Dry reforming of methane using DBD plasma reactor coupled with Ni/La_2O_3-$MgAl_2O_4$

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Carbon dioxide (CO_2) could be utilized with methane (CH_4) for the production of syngas through catalytic CO_2 reforming of CH_4 or dry reforming of methane (DRM)[1,2]. DRM offers a number of remarkable advantages such as mitigation of both greenhouse gases, and direct production of syngas. Ni/La_2O_3-$MgAl_2O_4$ has been investigated for dry reforming of methane (DRM) in a cold plasma dielectric barrier discharge (DBD) fixed-bed reactor [3]. Ni/La_2O_3-$MgAl_2O_4$ was prepared according to the modified coprecipitation assisted hydrothermal method.

The addition of La_2O_3 as a co-support enhances the Ni−support interaction. The web-like structure of La_2O_3 allows better Ni dispersion over the support as observed in the EDX mapping shown in Figure 1(a). Figure 1(b) illustrates the elemental mapping of Ni, La, Mg, Al and O of the Ni/La_2O_3-$MgAl_2O_4$. The catalytic DBD plasma reactor significantly improves the conversion of CH_4 and CO_2 to 86% and 84.5%, respectively. The selectivity for H_2 and CO is 50% and 49.5%, respectively. The syngas ratio (H_2/CO) increases from 0.86 to 1.01, while the overall energy efficiency is 26% higher than that of plasma only DRM (Figure 2).

The enhanced DRM activity is ascribed to the higher basicity for Ni/La_2O_3-$MgAl_2O_4$ at 0.9377 mmol/g compared to 0.8477 mmol/g for $MgAl_2O_4$. The dielectric properties of the Ni/La_2O_3-$MgAl_2O_4$ is 18.3 compared to 12.0 and 8.8 for NiO and $MgAl_2O_4$, respectively. The formation of intermediate carbonate ($La_2O_2CO_3$) (Figure 2) inhibited carbon deposition as evident by TGA and EDX mapping. Furthermore, the catalyst is also successfully regenerated, and stable DRM performance is maintained during cyclic runs.

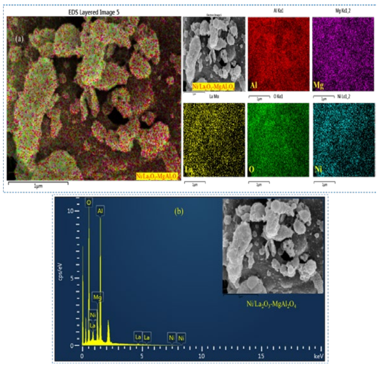

Figure 1. (a) EDX dot mapping of Ni/La_2O_3-$MgAl_2O_4$ (b) EDX elemental analysis Ni/La_2O_3-$MgAl_2O_4$

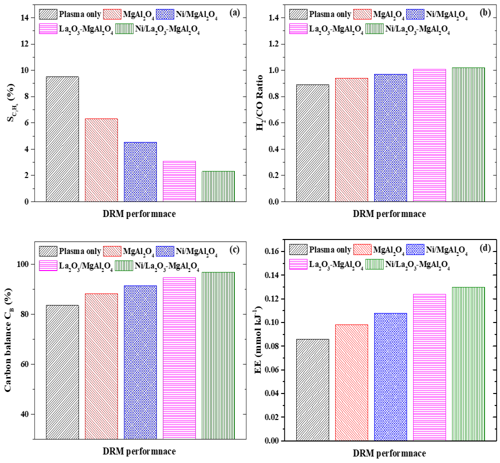

Figure 2. Catalyst performance analysis: (a) C_2H_6 selectivity (b) H_2/CO ratio (c) carbon balance C_B (d) energy efficiency (EE) mmol kJ^{-1}; GHSV 364 h^{-1}, SIE =300 J mL^{-1}, catalyst loading = 0.5 g, D_{gap}=3 mm, D_L=20 cm, V_D = 9.75 cm^3, T= 350 ºC

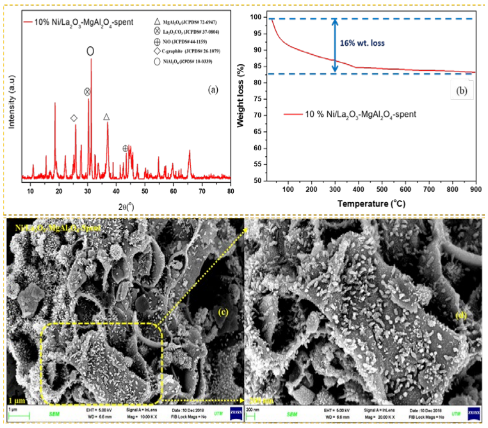

Figure 3. (a) XRD pattern of the spent Ni/La_2O_3-$MgAl_2O_4$ (b) TGA profile for spent catalyst after 15 h operation time (c-d) FESEM of the spent catalyst with different magnification

References
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[2] Abbas et al., Fuel Proc Tech 248, 107836 (2023)
[3] Khoja, A.H. Phd Thesis Dry Reforming Of Methane Using Cold Plasma Reactor For Different Dielectric Materials And Modified $MgAl_2O_4$ catalysts (2019)