4.3. H₂ production by catalytic steam gasification of HyperCoal.

SYNOPSIS: HyperCoal is a solvent extracted coal with ash content <0.1 %. Catalytic steam gasification of HyperCoal was carried out at 650-775 °C for production of H₂ and synthesis gas. The gasification results showed nearly 4 times higher gasification rate than raw coal. Deactivation of the catalyst was not observed even after 6 times recycling of the same catalyst amount. H₂/CO ratio was reduced from 10.5 to 1.4 by changing the partial pressure of steam. The preliminary results showed that HyperCoal, an ash less coal, could be a potential hydrocarbon resource for H₂ and synthesis gas production by catalytic steam gasification process.

INTRODUCTION

The gasification of coal at temperatures below 800 °C has been widely examined [1-3]. The results concluded that except some low rank coals steam gasification rates of the most of the coals were very low and not practical. The gasification rates at low temperature can be enhanced significantly by using a catalyst. The most favored catalysts are alkali metals especially K₂CO₃. However, a major drawback is the reaction of catalyst with the mineral matter present as ash in the coals. Such interactions lead to the formation of compounds that are insoluble in water making the recovery of catalyst impossible [4]. The catalyst lost is to be compensated by addition of fresh new catalyst leading to an enormous amount of catalyst requirement. To overcome the problem of loss of catalyst, one approach is to remove the ash from the coal before gasification. Recently, our research group has developed a process to remove ash from coal by solvent extraction [5, 6]. The extracted coal called HyperCoal, has less than 200 ppm of ash. A bench scale demonstration plant producing 0.1 ton/day HyperCoal has been constructed by Kobe Steel. This study is a part of ongoing investigations about the potential applications of HyperCoal in energy related fields [7].

This study presents the preliminary results about the low temperature K₂CO₃-catalyzed steam gasification of HyperCoal. The possibility of recovery and recycling of the catalyst have been investigated. Effect of partial pressure of steam and gasification temperature on the gasification rate is investigated.

EXPERIMENTAL

Oaky Creek (OC) coal from Australia was selected for investigation. HyperCoal was produced by extraction of the coal with 1-methylnaphthalene at 360 °C and subsequently separating the extract (HyperCoal) from solvent. The extraction yield was 69 %. The HyperCoal production method has been described in detail elsewhere [5, 6]. The gasification was performed on a thermogravimetric (TG-DTA 2020, MAC) apparatus. The experimental set up consists of an HPLC pump, Argon supply, steam generator, TG, cold trap, micro GC and film flow meter. The steam was generated at 250 °C and inlet/outlet flow lines were kept at 250 °C by using ribbon heaters. The steam partial pressure was 0.5 obtained by adjusting the Ar flow and water pumping rate. The experiments were carried out under non-isothermal conditions. First, about 20 mg of sample was heated in Ar for 10 min at 250 °C to remove moisture. The sample was then heated to desired temperature 650-775 °C at a heating rate of 20 °C/min. Steam was introduced at 300 °C. Weight loss vs time data was measured from the beginning at room temperature. Also gas evolved was analyzed every 3 min using micro GC. Instant flow rate was recorded by Film flow meter after every 6 min. Samples were prepared by mixing 3-20 % (wt % of coal, daf) of K₂CO₃ with coal and HyperCoal. Oaky Creek coal samples without catalyst and with catalyst were named as OCnc and OCwc, respectively. Similarly, HyperCoal samples were named as OCHPCnc and OCHPCwc.

RESULTS AND DISCUSSION

Figure 1 shows steam gasification conversions for OCnc, OCwc, OCHPCnc, OCHPCwc at 775 °C and 0.5 partial pressure of steam. In case of catalytic gasification, the amount of catalyst was 6 wt%. The gasification can be seen to begin at 42 min which corresponds to 650 °C. The results show that the addition of catalyst significantly increased the gasification rates of both OC and OCHPC. However, the effect of catalyst in case of OCHPC was much

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Figure 1 Gasification profiles of OC and OCHPC.
higher than OC. The rate of OCHPCwc was 4 times higher than the rate of OCwc.

Figure 2 shows the change in gasification rate of OC and OCHPC when the catalyst is recycled. For OCwc, when the residue is mixed with the fresh coal the gasification rate decreased and becomes almost similar to OCnc rate. While, in OCHPCwc case the rate was same even after the catalyst was recycled three times.

**Figure 2** Gasification rate and catalyst recycling.

This shows that the catalyst was active in case of OCHPC even after repeated use while in OC it became inactive after one time use. This is because of the formation of inactive compounds by reaction between K$_2$CO$_3$ and mineral matter in coal. But in HyperCoal, ash content is less than 1000 ppm and therefore, complex K-compounds are not formed.

**Figure 3** Gasification rate vs partial pressure of steam at 775 °C for OCHPC.

Figure 3 shows the effect of partial pressure of steam on the gasification rate of the OCHPC. The steam partial pressure was changed from 0.5 to 0.01. H$_2$/CO ratio was reduced from 10.5 to 1.4. The curves show that from 0.5 to 0.1 the rate change is very little. From 0.05 the rate change is appreciable and becomes significant at 0.01. The results suggest that the reaction is nearly independent of steam amount up to 0.1. For partial pressure (pp) less than 0.1, the reaction becomes dependent on steam amount.

**Figure 4** shows the effect of the temperature on the gasification reactivity of OC HyperCoal. All gasification runs were done with same amount of catalyst. As can be seen from figure 4, gasification rate decreases as the temperature decreases. At 650 °C, the rate is extremely slow even in the presence of catalyst.

**CONCLUSIONS**
Catalytic gasification of HyperCoal at low temperature has been investigated. The results show HyperCoal can be used as feedstock for low temperature catalytic gasification. Recycling of catalyst is possible when HyperCoal is used because of its ash less nature. Both hydrogen and synthesis gas can be obtained from HyperCoal gasification by changing the process conditions. The reactivity of HyperCoal reduces with temperature even when a catalyst is used.

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**REFERENCES**