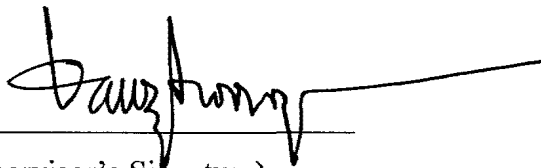


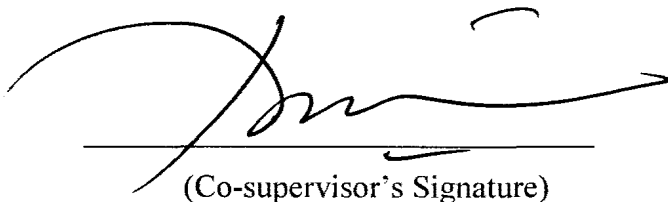
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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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**SYNGAS PRODUCTION FROM METHANE REFORMING PROCESSES OVER  
BORON PROMOTED Ni/SBA-15 CATALYSTS**

**SHARANJIT SINGH**

Thesis submitted in fulfillment of the requirements  
for the award of the degree of  
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## ABSTRAK

Bahan bakar fosil sedang dieksploitasi pada kadar yang belum pernah terjadi sebelum ini bagi memenuhi permintaan penduduk global yang semakin meningkat. Penurunan sumber minyak mentah dan peningkatan pengeluaran gas antropogenik rumah hijau (iaitu, CH<sub>4</sub> dan CO<sub>2</sub>) telah mendorong inisiatif untuk meneroka tenaga alternatif yang mesra alam, yang boleh menggantikan bahan bakar fosil yang tidak boleh diperbaharui dan turut mengurangkan pelepasan gas rumah hijau. Dalam hal ini, pembaharuan kering metana (MDR) baru-baru ini mendapat perhatian yang ketara kerana menukarkan gas rumah hijau kepada produk berharga. Walau bagaimanapun, MDR menggunakan pemangkin yang mahal dan kompleks, menjadikan teknologi ini kurang ekonomi dalam aplikasi dunia sebenar. Oleh itu, pembangunan bahan pemangkin yang kos efektif dan kejuruteraan proses maju adalah sangat wajar untuk menjadikan teknologi ini ekonomik untuk menghasilkan bahan bakar tenaga lestari. Dalam hal ini, pemangkin berasaskan Ni telah dikaji dengan teliti untuk MDR dan telah menunjukkan hasil penukaran yang berjaya untuk pengeluaran syngas. Tetapi pemangkin ini juga memperlihatkan kecenderungan pembentukan karbon yang akhirnya menyebabkan pnyahaktifan pemangkin. Kajian menunjukkan bahawa pnyahaktifan pemangkin oleh pembentukan karbon boleh dikurangkan dengan meningkatkan bekalan oksidan (iaitu H<sub>2</sub>O dan CO<sub>2</sub>) dan penggabungan penggalak. Oleh itu, tesis ini menyiasat kesan parameter operasi untuk pembaharuan metana kering dan penggabungan pembaharuan stim dan metana kering. Di samping itu, kajian kinetik dilakukan dengan menggunakan undang-undang kuasa dan enam ungkapan kinetik Langmuir-Hinshelwood yang berbeza melebihi 10% Ni / SBA-15 pemangkin. Selain itu, pengaruh kuantiti boron (1%, 2%, 3% dan 5%), promoter berbeza (B, Ce, La dan Y) dengan disiasat untuk tindak balas MDR. Sokongan SBA-15, pemangkin yang tidak digalakkan dan digalakkan telah dihasilkan dengan menggunakan teknik hidrotermal, kaedah impregnasi yang kering. Hasilnya mendapati korelasi yang kuat antara aktiviti pemangkin, suapan setiap reaktan dan pembentukan spesies karbon untuk tindak balas MDR terhadap SBA-15 yang disokong nikel. Malah, pemangkin mendapati kedua-dua jenis karbon amorfus dan grafit pada pemangkin yang digunakan. Pada mulanya, tenaga pengaktifan sebanyak 91 kJ mol<sup>-1</sup> untuk kadar penggunaan metana dikira dengan menggunakan ungkapan Power-law. Tambahan pula, satu mekanisme kinetik Langmuir-Hinshelwood disosiasi satu tapak dicadangkan, dengan tenaga pengaktifan kira-kira 72 kJ mol<sup>-1</sup>. Pemangkin menunjukkan rintangan yang tinggi terhadap pembentukan karbon grafit dalam kehadiran agen pengoksida H<sub>2</sub>O dan CO<sub>2</sub> dalam tindak balas pembaharuan, sementara re-pengoksidaan spesies logam nikel diperhatikan dalam tindak balas. Hasil yang diperolehi untuk pemuatan boron yang berbeza mencadangkan bahawa 2% boron mencukupi untuk mencapai aktiviti yang tinggi dan ketahanan daripada spesies karbon. Oleh itu, pemangkin 2% B-mencukupi lebih banyak dibandingkan dengan pemangkin lain yang digalakkan (La, Ce dan Y) di bawah syarat-syarat pembaharuan kering metana untuk 72 h kajian kestabilan. Keputusan menunjukkan bahawa pemangkin yttrium dipromosikan dengan ketara meningkatkan aktiviti pemangkin, sedangkan pemangkin yang dipromosikan oleh boron menunjukkan rintangan koko tertinggi di kalangan rakan-rakan.

## ABSTRACT

Non-renewable fossil fuels are currently being exploited at an unprecedented rate to meet the global energy demand. In addition, the increasing depletion of crude oil resources and growing anthropogenic greenhouse gases (i.e., CH<sub>4</sub> and CO<sub>2</sub>) emissions have spurred an initiative to find renewable and environmentally friendly energy sources which can substitute the existing non-renewable fossil fuels and simultaneously reduce greenhouse gas emissions. In this regard, methane dry reforming (MDR) has gained substantial interest since it can be used to convert these greenhouse gases to valuable products. However, MDR utilizes the expensive and complex catalysts, making the technology less economic in the real-world applications. Therefore, the development of cost-effective catalysts materials and advanced process engineering is highly desirable to make this technology economical to produce sustainable energy fuels. In this regard, Ni-based catalysts have been thoroughly investigated for MDR and have shown successful conversion results for syngas production. But these catalysts also exhibit the tendency for coke formation which ultimately results in the catalysts deactivation. Literature suggests that the catalyst deactivation by coke formation can be considerably reduced by increasing the supply of oxidants (i.e. H<sub>2</sub>O and CO<sub>2</sub>) and incorporation of promoters. Therefore, this thesis investigates the effect of operating parameters for methane dry reforming and bi-reforming of methane reactions. In addition, the kinetic studies were performed for methane dry reforming by using power law and six different Langmuir-Hinshelwood kinetic rate expressions over 10%Ni/SBA-15 catalyst. Besides, the influence of boron loading (1%, 2%, 3% and 5%) and different promoter (B, Ce, La and Y) was investigated for MDR reaction. The SBA-15 support, unpromoted and promoted catalysts were synthesized by using hydrothermal, incipient-wetness impregnation and sequential impregnation techniques, respectively. The results found a relation between the catalyst activity, feed of each reactant and formation of carbonaceous species for MDR reaction over SBA-15 supported nickel. In fact, the catalysts found both amorphous and graphitic kind of carbonaceous species on the spent catalyst. Initially, activation energy of 91 kJ mol<sup>-1</sup> for methane consumption rate was calculated by using the Power-law expression. Furthermore, a single-site dissociative Langmuir-Hinshelwood kinetic mechanism was proposed, with associated activation energy of about 72 kJ mol<sup>-1</sup>. The catalyst showed high resistance to graphitic carbon formation in the presence of H<sub>2</sub>O and CO<sub>2</sub> oxidizing agents in bi-reforming reaction, whilst re-oxidation of nickel metallic species was observed in the reaction. The results obtained for different boron loading suggested that the 2% of boron was sufficient to achieve high activity and resistance from carbonaceous species. Therefore, the 2%B-promoted catalyst was further compared with other promoted catalysts (La, Ce and Y) under methane dry reforming conditions for 72 h stability test. The results revealed that the yttrium promoted catalyst significantly enhanced the catalytic activity, whereas the boron promoted catalyst exhibited the highest coke resistance among the counterparts.

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