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Effect of hydrogen spillover in decalin dehydrogenation over supported Pt catalysts

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ABSTRACT

One weight percent Pt impregnated on activated carbon (Pt/AC) and alumina supports (Pt/Al_2O_3) have been tested for decalin dehydrogenation for the production of clean H_2. CO pulse chemisorption results indicate nearly same Pt dispersion in both Pt/AC and Pt/Al_2O_3 catalysts. The comparison of activity between Pt/AC and Pt/Al_2O_3 catalysts revealed superior decalin dehydrogenation activity over Pt/AC both in terms of turn over frequency as well as the yield towards H_2 production which is due to the spill over of H_2 on Pt/AC. The Pt dispersion in Pt/Al_2O_3 is lost due to the phase transformation of support (boehmite phase of alumina to γ phase) during reduction treatment and this is the main reason for the loss of decalin dehydrogenation activity. It is concluded that spill over hydrogen is also an important aspect in addition to Pt dispersion for getting higher H_2 yield from decalin dehydrogenation.

1. Introduction

Hydrogen is regarded as a futuristic clean fuel which could have a huge industrial impact and potential effect on fuel economy through fuel cell technology. The serious problem faced currently in proton exchange membrane fuel cell (PEMFC) relates to purity of hydrogen particularly in transport sector. The presence of CO as impurity usually leads to poisoning of the Pt electrodes of the fuel cell. In addition to purity the PEMFC also requires rapid hydrogen supply, low energy consumption, and high energy content apart from safety in handling hydrogen. The current status of hydrogen energy with different methodologies of H_2 production has been reported recently [1]. Brown made a comparative study of fuels for on-board hydrogen production for fuel-cell powered automobiles [2]. Recently, decalin dehydrogenation to produce clean hydrogen has been proposed for PEMFC powered vehicles [3–5]. The advantages associated with decalin dehydrogenation are: it gives a CO_X free hydrogen, the theoretical amount of hydrogen produced from each decalin mole is very high (5 moles of H_2) and the final organic product, i.e., naphthalene in the decalin dehydrogenation can easily be reconverted back into decalin by catalytic hydrogenation, by a well established commercial process [6]. Thus on board decalin dehydrogenation reaction can be treated as an option for H_2 carrier rather than H_2 source.

The most promising catalyst for decalin dehydrogenation is a Pt based catalyst with or without promoters. Alumina and/or activated carbon have been shown as the commonly used supports for Pt [7,8]. The amount of Pt that has been reported for the decalin dehydrogenation under super heated liquid film concept or reactive distillation conditions is about 5 wt% [4,9–11]. On the other hand, under vapor phase conditions the Pt loading on the support has been reported to be at around 1 wt% [12,13]. However, in the literature, there is no systematic comparison between the characteristics of Pt/Al_2O_3 and Pt/C catalysts with regard to the decalin dehydrogenation under vapor phase conditions. The present investigation is aimed to answer some of the characteristics of Pt supported on alumina and carbon supports which influences the decalin dehydrogenation reaction.

2. Experimental

Both alumina (Al_2O_3) and activated carbon (AC) supports were obtained from M/s. Aldrich Chemical Co. and M/s. Cabot Corporation respectively.

2.1. Preparation of catalysts

These supports were dried overnight at 393 K before deposition of Pt. Impregnation of these supports (9.9 g each of Al_2O_3

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