

lyst as a function of contact time and temperature. At temperatures in the range of 240-270° C. and a contact time of 120 ms, less than 50 ppm CO was achieved.

Steam Reforming Catalyst Testing

For comparison purposes, a supported Pd—ZnO catalyst was fabricated via the formation of zinc oxide on a γ -alumina support by the precipitation of zinc hydroxide from a zinc nitrate solution with ammonia at pH about 8 and calcination at 350° C., followed by Pd incipient wetness impregnation. The catalysts prepared by this precipitation route are hereafter referred to the baseline catalyst.

Other supported Pd—ZnO catalysts were prepared by a one-step co-impregnation method. In this method, a concentrated palladium nitrate solution (containing about 20 wt % Pd in nitrate acid solution) was mixed with solid $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ at 50 to 80° C. in order to obtain a solution containing Pd and Zn as concentrated as possible. The ZnO/Pd ratio in the resultant solution was varied from 0.7 to 25 in order to obtain the final products having Pd loadings of 1 to 15 wt %. A given support was impregnated at 50 to 80° C. with the amount of solution adjusted according to the pore volume of the support. The wet sample was kept at 60° C. at least one hour that allowed completing the impregnation process. The samples were dried in air and then calcined at 350° C. for 3 hours. A series of Pd—ZnO/ Al_2O_3 with varying Pd loadings using the one-step co-impregnation method have been fabricated. In order to investigate the support effects, ZrO_2 and Ce_2O_3 supported Pd—ZnO catalysts were also be made.

A typical Pd—ZnO/ Al_2O_3 using the one-step co-impregnation method for fabricating is described as follows.

1. A mixture containing 13.10 g of $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 1.773 g of concentrated Pd nitrate solution (20.19 wt Pd) was heated at 60° C. in a water bath till the solids dissolved.
2. A 2.00 g of $\gamma\text{-Al}_2\text{O}_3$ powder with 60-120 mesh (preheated at 500° C. for 2 h) was impregnated with 3.077 g above solution in a glass vial and kept at 60° C. for at least one hour.
3. The wet sample was dried at 100° C. and then calcined at 350° C. at 20° C./min for 2 hours.

The final composition of the catalyst contained 2.6 wt % of Pd, 26 wt % of ZnO, and 70.2 wt % of Al_2O_3 . Other catalysts were fabricated in similar ways and their compositions are listed in Table 1.

TABLE 1

Composition of supported Pd/ZnO catalysts.				
Sample	Description	Composition, wt %		
		Pd	ZnO	Support
(baseline)	ZnO: Pd = 8, on Al_2O_3	8.6	70	21
PdZnAl-10	ZnO: Pd = 10, on Al_2O_3	2.6	26.4	71.0
PdZnAl-4.5	ZnO: Pd = 4.5, on Al_2O_3	5.0	22.8	72.2
PdZnAl-2	ZnO: Pd = 2.0, on Al_2O_3	8.9	17.8	73.3
PdZnAl-1	ZnO: Pd = 1.0, on Al_2O_3	12.9	13.0	74.1
PdZnAl-0.7	ZnO: Pd = 0.7, on Al_2O_3	15.7	10.9	73.4
PdZnZr-10	ZnO: Pd = 10, on ZrO_2	1.9	19	79.1
PdZnCe-10	ZnO: Pd = 10, on Ce_2O_3	1.4	14	84.6

The steam reforming catalysts were tested in the fixed-bed quartz tubular reactor with a 4 mm I.D. In each case, the packed catalysts were reduced at a hydrogen-containing gas at 400° C. for 3 hours prior to reaction. The feed liquid consisted of methanol and water at a weight ratio of 1. In most cases, about 0.192 g of the catalyst was used at a feed rate of 2 ml/h.

Table 2 list some results for steam reforming of methanol over selected catalysts fabricated according the reported method. The results show that all the examined catalysts, except for the Ce_2O_3 supported catalyst, demonstrate the high activity (80% conversion of methanol at below 300° C.) and very low CO selectivity (less 0.8% CO in the dry product steam) while retaining fairly high H_2 productivity.

TABLE 2

Reaction results at a 80% of methanol conversion for supported Pd—ZnO.				
Results at Conversion of 80%				
Sample	T ₈₀ [° C.]	Selec. of CO ₂ %	% CO in Dry Product Stream	H ₂ Productivity [Nm ³ /kg cat · h]
(baseline)	297	97.7	0.52	8.2
PdZnZr-10	295	97.8	0.80	8.2
PdZnCe-10	305	86.0	3.60	2.7
PdZnAl-10	275	97.8	0.58	8.9
PdZnAl-4.5	265	98.3	0.42	7.3
PdZnAl-2	250	98.6	0.36	8.5

With a similar amount of Pd content, the catalyst (PdAlZn-2, 8.9 wt % Pd) which was made by the co-impregnation method exhibits much higher activity than that of the baseline catalyst (8.6 wt % Pd), which was synthesized by a precipitation and impregnation process. At the 80% conversion of methanol, the reaction temperature difference is about 50° C. Conversion of methanol over PdAlZn-2, PdZnAl-4.5, PdZnAl-10, and the comparative baseline, as a function of temperature is shown in FIG. 10. As can be seen in the figure, the coprecipitated catalyst exhibits significantly superior activity over a broad range of temperatures.

Furthermore, over the catalysts supported by different substrates, such as Al_2O_3 , ZrO_2 , and Ce_2O_3 prepared using the improved method, the H_2 productivities of steam reforming of an alcohol (in this case, methanol) are much higher than that of the baseline catalyst. The results are shown in FIG. 11.

FIG. 12 shows the conversion of methanol and selectivity to CO at varying Pd loadings (wt %) and Pd to Zn ratios at 260° C. Surprisingly, it was observed that alcohol conversion decreased with increasing Pd levels above about 9% and above a Pd:Zn ratio of about 0.4.

CLOSURE

While preferred embodiments of the present invention have been shown and described, it will be apparent to those skilled in the art that many changes and modifications may be made without departing from the invention in its broader aspects. For example, although devices are shown with one combustor and one reformer, numerous variations such as two combustors sandwiching one reformer, and these variations are included within the scope of the invention. The appended claims are therefore intended to include all such changes and modifications as fall within the true spirit and scope of the invention.

We claim:

1. A steam reformer comprising: a first reaction chamber connected to a first inlet and a first outlet; a second reaction chamber connected to a second inlet and a second outlet; the first reaction chamber and the second reaction chamber being in thermal communication, wherein the first reaction chamber comprises a combustion catalyst or a steam reforming catalyst, wherein, if the first reaction chamber comprises a combustion catalyst, the second reaction chamber comprises