1   # Catalytic hydrolysis of carbonyl sulfide in blast furnace gas

2   # over Sm-Ce-O$_x$@ZrO$_2$ catalyst

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21 **Reagents**

22   All chemicals were purchased from commercial sources and used without further

23 treatment. $Ce(NO_3)_2 \cdot 6H_2O$, 99.5%, Shanghai Macklin Biochemical Co., Ltd;

24 $Sm(NO_3)_3 \cdot 6H_2O$, 99.9%, Shanghai Macklin Biochemical Co., Ltd; $Y(NO_3)_3 \cdot 6H_2O$,

25 99.9%, Shanghai Macklin Biochemical Co., Ltd; Na_2CO_3, 99%, Shanghai Macklin

26 Biochemical Co., Ltd; K_2CO_3, 99%, Shanghai Macklin Biochemical Co., Ltd; ZrO_2,

27 99.99%, Sinopharm Chemical Reagent Co., Ltd.

28 **Catalytic activity and selectivity measurement**

29   The catalyst (1 mL) was added to a fixed-bed quartz reactor (inner diameter 8 mm)

30 to study the catalytic activity of COS hydrolysis. First, the mixture gas (COS: 200 ppm,

31 H_2O: 2%, N_2 as carrier gas) was introduced into the reactor, and water was injected into

32 the gas mixing tank through a micro-injection pump, conversion of liquid water into

33 water vapor into the reactor by heating in an oil bath. The total flow rate of flue gas was

34 200 mL/min. The temperature range of the activity test was 60~180 °C, and the

35 temperature was programmed. The experimental setup involves gradually increasing

36 the temperature at a rate of 1 °C per minute, commencing from an initial temperature

37 of 60 °C. Each subsequent temperature point for evaluating the catalytic performance

38 is spaced apart by 30 °C. The exhaust gas was analyzed after heating to the target

39 temperature and stabilizing for 30 min. Gas chromatography (Flame Photometric

40 Detector (FPD) from Agilent for sulfide detection) was used to detect the concentration

41 of COS and product H_2S at the inlet and outlet of the reactor.

42   COS conversion and H_2S selectivity are defined as follows:

$$COS\ Conversion(\%) = \frac{COS_{in} - COS_{out}}{COS_{in}} \times 100\%$$

43   Eq. (1)

$$H_2S\ Selectivity(\%) = \frac{H_2S_{out}}{COS_{in} - COS_{out}} \times 100\%$$

44   Eq. (2)

45   Among them, COS_{in} represented the COS concentration at inlet of the reactor,

46 COS_{out} represented the COS concentration at outlet of the reactor, and H_2S_{out}

47 represented the H$_2$S concentration at the outlet of the reactor.
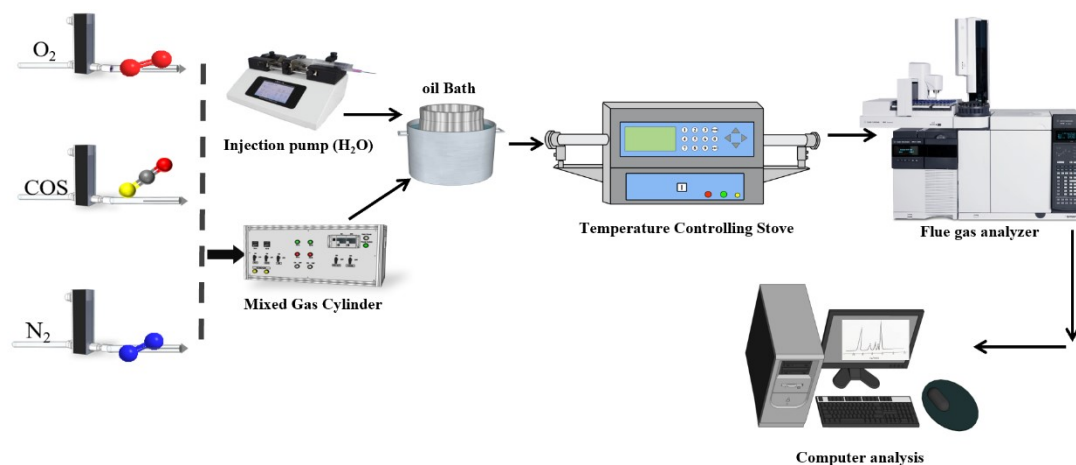
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49   Fig.S1 Simulation diagram of experimental device.

Characterization

51   The structure of the catalyst was analysed using a Rigaku D max/RB X-ray

52 diffractometer from Rigaku, Japan. The 2θ scanning range was from 5 to 85 °, starting

53 at 5° and scanning to 85° at a scanning speed of 10 °/min.

54   The amount of N$_2$ adsorbed on the surface of the solid material was determined at

55 different relative pressures at a constant temperature, and the data on the surface

56 structural properties of the solids were calculated on the basis of the multilayer

57 adsorption theoretical equations of Brunner-Emmet-Teller (BET) as well as the HK,

58 BJH, and DFT equations. The BET data of the catalysts were obtained using an

59 American ASAP 2020M V3.00H specific surface area analyzer. The catalysts were

60 pretreated at 100 °C for 5 h under vacuum, and then the adsorption and desorption tests

61 were carried out with N$_2$ as the adsorbent, and the adsorption temperature was

62 maintained at -196 °C by using liquid nitrogen.

63   The surface acidity and surface oxygen species of the catalyst were tested and

64 analyzed by a CHEMBET-3000 automatic chemical adsorption instrument. The

65 catalysts were preheated in He atmosphere at 400 °C for 1 h and then cooled to 50 °C.

66 After the catalyst was saturated with NH$_3$ at 50 °C, the catalyst was heated up to 800

67 °C in a N$_2$ stream at a constant rate of 10 °C/min, the change in NH$_3$ desorption with

68 temperature was recorded continuously.

69   The redox performance of the catalysts was tested and analyzed by a

70 Micromeritics TPD/TPR 2900 instrument. All catalysts were preheated at 400 °C for 1

71 h and then cooled to 50 °C under Ar flow, and increased the temperature from 50 °C to

72 800 °C at a heating rate of 10 °C/min.

73   The temperature programmed desorption of carbon dioxide (CO_2-TPD) was

74 conducted on the CHEMBET-3000 (Quantachrome) to obtain the surface alkaline

75 properties. All the catalysts were preheated at 400 °C under a helium stream for 1 h,

76 and then cooled to 50 °C for the carbon dioxide adsorption. Afterwards, the CO_2

77 adsorption was determined by a TCD detector from 50 °C to 800 °C in a 10% CO_2/He

78 atmosphere at a temperature increase rate of 10 °C/min.

79   Via AXIS ULTRA DLD instrument (Al-Kα radiation, 1486.6 eV) to obtain X-ray

80 photoelectron spectroscopy (XPS) maps. The vacuum was maintained at 10^{-7} Pa. The

81 samples were dried at 100 °C for 24 h to remove H_2O. The obtained curves were fitted

82 using XPSPEAK 4.1 software with Shirley background.