

Low temperature complete combustion of CH₄ emitted from CNG vehicles over Ce-doped Nano Co-Mn catalyst

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Compressed natural gas (CNG) is becoming a popular cleaner alternative fuel to gasoline/diesel and is used widely throughout the world owing to its several advantages. However, due to incomplete combustion, CNG vehicles emit significant amount of methane (CH₄) in the exhaust which is of great environmental concerns. CH₄ is a greenhouse gas having 21 times more global warming potential than CO₂. It is therefore essential to reduce anthropogenic CH₄ emissions. Catalytic oxidation is one of the best methods to control CH₄ emission. Platinum group metals (PGM) catalysts are highly active for CH₄ oxidation. Due to their expensiveness and rare availability, it is vital to find a substitute of PGM catalyst.

The aim of present work was to develop a cost effective, Ce doped Co-Mn catalysts (Co₅Mn_{1-x}Ce_x, x=0.1-0.5) for CH₄ oxidation at low temperature within the range of vehicle exhaust temperature (150-450°C). The catalysts (A, B, C, & D) with definite doping of Ce were prepared by co-precipitation method and calcined in a reactive 4.5%CO-air mixture to produce oxygen deficient catalyst (Table 1). The catalysts were characterized by XRD, FTIR and N₂-physisorption. The activity test of catalysts were performed in a fixed bed tubular reactor under the following conditions: 500 mg catalyst, 1.5% CH₄ in air, total flow rate 100 ml/min, heating rate 1°C/min, temperature range: ambient to 450°C. The gaseous reactants and products were analysed by an on-line GC equipped with porapack q-column, methanizer and FID detector.

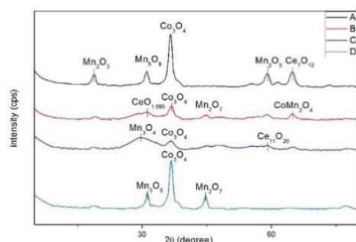


Figure 1: XRD Pattern of catalyst A, B, C and D

XRD pattern of the catalysts are presented in Figure 1. The intensity of XRD peaks increase in order C < B < A < D. The diffraction peaks of cat-C (Co₅Mn_{0.9}Ce_{0.1}O_x) show that it is almost amorphous. The crystallite sizes of the various cobaltite catalysts

were calculated using the Debye-Scherrer eq. 1, as given in Table 2.

$$d = 0.89\lambda / \beta \cos \theta \quad (1)$$

Table 1: Crystallite size of prepared catalyst samples

Catalyst ID	Catalyst	Crystallite Size (nm)
A	Co ₅ Mn _{0.5} Ce _{0.5} O _x	4.72
B	Co ₅ Mn _{0.7} Ce _{0.3} O _x	9.03
C	Co ₅ Mn _{0.9} Ce _{0.1} O _x	14.59
D	Co ₅ MnO _x	10.62

The activity test results showed that performance of Ce doped catalysts was better than un-doped one. The lowest temperature for total combustion of CH₄ over the best catalyst was 407°C (Figure 2). Table 2 shows light off characteristics of the catalysts. The activity order of the catalysts is as follows: Co₅Mn_{0.9}Ce_{0.1}O_x (407°C) > Co₅Mn_{0.7}Ce_{0.3}O_x (465°C) > Co₅Mn_{0.5}Ce_{0.5}O_x (495°C) > Co₅MnO_x (545°C). Doping appropriate amount of Ce enhances the activity of catalyst, Ce provides extra O for CH₄ oxidation.

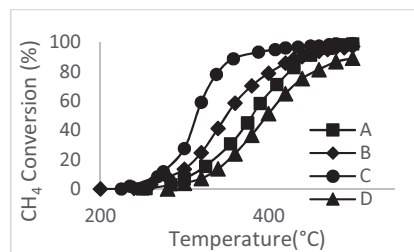


Figure 2: Activity test performance of Ce-promoted catalysts for CH₄ oxidation

Table 2: Light off temperature for Ce promoted catalysts.

Catalyst ID	Catalyst	T ₁₀ (°C)	T ₅₀ (°C)	T ₉₀ (°C)
A	Co ₅ Mn _{0.5} Ce _{0.5} O _x	310	380	450
B	Co ₅ Mn _{0.7} Ce _{0.3} O _x	290	350	440
C	Co ₅ Mn _{0.9} Ce _{0.1} O _x	270	315	360
D	Co ₅ MnO _x	330	400	510

References

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