O, Ni(NO₃)₂· 6H₂O, (NH₄)₆Mo₇O₂₄·4H₂O, and NH₄VO₃ were purchased from

Supported samples, including 21.6% copper supported on ZnO, 5% vanadium supported on ZnO, and pure ZnO supported on γ -Al₂O₃, were prepared using the standard incipient wetness technique, dried at 120 °C, and calcined at 400 °C in air. A NiO-MoO₃ absorbent was prepared by coprecipitation using the following procedure. Thirty-three grams of nickel nitrate (Ni(NO₃)₂·6H₂O) was dissolved in 300 mL of H₂O at room temperature. Ammonium heptamolybdate ((NH₄)₆Mo₇O₂₄· 4H2O, 51.44 g) was dissolved in 1000 mL of H2O at room temperature. The two solutions were mixed together under constant stirring and boiled for 8 h. The precipitate was filtered, washed with 2 L of deionized water, and wash-coated using the procedure described below.

ZnO, SnO₂, coprecipitated NiO-MoO₃, ZnO-γ-Al₂O₃, and CuO-ZnO absorbents were wash-coated onto a piece of monolith using the procedure described below. All the absorbents were used without binder in monolith preparation. The wash-coated ZnO-containing samples were used in the form of cylindrical monoliths with the same diameter (19.05 mm (3/4 in.)) but with different lengths (9 mm (3/8 in.) or 76.2 mm (3 in.)), different numbers of cells per square inch (CPSI), and different loadings of absorbent.

A piece of monolith with a prescribed size and CPSI value was cut out and weighted. A slurry composed of 40% solid material in water was prepared in a blender. The slurry was milled until 90% of the particles were smaller than 0.01 mm. The slurry was placed in a container of sufficient depth such that a monolith carrier can be fully immersed in the slurry. The monolith carrier was dipped in the slurry, and the freeflowing excess liquid was allowed to drain off. Blocking of the channels of the monolith carrier was minimized by blowing air across the face of the carrier through the channels. The monolith was then dried as 120 °C for 1 h in an oven. The coated monolith carrier was then cooled and weighed, to estimate the ZnO loading. Thereafter, the process (immersion in the slurry, draining off the excess liquid, blowing air through the channels, followed by drying) was repeated to attain the desired coating thickness. The coated monolith carrier was

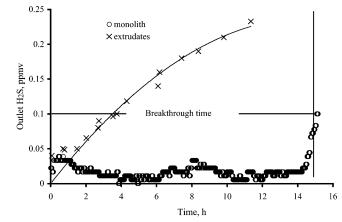


Figure 1. Performance of a wash-coated ZnO monolith and 3-mm ZnO-based extrudates for H₂S capture. Conditions: 400 °C; 8 ppmv H₂S, 34.4% H₂, and 13% H₂O, with the balance being \hat{N}_2 . The 3-mm extrudates were tested at a space velocity (SV) of 8775 h⁻¹, and the monolith (400 CPSI) with a ZnO loading of 1.07 g/in.3 was tested at SV = 11 000 h⁻¹.

placed in a fused silica tray and calcined at 400 °C in air in a furnace for 2 h and weighed again. The loading was calculated by the weight difference:

$$\text{Loading (g/in.}^3) = \\ \frac{m_{\text{coated monolith}} - m_{\text{starting monolith}}}{m_{\text{starting monolith}}} \times D_{\text{starting monolith}}$$

where $m_{\text{starting monolith}}$ and $m_{\text{coated monolith}}$ are the weight of the starting monolith and coated monolith, respectively. D_{starting monolith} is the bulk density of the starting monolith (in units of g/in.3).

The sample performance was tested in a flow reactor. The reactor tube was made of Pyrex with an inner diameter of 20 mm. The adsorbent was maintained isothermal, and the temperatures in the top and bottom of the absorbent bed were also monitored. Feed gas contained 1-8 ppmv H₂S, 43% H₂, with the balance being N_2 , to which 13% or 20% of water (H_2O) vapor was added. The flow rate, based on dry gas, was 200-2000 mL/min. When the dry gas flow rate was 200 mL/min, the addition of 13% H_2O means adding 30 mL/min of stream to give a total flow rate of 230 mL/min. The outlet H₂S concentration was monitored using an ONIX CVI H2S analyzer, which has a lower detection limit of 0-20 ppbv and an upper limit of 10 ppmv. The details of the reactor design, analytical procedures for outlet H₂S analysis with calibration, and capacity calculation method were described in the companion paper.1

Results and Discussion

Figure 1 compares the profiles for H₂S removal by two types of ZnO absorbents from Engelhard: 3-mm extrudates and a wash-coated monolith. The use of a ZnO wash-coated monolith led to a major performance improvement, in that the breakthrough time (BT) was extended from less than 4 h to more than 14 h. Despite the higher space velocity for the test with the washcoated monolith, the outlet H₂S concentration was less than that with the extruded material. Monoliths also increased the dynamic capacity, from 0.047 g S/100 g ZnO for the 3-mm extrudates (400 °C; SV = 8775 h^{-1}) to 3.54 g S/100 g ZnO for the monolith (400 °C; SV = 11 000 h^{-1}) that was wash-coated with the same type of Engelhard ZnO that was modified by ammonium carbonate pretreatment. 1 Thus, wash-coated monoliths

Table 1. Dynamic Capacity of Several Types of H_2S Absorbents Wash-Coated onto the 400 CPSI Monolith^a

sample	capacity (g S/100 g absorbent)
ZnO^b	3.54
5% V-ZnO	1.51
$21.6\% \text{ Cu-ZnO}^c$	1.18
$13\% \text{ Ni-MoO}_3$	0.94
SnO_2	0

 a Conditions were as follows: 8 ppmv H₂S, 37% H₂, and 13% H₂O, with the balance being N₂; SV = 11 000 h⁻¹; 400 °C. b ZnO modified by ammonium carbonate pretreatment. c Absorbent was tested at 300 °C, SV = 6936 h⁻¹, and 20% H₂O.

could be much more effective in H_2S removal than extrudate absorbents.

Several other types of candidate absorbents have also been tested. The results are presented in Table 1. The capacity of the materials tested decreases in the following order: $ZnO > 5\%~V-ZnO > 21.6\%~Cu-ZnO > 13\%~Ni-MoO_3 > SnO_2$.

Vanadium oxide was reported to be active only at temperatures of >400 °C,4 so it may be an inactive component in the temperature range studied here, because 5% V-ZnO gave a lower value than ZnO. Nickel is known to have an affinity for sulfur compounds. The organic and inorganic sulfur compounds may undergo dissociative absorption on the activated nickel-containing materials. However, nickel forms bulk sulfides after absorbing sulfur. In the presence of hydrogen and steam, bulk nickel sulfides become unstable at temperatures of >200 °C, which leads to a decrease in the dynamic capacity of nickel-based absorbents. That phenomenon perhaps explains why the performance of the nickel-containing trap was low. Molybdenum oxide alone could not provide enough dynamic capacity to be effective under the conditions used.

Tin dioxide (SnO_2) was the worst absorbent candidate, with zero dynamic capacity. Visually, there was no color change after the reaction, which suggests that (i) SnO_2 was not reduced under the reaction conditions that were used, and (ii) SnO_2 does not react with H_2S in the presence of water.

A copper-containing absorbent was tested as a potential candidate material, along with ZnO, based on consideration of the thermodynamics shown in Figure 2. (A software system, HSC Chemistry, from Outokumpu Research Oy in Finland, was used to perform the thermodynamic equilibrium calculations, giving the results shown in Figure 2.) Under the reaction conditions, copper can form two bulk oxides and sulfides with Cu⁺ and Cu²⁺ oxidation states. Copper sulfides are more stable under the reaction conditions than ZnS, because they could not be hydrolyzed as easily as ZnS (see Figure 2). However, the dynamic capacity of the copper supported on ZnO was lower than the capacity of the pure ZnO-based trap. The difference might be attributed to the formation of a solid solution, because all the copper-containing samples were blue before use. Bulk copper oxide is well-known to be black; therefore, copper was not present completely as a bulk oxide on the surface of ZnO.

On the basis of the aforementioned results and discussion, the monolith that was wash-coated with

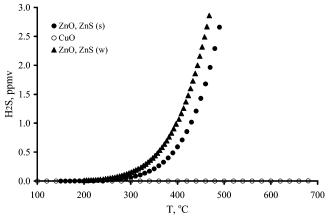


Figure 2. Calculated equilibrium H_2S concentration at different temperatures in the $ZnO-ZnS-H_2O-H_2S$ and $CuO-CuS-H_2O-H_2S$ systems. The initial concentrations were 34.4% H_2 , 8 ppmv H_2S , and 20% H_2O , with the balance being N_2 . Activity coefficients of all compounds were equal to 1. "ZnS (s)" denotes a sphalerite structure, and "ZnS (w)" denotes a wurtzite structure.

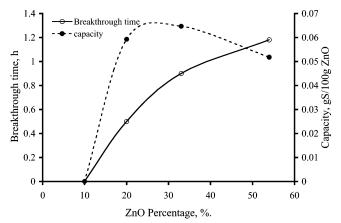


Figure 3. Performance of the γ -alumina-supported ZnO wash-coated onto a monolith. Conditions: 400 °C; 8 ppmv H₂S, 37% H₂, and 13% H₂O, with the balance being N₂; absorbent loading of 1 g/in.³; SV = 11 000 h⁻¹.

Table 2. Physical Properties of Alumina-Supported ZnO
Adsorbent

adsorbent	surface area (m²/g)	pore volume (cm³/g)
γ-Al ₂ O ₃	95	0.55
20% ZnO/ γ -Al ₂ O ₃	65	0.41
33% ZnO/ γ -Al ₂ O ₃	56	0.35
54% ZnO/v-Al ₂ O ₃	41	0.25

pure ZnO was the best among the various materials. Consequently, our subsequent work focused on the monolith samples that were wash-coated with ZnO.

The dynamic capacity of ZnO-based absorbents is dependent on particle size. One of the common methods to create and stabilize small particles of the active component is to support it on a porous carrier, such as alumina. As shown in Figure 3, with 10 wt % ZnO supported on alumina, H₂S breakthrough occurred immediately after the beginning of the reaction. In the 10–25 wt % range of ZnO concentration, the BT value and the capacity gradually increased (see Figure 3), despite a partial loss of pore volume (Table 2). At higher ZnO concentrations, the capacity started to decrease and BT continued its growth. The decrease in dynamic capacity could be attributed to the pore volume loss,

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Table 3. Dynamic Capacity of ZnO with Different Particle Sizes and Morphologies, Wash-Coated onto the Monolitha

sample	surface area (m²/g)	morphology and size of ZnO crystals	loading (g ZnO/in.³)	dynamic capacity (g S/100 g ZnO)
AZO-66	7	cubes and prisms, 200-500 nm	1.13	0.08
Nanotek	18	elongated, 24–71 nm	1.21	0.78
Engelhard	29	small flat plates, \sim 30–50 nm thick	1.07	3.54

^a Testing conditions: 8 ppmv H₂S, 37% H₂, and 13% H₂O, with the balance being N₂; SV = 11 000 h⁻¹; 400 °C.

which prevents H₂S from penetrating into the depth of the alumina channels, leaving some of the ZnO unreacted. Increasing the total loading of ZnO leads to an increase in the amount of ZnO on the external surface of γ -Al₂O₃, which can still react with H₂S. This is the reason why BT continued to grow when the dynamic capacity began to decrease upon further increasing the amount of ZnO on alumina from 33% to 55%. A decrease in the dynamic capacity was also reported when titanium dioxide (TiO₂) was used as a stabilizer.⁵ TiO₂ was used to reduce zinc volatility, 6-8 but, at the same time, it decreases the reaction rate between ZnO and H₂S.⁸ The decrease of the reaction rate might be the reason for the decrease in capacity in the case of the supported ZnO samples. The second reason may be that the capacity of ZnO particles on the external surface of γ-alumina at a low ZnO percentage was too small to absorb all the H₂S above the 0.1 ppmv level. These account for the much shorter BT values in the case of supported samples. Extrapolation of the data to 100% ZnO would indicate that the best absorbent should be pure ZnO with small particles wash-coated onto the monolith.

Several types of pure ZnO samples with different particle sizes and morphologies were comparatively examined, and the results are shown in Table 3. A comparison of the AZO-66 and Nanotech ZnO samples shows that changing the ZnO crystal particle size from 200-500 nm to 24-71 nm could improve the absorbent dynamic capacity by a factor of 10. Comparison of the Engelhard and Nanotek samples with similar ZnO crystal particle size but different morphologies shows that changing from an elongated morphology to small flat plates¹ could increase the dynamic capacity of ZnO by a factor of >3. These comparisons are semiquantitative, because the samples were from different sources and formed using different preparation methods. However, these results clearly establish that the dynamic capacity of wash-coated ZnO for H₂S capture is affected by the particle size and morphology of ZnO particles. This is consistent with our previous results for particulate ZnO samples.1

Figure 4 illustrates the influence of inlet H₂S concentration on the dynamic capacity of wash-coated ZnO monoliths. The capacity of the monolith coated with coarse particles (AZO-66) decreased from 0.116 g S/100 g ZnO at 8 ppmv of H₂S to 0.019 g S/100 g ZnO at 0.5 ppmv H₂S (see Figure 4). The dynamic capacity of the monolith that contained Engelhard ZnO was also dependent on inlet H₂S concentration at 300 °C and SV =

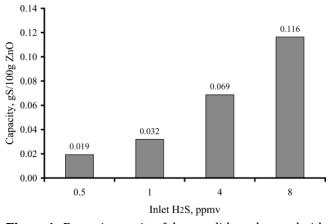


Figure 4. Dynamic capacity of the monolith wash-coated with AZO-66 ZnO at different H2S inlet concentrations. Conditions: 400 °C; 37% H₂, and 13% H₂O, with the balance being N_2 ; SV = 11 000 h^{-1} ; ZnO loading of 0.98–1.13 g/in.³.

3468 h⁻¹. The capacity decreased from 2.96 g S/100 g ZnO at 8 ppmv of H₂S to 0.89 g S/100 g ZnO at 1 ppmv of H₂S. Generally, the capacity is higher at higher H₂S inlet concentrations. In our recent work, the capacity of the 3-mm extrudates was 0.08 g S/100 g ZnO (Engelhard ZnO) at 1 ppmv of inlet H_2S , $SV = 8775 h^{-1}$ and 0.14 g S/100 g ZnO at 8 ppmv of H_2S , SV = 8775h⁻¹ at 300 °C.¹ The response of the wash-coated monolith to inlet concentration is similar to the case with the particulate samples, but the inlet H₂S concentration impacts the dynamic capacity more in the case of washcoated monoliths than in the case of particulate samples.

The following questions arise: (i) Why does a higher inlet H₂S concentration lead to a higher capacity of ZnO for H₂S capture, and (ii) why does the inlet H₂S concentration have a greater impact on the wash-coated ZnO monolith than in the case of particulate ZnO? The answer to the first question is that the higher inlet concentration favors the diffusion of H2S into the interior pores of the ZnO absorbent, which leads to higher utilization of the internal surface related to the geometric surface of the particles. On the other hand, at a lower inlet concentration, more of the H₂S would be reacted at the ZnO particle surface, resulting in a dense ZnS layer that envelopes ZnO and prevents subsequent diffusion of gas molecules into the bulk. To answer the second question, one must consider the difference in flow characteristics between the monolith and the packed bed of particulate.

The use of a fixed bed allows more ZnO-based absorbent particulate within a given volume. The length of the gas diffusion path is shorter when the particle size becomes smaller for a given material; however, a smaller particle size can lead to a greater decrease in pressure across the bed. Certain packing patterns may help to reduce the H₂S concentration gradient between the bulk phase and the gas film that surrounds the particle. On the other hand, wash-coating onto the

⁽⁵⁾ Hatori, M.; Sasaoka, E.; Uddin, M. A. Ind. Eng. Chem. Res. 2001, 40, 1884-1890.

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	Dynamic Capacity				
temp, T(°C)	$SV = 3468 h^{-1}$	$SV = 6936 \ h^{-1}$	$SV = 11000 \text{ h}^{-1}$	$SV = 23400 \ h^{-1}$	$SV = 46800 \ h^{-1}$
300 400	2.96	2.55	0.134	0.055	0.047

 $^{\it a}$ Conditions were as follows: 8 ppmv $H_2S,\,34.4\%$ $H_2,$ and 20% $H_2O,$ with the balance being $N_2.$

monolith allows the use of smaller ZnO particles for treating a gas flow without a decrease in pressure. When the absorbent is used in the form of wash-coated monoliths, the total flow is evenly distributed among the channels of a monolith. In our case, the ratio of channel size to channel length was 1:34.2. The flow was laminar, because the Reynolds number was 98 and 56 at 300 and 400 °C, respectively, which is much lower than the Reynolds number (2100) that corresponded to the border region for the flow through a circular tube.9 Turbulent flow, which may be caused by molecules that are encountering an obstacle (such as monolith walls), could occur only in the front portion of the monolith, equalizing the H2S concentration between the bulk and surface gas layers. In this case, the reaction rate would be more likely limited by H₂S diffusion into the bulk of unreacted ZnO through a product (ZnS) layer. When the flow is stabilized in the later (and tail) part of a monolith channel, gas-film diffusion would begin to have a more significant role. In this case, the concentration gradient becomes more influential. These differences in flow characteristics are the possible reasons why H₂S concentration has a relatively greater impact on the performance of the wash-coated ZnO samples.

Table 4 shows the effect of space velocity on the sample performance for the feed with the higher steam concentration (20% H₂O, vs 13% H₂O for data in the other tables). The increase in space velocity leads to turbulent growth in the flow through a monolith channel. With the present case of the monolith (19.05 mm in diameter and 38 mm in length), the flow through the channel will become completely turbulent at a flow rate of 72-75 L/min. Thus, in all our experiments with a monolith 19.05 mm in diameter and 38 mm in length, the flow rate was much smaller than this value; therefore, the flow was not turbulent in the monolith channel. It is laminar at the later part and the tail end of the channel. To increase the space velocity, the monoliths were cut into smaller pieces (9 mm in length). In this case, the ratio of channel length to channel size is not enough to form a laminar flow and fluxes must occur. Probably for this reason, the influence of space velocity on the dynamic capacity is not as significant as in the case with a fixed bed of particulate absorbents. This is an advantage of the monolith, compared to the case of a fixed bed of particulate ZnO, because it gives more flexibility in adjusting the space velocity without major changes in the dynamic capacity. Nevertheless, a lower capacity was observed at higher space velocities for the wash-coated ZnO monolith.

Monoliths have several special characteristics that the particulate ZnO samples do not have. They are (i) the

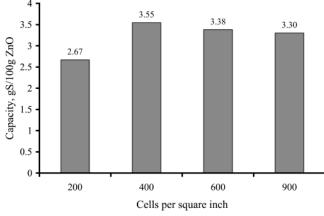


Figure 5. Influence of the number of cells per square inch (CPSI) on the dynamic capacity of the monolith wash-coated with modified ZnO from Engelhard. Conditions: 400 °C; 37% H₂ and 13% H₂O, with the balance being N₂; SV = $11~000~h^{-1}$; loading of $0.6-1.00~g/in.^3$.

Table 5. Dynamic Capacity and Breakthrough Time of Monoliths Wash-Coated with Engelhard ZnO^a

ZnO loading (g/in³)	breakthrough time, BT (h)	capacity (g S/100 g ZnO)
0.55	0.8	1.45
0.77	0.73	0.95
0.93	1	1.07
1.15	1.3	1.13

 a Conditions were as follows: 400 °C; 37% H_{2} and 13% $H_{2}O,$ with the balance being $N_{2};\,SV=46\;800\;h^{-1}.$

number of cells per square inch (CPSI) of the cross section, and (ii) the surface loading of the active component per unit volume (in grams per cubic inch, g/in.³) of monolith. They can also affect the capacity of wash-coated absorbents. Figure 5 shows the effect of CPSI on the dynamic capacity of wash-coated ZnO, and Table 5 shows the influence of ZnO loading at higher space velocity (SV = 46 800 h $^{-1}$ vs 11 000 h $^{-1}$ for data in the other tables). Increasing the CPSI from 200 to 400 significantly increases the dynamic capacity from 2.67 g S/100 g ZnO to 3.55 g S/100 g ZnO. However, further increasing the CPSI to 600 and 900 caused a slight decrease in the dynamic capacity. The best performance was achieved using 400 CPSI monoliths.

Finally, note that different batches of monoliths that have been wash-coated with ZnO can give somewhat different capacity values, because of some minor differences in ZnO pretreatment, particle milling, monolith dimension, wash-coating, absorbent loading, and calcination temperature, because of the exploratory nature of this work in the laboratory. However, the trends observed clearly demonstrate the suitability of monoliths for wash-coating very fine ZnO particles to make a H₂S absorbent with high dynamic capacity without a decrease in pressure.

Conclusions

This work is an experimental study on H_2S capture from steam-containing gas mixtures with low H_2S concentrations using wash-coated monolith absorbents for fuel cell applications, and the work has led to the following conclusions:

(1) For the first time, wash-coated monolithic absorbents of different types have been comparatively studied

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for H₂S capture at low H₂S concentrations (in the range of 0.5-8 ppmv), low temperature (in the range of 300-400 °C), and in the presence of steam (13–20 vol % H₂O) at medium-to-high space velocities (3468–46 800 h⁻¹). The use of wash-coated monoliths allows one to achieve greater dynamic capacity for H_2S capture with little or no decrease in pressure, as compared to a fixed bed of particulate absorbents.

- (2) A comparison of the various materials tested showed that ZnO (without any other additives, such as copper or alumina) exhibited the best performance in the form of a wash-coated monolith.
- (3) The morphology and size of the ZnO particles each have great impact on the dynamic capacity of a washcoated monolith at similar ZnO loading. ZnO that had been modified by a ammonium carbonate treatment (which shows a flat, platelike morphology) gave the best performance.
- (4) The inlet H₂S concentration impacts the dynamic capacity more in the case of wash-coated monoliths than in the case of a fixed bed of particulate samples. Space velocity also has an impact on the capacity of the washcoated monolith; however, the effect seems to be less, compared to the fixed-bed case.

(5) The monolith with 400 cells per square inch (CPSI) showed better performance for wash-coated ZnO, in regard to H₂S capture, compared to the monoliths with a lower or higher number of cells (200, 600, and 900 CPSI).

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Note Added after ASAP Posting. This article was released to the Web on 2/12/2004, before all the author corrections were received and incorporated. (Specifically, the dynamic capacity of the Engelhard sample in Table 3 has been corrected and the in-text mentions of Figure 5 and Table 5 have been reworded.) The corrected version was posted on 2/20/2004.

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