METHANE DRY REFORMING OVER COPPER AND IRON SUBSTITUTED STRONTIUM COBALT OXIDE PEROVSKITE CATALYST

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ABSTRACT

With growing concern on global warming arising from greenhouse gases (GHG) emission, dry reforming of methane (DRM) for synthesis gas (syngas) production has emerged as a promising technology by converting the main GHG, methane (CH_4) and carbon dioxide (CO_2), into hydrogen (H_2) and carbon monoxide (CO). Previous research works have been carried out to develop perovskites (ABO_3) as reforming catalysts that exhibit high catalytic performance, excellent stability and coke deposition prevention. However, those studies are limited to the common use of perovskite lanthanum nickel-based ($LaNiO_3$) oxides in the DRM reaction. Literature reports that cobalt based catalysts, which are less prone to carbon formation are more active than nickel-based catalysts. Among the various promoters, the substitution of alkaline earth metals such as calcium and strontium for the A-site of perovskites enhances high metallic dispersion and basicity over the catalyst and minimizes carbon formation. In addition, the partial substitution of the 'B-site' improves the structural stability of perovskites and catalytic performance in methane reforming reactions. Therefore, this work aimed to synthesize and characterize perovskite strontium cobalt-based $SrCo_{0.8}M_{0.2}O_{3-\delta}$ for DRM reaction where M = iron (Fe) or copper (Cu). These perovskites, $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$, $SrCo_{0.8}Cu_{0.2}O_{3-\delta}$ and $SrCoO_{3-\delta}$ catalysts were prepared via a citrate sol-gel method. Various techniques were then employed to characterize the catalysts such as N_2 physisorption, XRD, SEM-EDX and FT-IR. For the kinetics of DRM, the synthesized catalysts were tested in a fixed-bed tubular reactor over a reactant ratio of 1:1 and reforming temperature of 1023 K to obtain the reactant conversion and product yield. From the results obtained, H_2:CO product ratios of less than 1.0 were catalytically produced over perovskite strontium cobalt-based $SrCo_{0.8}M_{0.2}O_{3-\delta}$ oxides. $SrCo_{0.8}M_{0.2}O_{3-\delta}$ materials were determined as effective perovskite catalysts for methane dry reforming with substantial high methane conversion.

ABSTRAK

Dengan keprihatinan pemanasan global berpunca daripada gas rumah hijau (GHG), Cucian reformasi dari metana (DRM) untuk pengeluaran gas sintesis (syngas) muncul sebagai satu teknologi yang memberangsangkan dengan menukar GHG utama, metana (CH_4) dan karbon dioksida (CO_2) ke dalam (H_2) hidrogen dan karbon monoksida (CO). Kajian-kajian yang lepas dan kerja-kerja telah dijalankan untuk membangunkan perovskites (ABO_3) sebagai pemangkin reformasi itu mempamerkan prestasi perangsang tinggi, kestabilan cemerlang dan mencegah pemendapan coke. Walau bagaimanapun, kajian tersebut adalah terhad kepada penggunaan biasa perovskite lanthanum berasaskan nikel ($LaNiO_3$) oksida sebagai reaksi DRM. Sastera dilaporkan kobalt berasaskan pemangkin, seperti kurang terdedah kepada pembentukan karbon yang lebih aktif daripada pemangkin berasaskan nikel. Antara penganjur pelbagai, gantian logam alkali bumi seperti kalsium dan strontium untuk A-tapak perovskites meningkatkan penyebaran logam yang tinggi dan basicity ke atas pemangkin dan mengurangkan pembentukan karbon. Di samping itu, sebahagian daripada penggantian 'B-tapak' meningkatkan kestabilan struktur dan tingkah laku pemangkin dalam reformasi reaksi metana. Justeru itu, kerja-kerja ini bertujuan untuk mensintesis dan mencirikan perovskite strontium berasaskan kobalt $SrCo_{0.8}M_{0.2}O_{3-\delta}$ oksida untuk tindak balas DRM di mana M = besi (Fe) atau Kuprum (Cu). Perovskite jenis $SrCo_{0.8}Fe_{0.2}O_{3-\delta}$, $SrCo_{0.8}Cu_{0.2}O_{3-\delta}$ dan $SrCoO_{3-\delta}$ oksida pemangkin telah disediakan melalui kaedah sol-gel citrate. Pelbagai teknik telah kemudian digunakan untuk mencirikan pemangkin reformasi seperti N_2 physisorption, XRD, SEM-EDX dan FT-IR. Bagi kajian kinetik DRM, pemangkin synthesized diuji di reaktor tiub tetap-katil yang lebih reactant nisbah 1:1 dan reformasi suhu 1023 K untuk mendapatkan penukaran reactants dan menghasilkan produk. Berdasarkan keputusan, H_2:CO produk nisbah kurang daripada 1.0 dihasilkan melalui perovskite strontium berasaskan kobalt $SrCo_{0.8}M_{0.2}O_{3-\delta}$ oksida. $SrCo_{0.8}M_{0.2}O_{3-\delta}$ telah ditentukan sebagai pemangkin perovskite berkesan untuk metana kering pembaharuan dengan penukaran metana yang tinggi.