Supporting Information

A Mullite Oxide Catalyst of $SmMn_2O_5$ for Three-Way Catalysis: Synthesis, Characterization, and Catalytic Activity Evaluation

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S-1. Preparation of the Oxides and Pt Reference Catalyst

The $SmMn_2O_5$ mullite oxide and $LaCoO_3$ perovskite oxide was synthesized using co-precipitation method[1] and citrate method[2], respectively. The samples synthesized using Pluronic F127 was designed as SMO-F127-1 and LCO-F127-1. Meantime, the sample synthesized without Pluronic F127 designed as SMO-F127-0 and LCO-F127-0.

2 wt.% Pt on γ-Al_2O_3 was prepared by the incipient wetness method using $Pt(NO_3)_2$, calcined for 2 hours at 500 °C.

The prepared laboratory reagent of Sm_2O_3 and Mn_3O_4 respectively belong to extra pure reagent (3N) and analytical reagent (AR).

S-2. X-Ray Diffraction of Sm_2O_3 and Mn_3O_4

The XRD spectra of Sm_2O_3 (SO) and Mn_3O_4 (MO) oxides were showed in Fig. S1. The major peaks of the XRD patterns of the samples are consistent with the characteristic peaks of tetragonal structured Mn_3O_4 (JCPDS 76-0601) and monoclinic structured Sm_2O_3 (JCPDS 89-4837), respectively.

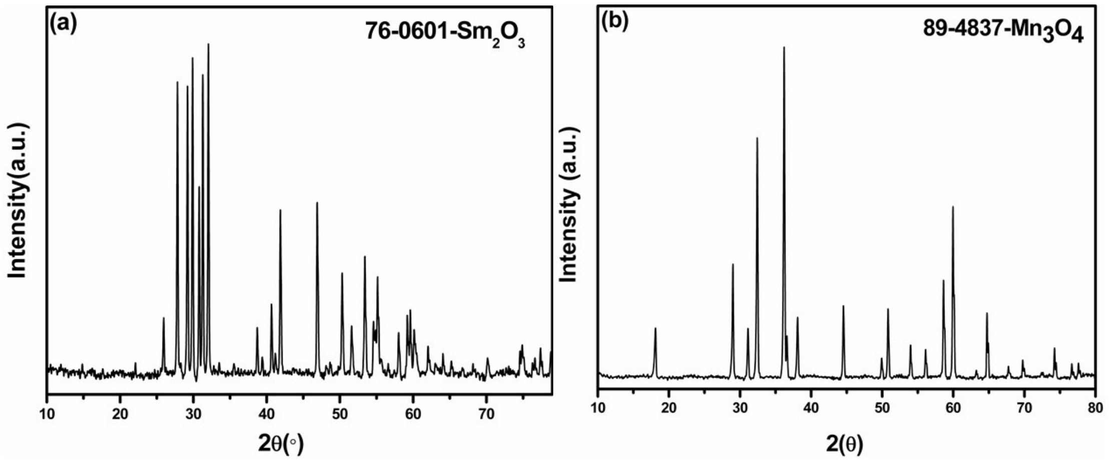

Fig. S1 The XRD profile of the simple samples. a: Sm_2O_3; b: Mn_3O_4.

S-3. CO Catalytic Activity

Fig. S2 showed the CO oxidation performance of the mullite oxide SMO and perovskite oxide LCO. We also prepared the 2 wt.% Pt/Al_2O_3 sample using impregnation method as a reference and investigated its CO oxidation activity in same condition. The CO to O_2 ratio was about 1/10 in the CO reaction in our experience. A mixture of reactants $CO/O_2/N_2$ = 2/20/78 at a total flow rate of 200ml/min (space velocity (SV) = 120000 $mlg^{-1}h^{-1}$) was passed through the catalyst bed containing 100 mg of the catalyst in three-way catalysis experiment. It can be seen that the CO conversion result was similar for SMO and 2 wt.% Pt/Al_2O_3, reaching a conversion efficiency of 100 % at 130 °C. It was obvious that mullite oxide SMO has comparable CO oxidation performance with 2 wt.% Pt/Al_2O_3, better than LCO sample.

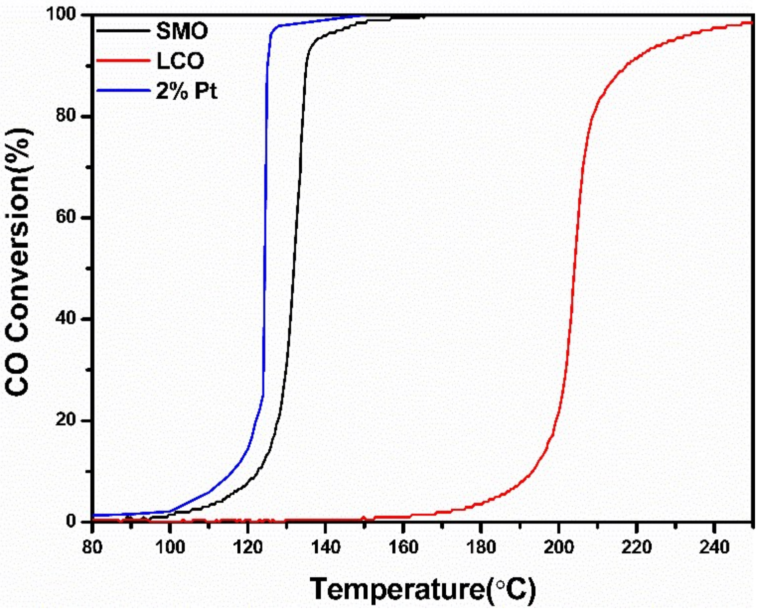

Fig. S2 The CO conversion as a function of temperature for the prepared samples.

S-4. Pluronic F127 Effect for Three-way Catalytic Performance

In order to distinct the effect of surfactant Pluronic F127, We also prepared the SMO sample without Pluronic F127 and LCO sample using F127. The three-way catalytic performance was tested for the all samples as well in same condition ($CO/NO/C_3H_8/O_2/N_2$ = 2.0/0.2/0.05/1.0/96.75 at a total flow rate of 200 ml/min (space velocity (SV) = 120000 $ml{\cdot}g^{-1}h^{-1}$)). Fig. S3a showed the CO conversion of the prepared samples, the results showed that surfactant F127 could not improve the CO oxidation catalytic activity for perovskite oxide LCO, in contrary, slightly inhibit CO oxidation activity. However, SMO still obtained a better catalytic performance than LCO, regardless of the surfactant F127 effect. The two samples of SMO have a similar starting conversion temperature for C_3H_8 oxidation, as demonstrated in Fig. S3b. The same tendency was observed for T_{50} and T_{90} as well. Compared to the two SMO samples, surfactant Pluronic F127 improved the C_3H_8 oxidation catalytic activity for LCO sample to some extent, as the T_{50} decreased dramatically for LCO-F127-1 than that of LCO-F127-0. Fig. S3c exhibited the NO conversion behaviors of four samples. Compared with SMO sample, Pluronic F127 hindered NO reduction catalytic activity for LCO, which improved nearly 100 ^{o}C for the starting conversion temperature, T_{50} and T_{90}, similarly. Based on the results, we can see that SMO-F127-1 and LCO-F127-0 have a better overall three-way catalytic performance compared with SMO-F127-0 and LCO-F127-1. In a word, Pluronic F127 could improve the three-way catalytic performance for mullite oxide SMO sample to a certain degree, but for perovskite oxide LCO, F127 would reduce its catalytic performance. That means surfactant F127 not the key factor make the SMO has a better three-way catalytic performance than LCO in our experimental research.

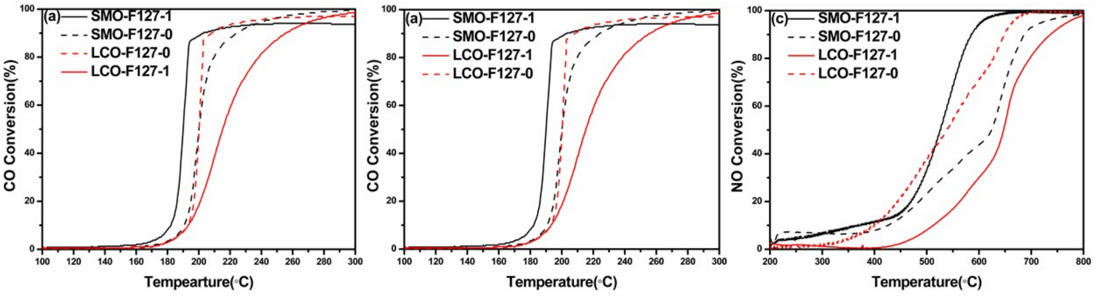

Fig. S3 (a) CO, (b) C_3H_8, (c) NO conversion of the oxides as a function of temperature under the simulated exhaust gas. ($CO/NO/C_3H_8/O_2/N_2$ = 2.0/0.2/0.05/1.0/96.75)

S-5. Three-way Catalytic Performance for Simple Oxide

The simple oxides Sm_2O_3 and Mn_3O_4 were prepared and evaluated for three-way catalytic performance as a reference to compare with mullite oxide $SmMn_2O_5$ in same condition ($CO/NO/C_3H_8/O_2/N_2$ = 2.0/0.2/0.05/1.0/96.75 at a total flow rate of 200 ml/min (space velocity (SV) = 120000 ml·g^{-1}h^{-1})). From Fig. S4a, c, it obvious that the oxides SO and MO exhibited barely no activity for three-way catalysis when compared with SMO for CO oxidation and NO reduction. For SMO sample, the CO conversion already reach 100% When SO and MO start catalyze CO oxidation, and even not show any activity for NO reduction after the temperature up to 800 ^{o}C. As for the C_3H_8 oxidation showed in Fig. S4b, all samples showed catalytic activities, among them SMO obtained the best performance. The starting conversion temperature was about 200 ^{o}C for SMO, 300 ^{o}C lower that for SO and 400 ^{o}C lower than MO. and T_{50} and T_{90} were also lower for SMO than SO and MO. That is to say, based on the results, the SMO obtained an overall better three-way catalytic performance than simple metal oxides SO and MO. In addition, simple metal oxides have a poor stability, leading to an oxide phase change with different stoichiometry in redox reactions. [3] And metal oxides were accepted to experience surface phase change and the corresponding loss of catalytic activity during an extended period of catalytic reactions. [4] The conclusion can be obtained that mullite oxide SMO not only has a better catalytic activity but show a commendable stability at an extended period of catalytic reactions than simple metal oxides. So SMO could become a promising material for three-way catalysts.

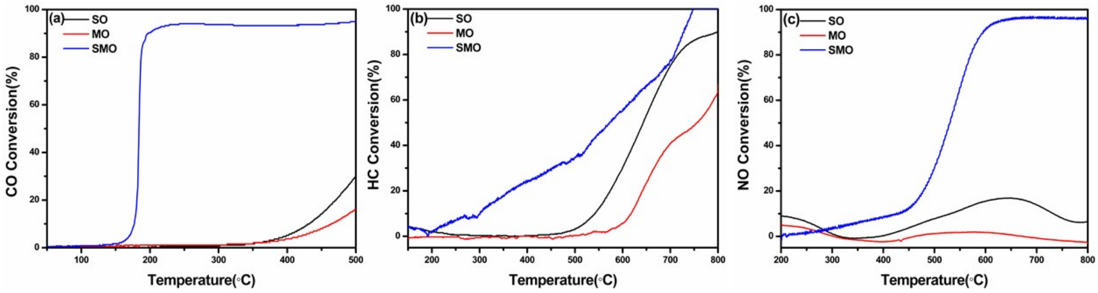

Fig. S4 (a) CO, (b) C_3H_8, (c) NO conversion for SO, MO and SMO oxides as a function of temperature under the simulated exhaust gas. ($CO/NO/C_3H_8/O_2/N_2$ = 2.0/0.2/0.05/1.0/96.75)

References

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