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The effect of microwave treatment on potassium promoted iron Fischer-Tropsch catalysts

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Chapter 6
General conclusions and recommendations

This study describes the effect of potassium loading on iron-based Fischer-Tropsch catalysts that were modified in the solid state using microwave radiation. Unsupported precipitated catalysts and silica supported catalysts have been studied in this work. The effect of microwave modification on the bulk properties of these catalysts has been studied as a function of the potassium loading. Bulk properties that were studied included the surface area, the morphology of the catalysts, the reducibility of iron and the phase composition of the samples. These parameters were found to be unchanged after microwave pre-treatment of all the samples. This observation suggested that the microwave pre-treatment effect is a surface phenomenon and not a bulk one. The study then focused on surface sensitive techniques, namely temperature programmed surface reaction-mass spectrometry and Fischer-Tropsch synthesis.

Effects of microwave pre-treatment on the surface carbon phase transformation were systematically studied in this work. Following exposure to MW radiation as a function of the potassium loading, marked differences in the total CH_4 evolved as revealed by TPSR are evident for the K/Fe and K/Fe/SiO_2 catalysts. For the K/Fe samples, MW induced increments in the total CH_4 evolved increased with the loading of potassium in the catalysts. The percentage increase started from 6%, attained an optimum value of 35%, before tailing off to 14% as the loading of potassium was increased. For the K/Fe/SiO_2 catalysts, microwave induced increments of the total CH_4 evolved increased from 13%, reached an optimum increase of 66%, before tailing off to 64% as the potassium loading increased. This indicated that the microwave effect generally increases with the loading of potassium, attains an optimum increase before it starts to decline. This observation was made for both supported and unsupported catalysts. However, the percentage increase in CH_4 induced by MW is higher in K/Fe/SiO_2 than K/Fe samples.

Investigations on the effect of MW modification on the catalyst surface carbon species was done by fitting Gaussian curves to the TPSR profiles. In the unsupported

catalysts, MW pre-treatment increased the formation of adsorbed atomic surface carbon (C_α) and surface methylene chains or films (C_β) for all the potassium loadings that were studied. Bulk iron carbides (C_γ) and graphitic carbon (C_δ) either remained unchanged or decreased upon microwave exposure of the samples. In the silica supported samples, MW modification promoted the formation of bulk iron carbides and graphitic carbon, whereas formation of adsorbed atomic surface carbon and surface methylene chains was suppressed. This observation is evident at all the loadings of potassium in the supported samples. Thus, it appears that the surface carbon species formed after CO adsorption are different for supported and unsupported catalysts.

Increasing the MW irradiation time from 10 seconds to 600 seconds did not change the total amount of CH_4 produced from the K/Fe catalysts. An average of ~26 000 peak area units were produced irrespective of the irradiation time (see Figure 4.21). However, contrasting behaviour was observed in the K/Fe/SiO_2 catalysts. The total CH_4 produced from the supported catalysts increased with an increase in the microwave irradiation time (see Table 5.7). A 1.0K/10Fe/SiO_2 sample that was irradiated for 10 minutes produced CH_4 equivalent to 32 127 peak area units. This value is higher than the 26 000 units produced from the non-irradiated 1.0K/Fe catalyst. This is remarkable considering that the supported catalyst only contains 10% of the active iron, while the unsupported catalyst is 99% iron oxide. This clearly showed that the support also influences the microwave pre-treatment effect.

In future work it is recommended that the effect of the support on the MW pre-treatment effect be investigated. Studies also need to be conducted so that the influence of other supports like TiO_2, Al_2O_3 and CNTs on the MW effect can be understood. It is also recommended that other surface sensitive characterization techniques such as secondary ion mass spectrometry (SIMS) be developed to confirm the TPSR findings.