SUPPORT-INDUCED EFFECTS ON THE IRIDIUM NANOPARTICLES ACTIVITY, SELECTIVITY AND STABILITY PERFORMANCE UNDER THE CO_2 REFORMING OF METHANE REACTION

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The dry (CO_2) reforming of methane (DRM, Eq.1) for syngas production – a critical feedstock for the production of hydrogen, ammonia and liquid energy carriers – ranks among the top issues of applied catalysis in the light of environmental protection, renewable energy production and circular economy [1,2]. DRM involves the simultaneous reduction of two key greenhouse gases (CO_2 and CH_4) and provides an efficient way for CO_2 recycling as well as the direct utilization of biogas. Among others, these advantages make DRM (Eq. 1) a more favorable process compared to the other alternatives, steam reforming (SRM, Eq. 2) and oxy reforming (POM, Eq. 3):

DRM: $CH_4 + CO_2 \leftrightarrow 2CO + 2H_2$ $\Delta H^o = 247\ kJ{\cdot}mol^{-1}$ (Eq. 1)

SRM: $CH_4 + H_2O \leftrightarrow CO + 3H_2$ $\Delta H^o = 206\ kJ{\cdot}mol^{-1}$ (Eq. 2)

POM: $CH_4 + ½\, O_2 \leftrightarrow CO + 2H_2$ $\Delta H^o = -36\ kJ{\cdot}mol^{-1}$ (Eq. 3)

Here we report on the effect of the metal oxide supports (γ-Al_2O_3, alumina-ceria-zirconia (ACZ) and ceria-zirconia (CZ)) on the low temperature (ca. 500-750 ^oC) DRM activity, selectivity, resistance to carbon deposition as well as on the stability under high temperature oxidative aging of Ir nanoparticles dispersed on them. A variety of characterization techniques were implemented to provide significant insight into the factors (e.g. metal-support interactions and materials properties) that determine Ir intrinsic kinetics and stability during DRM. It was found that all Ir/γ-Al_2O_3, Ir/ACZ and Ir/CZ catalysts have a very stable time-on-stream DRM performance, although supports with high oxygen storage capacity (i.e. ACZ and CZ) promoted CO_2 conversion, yielding CO-enriched syngas. For all catalysts carbon deposition was low, although it is decreasing in the order Ir/γ-Al_2O_3>Ir/ACZ>Ir/CZ that is consistent with a bifunctional mechanism involving participation of oxygen vacancies on the surface of the support in CO_2 activation and carbon removal. The lower apparent activation energy for CO_2 consumption rate observed with CZ-containing catalysts (Ir/ACZ and Ir/CZ) suggests that CZ is a promising support for use in low temperature DRM.

Keywords: Dry reforming of methane; Ceria-zirconia mixed oxides; Iridium nanoparticles; resistance to carbon deposition.

References

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