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Catalytic Decomposition of SO₃ over Pt/BaSO₄ Materials in Sulfur–Iodine Cycle for Hydrogen Production

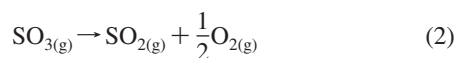
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Various platinum-supported barium sulfate catalysts have been employed for SO₃ decomposition in sulfur–iodine cycle to produce hydrogen. Before impregnation of platinum, barium sulfate as a support has been prepared by different methods, i.e., using ethylene glycol (EG) and water by dispersion method and EG and propanol by spray pyrolysis. The influence of the supports on catalytic performance was examined with three catalysts: 1.0Pt/BaSO₄ (PrOH), 1.0Pt/BaSO₄ (EG), and 1.0Pt/BaSO₄ (H₂O) at various temperatures for SO₃ decomposition. 1.0Pt/BaSO₄ (PrOH) prepared by spray pyrolysis showed high SO₃ conversion (62%) at low temperature (1023 K) compared with the other catalysts prepared by dispersion method. The effect of Pt loading on catalytic activity has been performed with BaSO₄ impregnated by various amounts of platinum. Under identical conditions, the 1.0Pt/BaSO₄ catalyst showed the highest catalytic performance because of high surface area and small particle size of platinum.

1. Introduction

Hydrogen production by water using a renewable energy power supply is a desirable solution by sulfur–iodine cycle. In H₂SO₄ decomposition SO₃ decay is a major step for H₂ production and breaking of SO₃ is a difficult task. Sulfur-based thermochemical cycles including the sulfur–iodine cycle,¹ hybrid sulfur cycle,² and sulfur–bromine hybrid cycle³ incorporate the sulfuric acid decomposition reaction as the high-temperature steps in their cycles. Sulfuric acid thermal decomposition is normally carried out at about 1023–1223 K in a process reactor that can withstand high-temperature heat from a nuclear supplying system. Usually, reaction is divided into two subreactions:



The sulfuric acid decomposition reaction takes place in two steps, a noncatalytic thermal decomposition of the acid to form gaseous SO₃ and H₂O at temperatures above 673 K, followed by a catalytic decomposition of the SO₃ to produce the SO₂ and O₂ products. At reasonable operating temperatures, between 873 and 1223 K the reduction of SO₃ does not take place without a catalyst. Most metal oxides had low activity due to the formation of sulfates on the catalysts.^{4–6} The preparation of catalyst, metal precursors, and supports has an important role for SO₃ decomposition. Ginosar et al. reported SO₃ decomposition using platinum-supported Al, Zr, and Ti at higher temperature.⁷ Kim et al. reported the catalytic decomposition SO₃ on the binary metal oxide catalysts.⁸ The formation of sulfates was more significant at lower reaction temperatures and was predicted to be problematic as reaction pressure increased to

Table 1. BET Surface Area (m²/g) of Dried and Calcined BaSO₄ Materials Prepared by Different (Dispersion and Spray Pyrolysis) Methods and Then 1 wt % Platinum Impregnation

catalysts		dispersion method		
		ethylene glycol (EG)	water	spray pyrolysis
BaSO ₄	dried	91.56	14.50	72.52
	calcined	13.52	13.70	18.13
1.0Pt/BaSO ₄	dried	12.53	13.87	14.19
	calcined	10.77	10.87	12.60

anticipated operating values. In addition, catalysts were found to fail due to formation of volatile acid salts,⁵ supporting poisoning.⁵ Although a few catalysts appeared promising under ideal conditions, primarily platinum supported on porous metal oxides, their long-term stability has not been reported.⁷ Abimanyu et al. reported Fe/Cu/Al₂O₃ composite granules catalysts

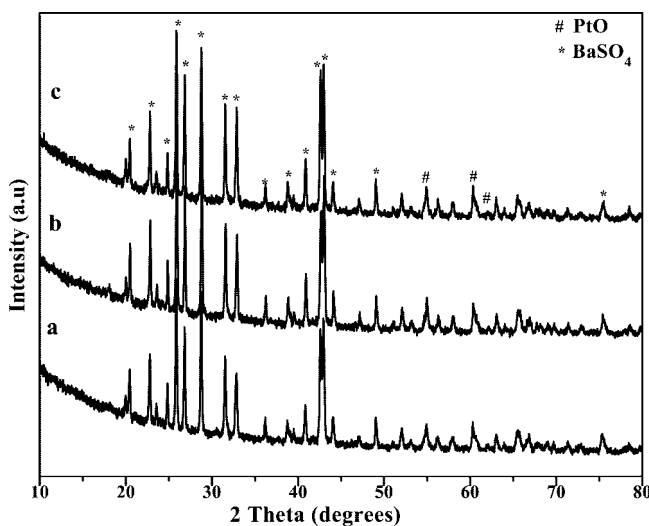


Figure 1. XRD patterns of Pt supported on BaSO₄ nanomaterial calcined at 700 °C prepared by dispersion and spray pyrolysis method: (a) 1.0Pt/BaSO₄ (EG), (b) 1.0Pt/BaSO₄ (H₂O), and (c) 1.0Pt/BaSO₄ (PrOH) catalysts.

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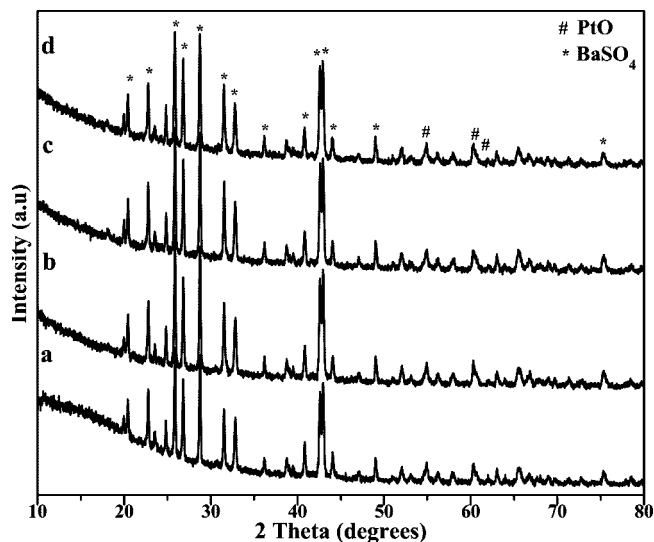


Figure 2. XRD patterns of calcined spray pyrolysis catalysts (BaSO_4 prepared by spray pyrolysis method) with different Pt loadings: (a) 0.5Pt/ BaSO_4 (PrOH), (b) 1.0Pt/ BaSO_4 (PrOH), (c) 1.5Pt/ BaSO_4 (PrOH), and (d) 2.0Pt/ BaSO_4 (PrOH).

were prepared by a combination of Yoldas sol–gel process and oil drop method for SO_3 decomposition to assist hydrogen production.⁹ Catalytic decomposition of sulfuric acid on mixed Cr/Fe oxide samples and its application in sulfur–iodine cycle for hydrogen production is reported by Banerjee et al.¹⁰

Barium sulfate is one of the most important fillers used in the plastics, rubber, and paint industries and is also used in

Table 2. Properties of Dried and Calcined Pt/ BaSO_4 Catalysts with Different Platinum Loadings

catalysts	BET surface area (m^2/g)		pore volume (cm^3/g)		average pore diameter (nm)	
	dried	calcined	dried	calcined	dried	calcined
0.5Pt/ BaSO_4 (PrOH)	10.19	10.33	0.0137	0.00742	5.36	2.87
1.0Pt/ BaSO_4 (PrOH)	14.19	12.60	0.0765	0.00707	2.17	2.67
1.5Pt/ BaSO_4 (PrOH)	12.05	11.25	0.0081	0.00509	2.56	2.81
2.0Pt/ BaSO_4 (PrOH)	11.12	12.79	0.0094	0.01203	3.39	2.76

pharmaceutical formulations. Recently, barite was incorporated into polymers to form electrical-insulating barite/epoxy composites with X-ray attenuation properties.¹¹ Li and Yuan reported the performance of barium sulfate nanotube arrays in supporting sulfates for low-temperature methane activation.^{12,13} In previous work we reported the high surface area mesoporous BaSO_4 prepared by the dispersion method.¹⁴

Spray pyrolysis is a useful method for producing multicomponent, high-purity powders that consist of nonagglomerated sub-micrometer-sized particles.¹⁵ We reported the novel method for the preparation of mesoporous BaSO_4 material with thermal stability by spray pyrolysis.¹⁶ Spray pyrolysis is a useful tool for large- or small-scale production with controlled particle size. The properties of the final product can be controlled through the choice of precursor and solution concentration or by changing of the conditions of aerosol decomposition. The micrometer-sized droplets of a precursor solution are dried and pyrolyzed in a furnace to form particles.^{17,18}

In the present work, we examined the catalytic performance of platinum supported on BaSO_4 prepared by dispersion and

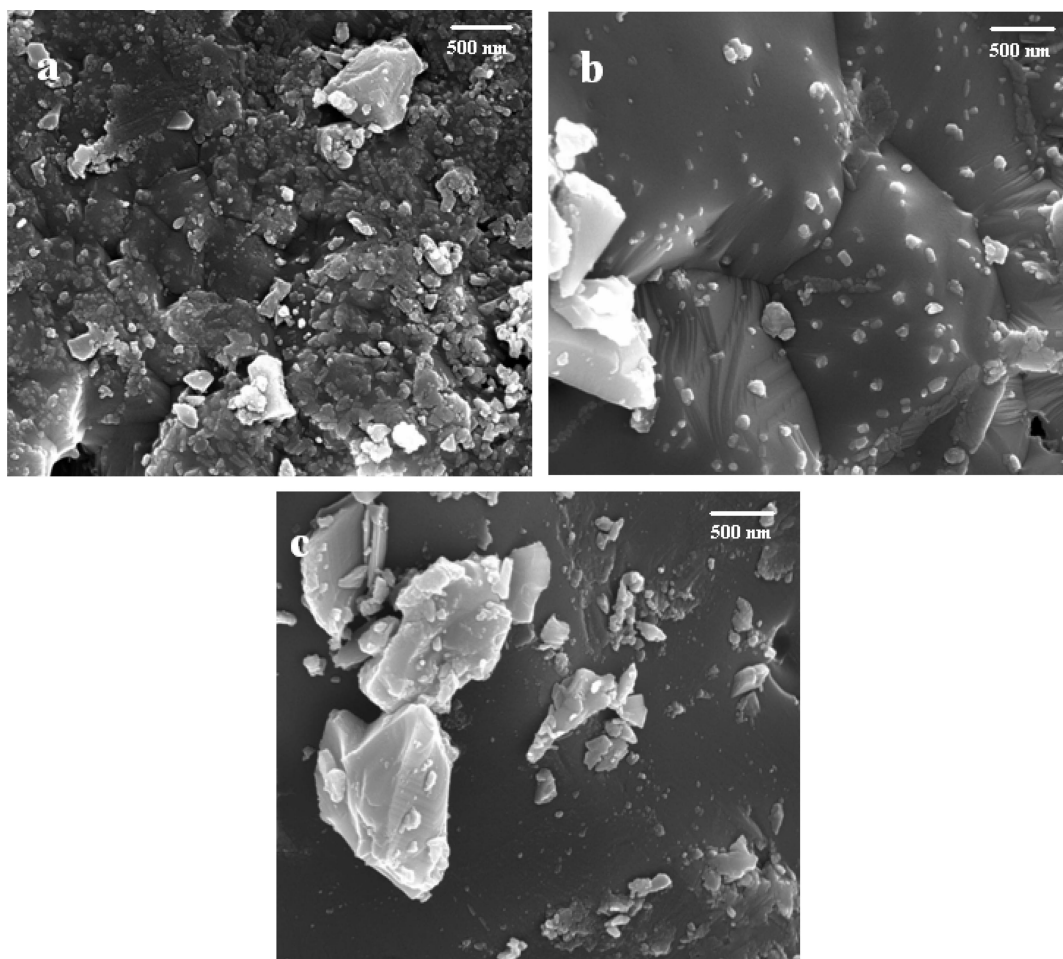


Figure 3. Scanning electron microscopy of 1.0Pt/ BaSO_4 calcined catalysts: (a) 1.0Pt/ BaSO_4 (PrOH), (b) 1.0Pt/ BaSO_4 (EG), and (c) 1.0Pt/ BaSO_4 (H_2O).

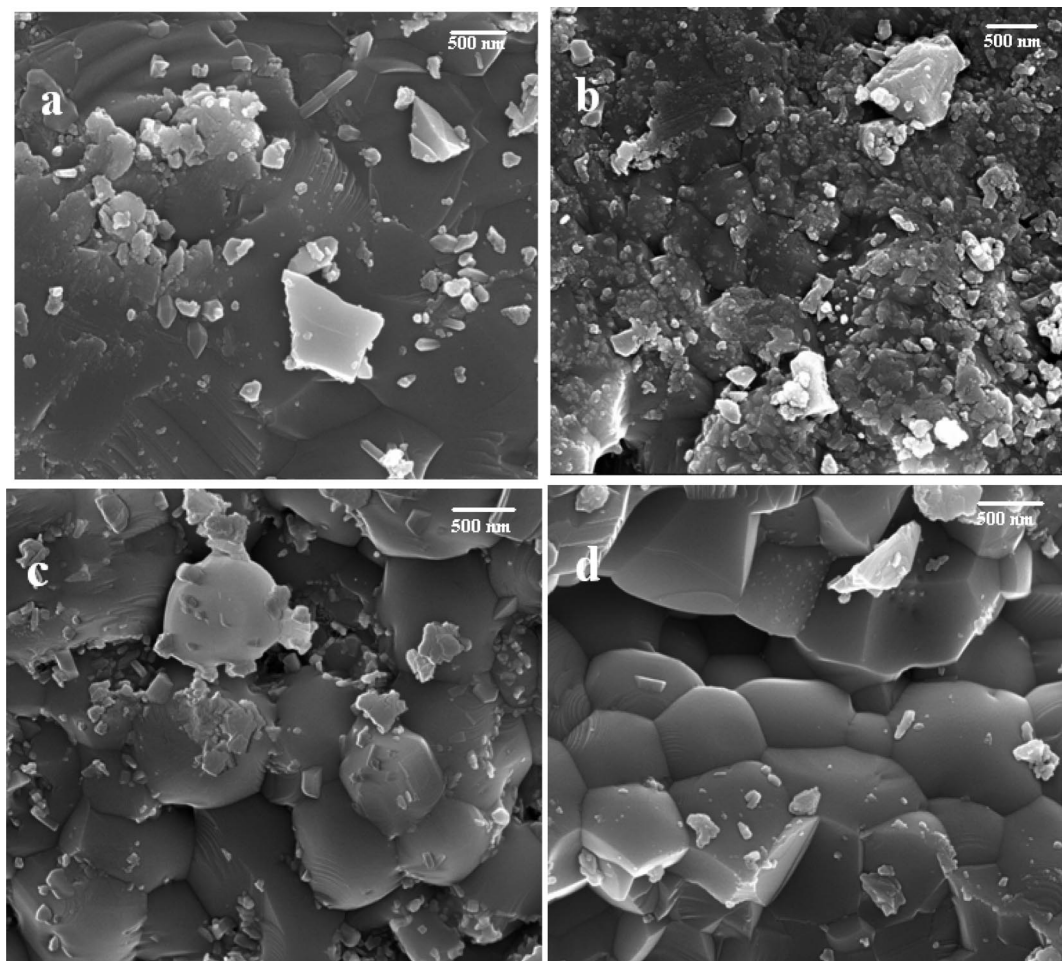


Figure 4. SEM images of calcined catalysts with different Pt loadings and BaSO₄ prepared by spray pyrolysis: (a) 0.5Pt/BaSO₄ (PrOH), (b) 1.0Pt/BaSO₄ (PrOH), (c) 1.5Pt/BaSO₄ (PrOH), and (d) 2.0Pt/BaSO₄ (PrOH).

spray pyrolysis methods. Propanol was used as a solvent for the preparation of BaSO₄ by spray pyrolysis because solvents have good inhibition effect for controlling the particle size. The catalytic activity measurement of these catalysts has been carried out in a gas-phase SO₃ reaction with temperatures of 873–1223 K.

2. Experimental Section

2.1. Preparation of Catalyst. The BaSO₄ nanomaterial was prepared by dispersion method using ethylene glycol (EG) and water as a precursor solution.¹⁴ In spray pyrolysis the BaSO₄ solution was dissolved in propanol and this solution was used for spray pyrolysis.¹⁶ This BaSO₄ powder material was calcined at 1173 K for 2 h. Platinum supported on BaSO₄ was prepared by impregnation method with different loadings, viz., 0.5, 1.0, 1.5, and 2 wt % of Pt. Finally, this catalyst was dried at 383 K for 12 h in air and calcined at 973 K for 2 h.

2.2. Characterization. The XRD patterns of platinum-supported BaSO₄ nanomaterials were recorded on a diffractometer (M/S, Shimadzu Instruments, Japan) with Ni-filtered Cu K α as a radiation source and at a 2θ scan speed of 2°/min. The N₂ adsorption–desorption measurements were performed at 77 K using an automated gas sorption system (M/S, Micromeritics ASAP 2000) utilized with Brunner-Emmett-Teller (BET) calculation for the surface area and Barrett-Joyner-Halanda (BJH) method for pore size distribution. The sample was pretreated at 473 K for 2 h before analysis. Scanning electron microscopy (SEM), an important characterization tool in this work, was

conducted by Nova 200 (image resolution, 1.0 nm; acceleration voltage, 200–300 kV; magnification, 20–100000 \times). FEI TecnaiF20, a high-resolution transmission microscope (HRTEM), was used (image resolution, 0.24 nm; accelerating voltage, 50–300 kV; magnification, 25–1030000 \times ; electron probe, 0.3 nm) using a Schottkey FEG electron gun to visually characterize Pt nanomaterials.

The CO chemisorption was carried out at 303 K using Micromeritics Autochem II instrument to determine the metal surface area. About 200 mg of the catalyst sample was placed in a U-tube microreactor of 8 mm i.d. and 250 mm long quartz reactor and the catalyst sample was first reduced under a 10% H₂/Ar flow at 723 K for 2 h (10 °C/min), pretreated at 723 K for 2 h under He flow, and finally cooled in He flow to 303 K. After cooling, pulses of 10% CO, balance He were injected at room temperature through a 1 mL loop connected to the six-port valve until no further change occurred in the intensity of the outlet CO.

2.3. Activity Measurement. The catalytic activity for the SO₃ decomposition was conducted in a fixed bed quartz reactor at atmospheric pressure. The mixture of SO₃ (20 cm³/min) and N₂ (40 cm³/min) was fed into the reactor and the gas hourly space velocity (GHSV) in the catalyst bed was calculated to be about 72000 cm³/(g cat·h). The product gases were analyzed by titration and absorption methods for SO₂ and SO₃ reported by Kim et al.⁸ The reaction was carried out in the temperature range of 873–1223 K at atmospheric pressure and 0.050 g of catalyst was used for the reaction.

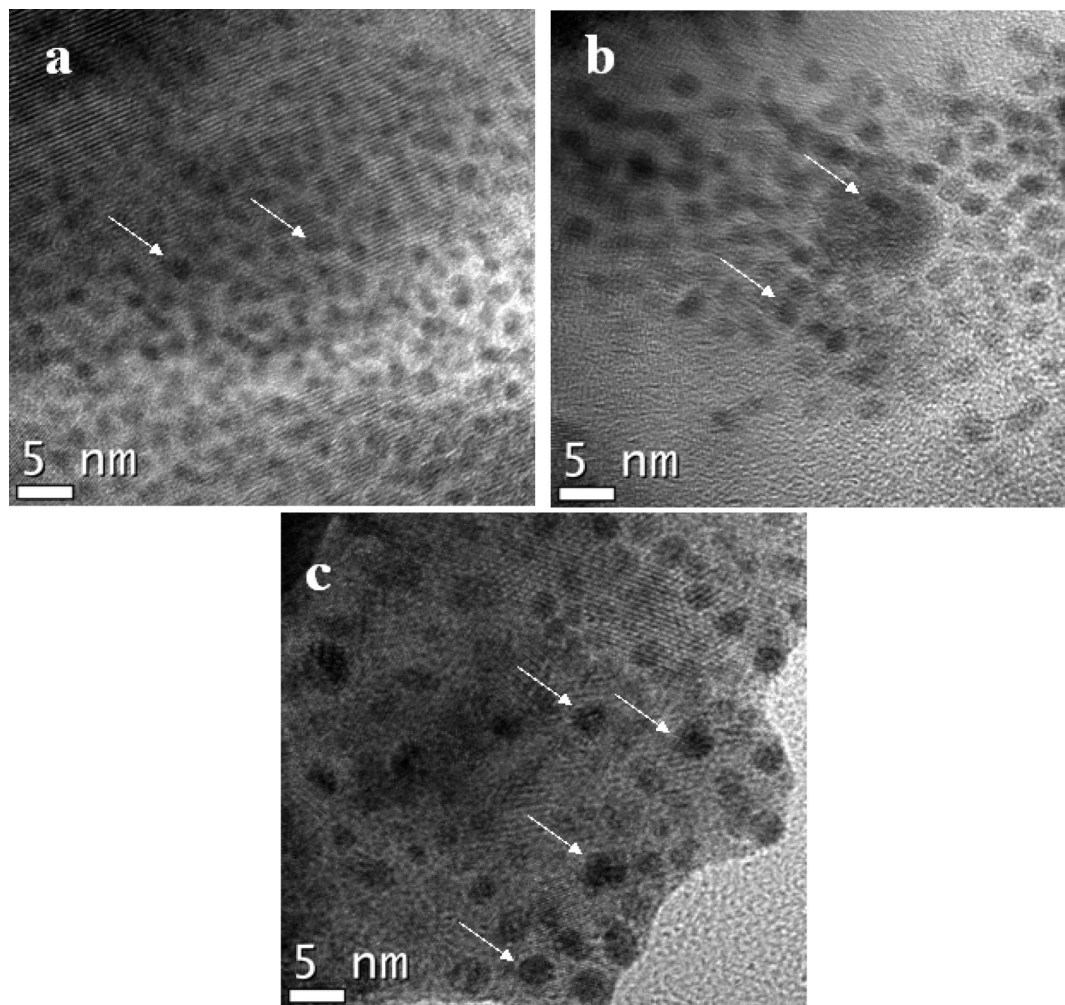


Figure 5. High-resolution transmission electron micrographs of (a) 1.0Pt/BaSO₄ (PrOH), (b) 1.0Pt/BaSO₄ (EG), and (c) 1.0Pt/BaSO₄ (H₂O) catalysts.

3. Results and Discussion

3.1. Characterization of the Catalysts. The surface areas of dried and calcined BaSO₄ samples prepared by different methods and 1 wt % platinum impregnated on BaSO₄ are presented in Table 1. The surface area of dried BaSO₄ prepared with ethylene glycol and H₂O by dispersion methods was 91.56 and 14.50 m²·g⁻¹, respectively. After calcination at 1173 K, the surface area was 13.52 and 13.70 m²·g⁻¹, respectively. In our previous work, the BaSO₄ materials were prepared by spray pyrolysis using different additives, i.e., methanol, ethanol, propanol, and butanol.¹⁶ The BaSO₄ material prepared at 1073 K in spray pyrolysis using propanol as the additive shows the highest surface area. Propanol was used as a solvent in spray pyrolysis because it has a good inhabitation effect and controls the particle size.¹⁹ In this study, the BaSO₄ by spray pyrolysis possessed the highest surface area after calcination.

For the catalytic reaction, 1wt % platinum was impregnated on BaSO₄ prepared with various methods and then calcined at 973 K. The surface area of BaSO₄ prepared by spray pyrolysis after impregnation (1.0Pt/BaSO₄ (PrOH)) is higher than that of BaSO₄ prepared by dispersion method using water (1.0Pt/BaSO₄ (H₂O)) and ethylene glycol (1.0Pt/BaSO₄ (EG)). These results indicate that the method of preparation of BaSO₄ using solvents had more effect on the bulk physical structure of the catalysts. Table 2 shows the surface areas, pore volumes, and pore sizes of dried and calcined catalyst samples with different platinum loadings impregnated on BaSO₄ prepared by spray pyrolysis.

The amount of platinum loadings mainly affected the pore volume of the catalyst.

The XRD patterns of the 1 wt % platinum-supported BaSO₄ materials prepared by dispersion method using ethylene glycol (EG) or H₂O and spray pyrolysis using propanol as the solvent are shown in Figure 1. All the samples were calcined at 973 K. The *d*-values of the BaSO₄ material are 4.34_x, 2.12₅, and 3.90₄ (JCPDS No. 83-1718), 3.44_x, 3.10_x, and 2.12₇ (JCPDS No. 83-2053) with *hkl* values of (101), (311), (111) and (210), (211), (401), and 1.96_x and 1.38₂ (JCPDS No. 87-0646) and the *d*-values of PtO are 1.67₃, 2.13₅, 1.53_{2,5}, and 1.49_{0,5} (JCPDS No. 27-1331). The XRD patterns of different catalysts with Pt loadings of 0.5, 1.0, 1.5, and 2.0 wt % of Pt supported on BaSO₄ are shown in Figure 2. The XRD patterns show PtO and BaSO₄ phases in all the catalysts.

The SEM images show the morphologies of calcined Pt/BaSO₄ particles. Figure 3 shows the particle distribution of platinum impregnated on BaSO₄ prepared by different methods. Figure 3a shows the spherical-shaped BaSO₄ nanoparticles prepared by spray pyrolysis using propanol as an additive. The platinum distribution is uniform and the BaSO₄ nanoparticles are agglomerated in a regular manner. The image (Figure 3a) revealed that the platinum dispersed on BaSO₄ had relatively good crystallographic orientation, suggesting the establishment of a strong metal–support interaction. Figure 3, parts (b) and (c), shows the morphology of Pt support on BaSO₄ using EG and H₂O by dispersion method. In these images the platinum

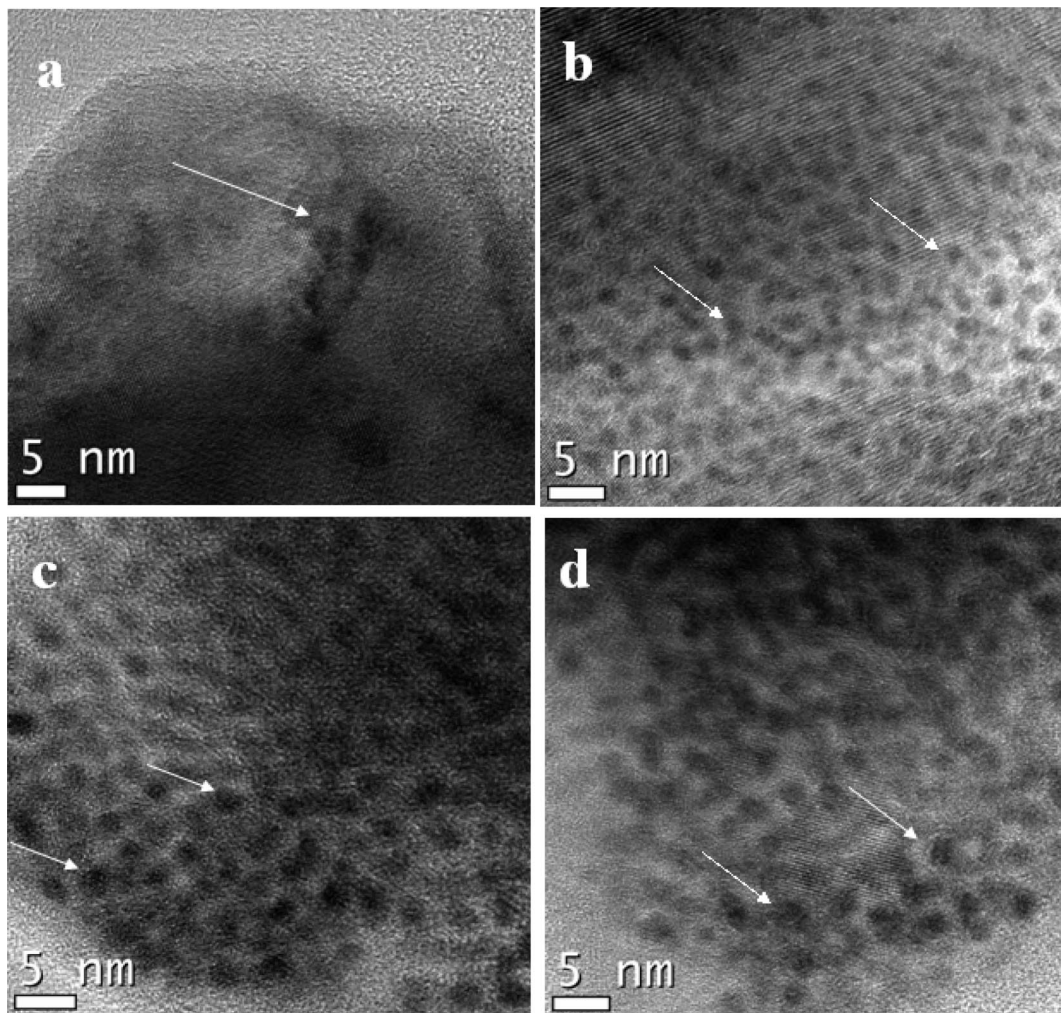


Figure 6. HRTEM images of calcined catalysts with different Pt loadings: (a) 0.5Pt/BaSO₄ (PrOH), (b) 1.0Pt/BaSO₄ (PrOH), (c) 1.5Pt/BaSO₄ (PrOH), and (d) 2.0Pt/BaSO₄ (PrOH).

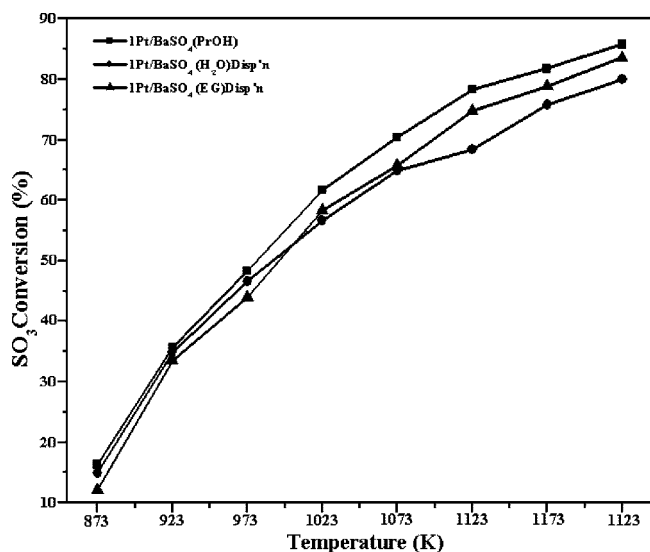


Figure 7. Activity of 1 wt % Pt supported on BaSO₄ with various methods (BaSO₄ prepared by dispersion and spray pyrolysis method) for SO₃ decomposition. Reaction conditions: catalyst, 0.050 g; GHSV, 72000 cm³/g cat·h).

and BaSO₄ become agglomerated completely in the form of bulk and the particle distribution was not uniform; there is no proper interaction between the metal and support.

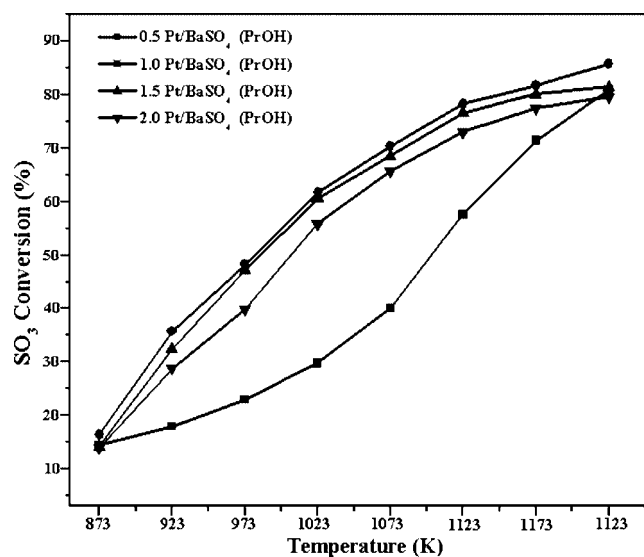


Figure 8. Effect of platinum loading supported on BaSO₄ for SO₃ decomposition. Reaction conditions: catalyst, 0.050 g; GHSV, 72000 cm³/g cat·h).

Figure 4 shows the SEM images of Pt/BaSO₄ with different loadings and 1.0Pt/Al₂O₃ commercial catalyst. Figure 4b shows that the 1 wt % Pt support on BaSO₄ was uniform and the platinum nanoparticles are deposited systematically with BaSO₄

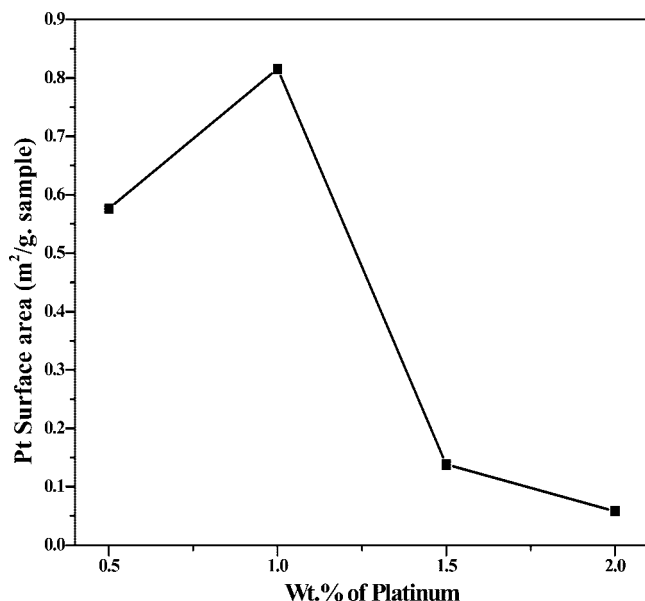


Figure 9. Platinum surface area of the catalyst.

particles. The uniform dispersibility of the Pt in BaSO₄ is because of the use of propanol as an additive for the preparation of BaSO₄ nanoparticles. The formation of the mesoporous structure is related to both nucleation and the growth mechanism of the BaSO₄ particles. In Figure 4, parts (c) and (d) (1.5 and 2 wt % Pt), the particles are in a hexagonal-like structure because the platinum was agglomerated with BaSO₄. This is because the dispersion of Pt on BaSO₄ is not uniform in higher loadings. It is well-known that high surface area (Table 1) of BaSO₄ support would help to increase platinum dispersion and effectively prevent extensive growth and aggregation of platinum particles.

Figure 5 shows HRTEM images of 1.0Pt/BaSO₄ catalysts prepared with different methods. It can be observed from Figure 5a that the Pt particles are uniformly distributed with a narrow size on the BaSO₄ material. Figure 5, parts (b) and (c), presents the HRTEM images of the 1.0Pt/BaSO₄ catalysts prepared by dispersion method using ethylene glycol and water. The distribution of Pt on BaSO₄ was not uniform because with calcination at higher temperatures, the BaSO₄ particles become agglomerated and the interaction between Pt precursor and support may also affect the metal dispersion. Figure 6 shows the HRTEM images of Pt/BaSO₄ with different loadings of Pt on BaSO₄ material prepared by spray pyrolysis method. The average metal particle sizes were 12, 3, 7, and 15 nm for 0.5, 1.0, 1.5, and 2 wt % Pt, respectively. The particle size 1.0 wt % Pt supported on BaSO₄ shows smaller particles compared with the others.

3.2. Activity Studies. SO₃ decomposition was performed on 1 wt % Pt supported on BaSO₄ prepared by various methods. The SO₃ decomposition efficiency was strongly affected by the temperature as shown in Figure 7. Among the catalysts, 1.0Pt/BaSO₄ (PrOH) catalyst, the BaSO₄ prepared by spray pyrolysis method, showed the highest conversion, about 62%, at 1023 K. The 1.0Pt/BaSO₄ (EG) and 1.0Pt/BaSO₄ (H₂O) catalyst showed 58% and 56% conversion, respectively, the BaSO₄ prepared by dispersion method. Ginossar et al. reported the Pt-supported Al, Zr, and Ti in sulfuric acid decomposition: the SO₂ yield at 1073 K was approximately 45, 30, and 20% and at 1123 K it was 52, 48, and 25%, respectively.⁷ Kim et al. reported the catalytic decomposition of SO₃ on the binary metal oxide catalysts containing Fe/Al and Fe/Ti was ~20% conver-

sion at 1023 K.⁸ The results were mainly attributed to smaller particle size of 1.0Pt/BaSO₄ (PrOH) as shown in HRTEM (Figure 5) compared with the other catalysts. At a reaction temperature of 1223 K the decreasing order of SO₃ decomposition was 1.0Pt/BaSO₄ (PrOH) > 1.0Pt/BaSO₄ (EG) > 1.0Pt/BaSO₄ (H₂O). The metal dispersions of 1.0Pt/BaSO₄ (PrOH), 1.0Pt/BaSO₄ (EG), and 1.0Pt/BaSO₄ (H₂O) are 33.03, 3.70, and 2.50% measured by CO chemisorption. The structure of BaSO₄ prepared by dispersion methods was completely decayed and the surface area decreased drastically. Moreover, platinum particles were agglomerated at higher temperature as shown in Figure 5.

The BaSO₄ material prepared by spray pyrolysis was calcined at 1173 K and then different amounts of Pt were impregnated on BaSO₄. The effect of Pt loading on BaSO₄ for SO₃ decomposition is shown in Figure 8. The conversion of SO₃ was enhanced with an increase in Pt content up to 1 wt % and after that there is a small variation in conversion. 0.5Pt/BaSO₄ (PrOH) catalyst shows 40% conversion at 1023 K and 81% at 1223 K. The 1.0Pt/BaSO₄ shows about 62% conversion at 1023 K and finally it reaches 86% conversion at 1223 K. This is due to small Pt particle size and uniform platinum dispersion on the surface of the support. This is due to small particle size and uniform platinum dispersion on the surface of the support having been observed in HRTEM. The metal dispersions of 0.5, 1.0, 1.5, and 2.0 wt % Pt were 46.68, 33.03, 3.72, and 1.18% measured by CO chemisorption. The HRTEM images show that BaSO₄ particles are in good contact with platinum particles with several Pt particles residing on each BaSO₄ particle as observed in Figure 6b. Figure 9 shows the Pt surface area of Pt/BaSO₄ increases linearly with the increase of the platinum loading and reaches 0.8158 m²/g sample at the maximum platinum content of 1 wt %. Then Pt surface area decreases drastically and reaches 0.0585 m²/g over the sample at platinum content of 2.0 wt %.

4. Conclusions

The activity studies over Pt/BaSO₄ impregnated catalyst indicate the presence of more particle dispersion and high platinum surface area is therefore responsible for the high activity toward SO₃ decomposition. The 1 wt % Pt/BaSO₄ (PrOH) catalyst, the BaSO₄ prepared by spray pyrolysis using propanol as a solvent, shows the highest catalytic activity at 1023 K compared with other catalysts. The scanning electron microscopy and high-resolution transmission emission microscopy results clearly indicate the necessity of the platinum dispersed on BaSO₄, which had relatively good crystallographic orientation, suggesting the establishment of a strong metal–support interaction. Thus, we conclude that these catalysts are highly active for SO₃ decomposition and are an environmentally stable material.

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