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Effects of Cobalt Oxide-Iron Oxide Ratios on the Catalytic Activity of Cobalt Ferrite Spinel Catalysts

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Abstract. The cobalt ferrite spinels $\text{Co}_x \text{Fe}_{3-x} \text{O}_4$ ($0.6 < x < 2.4$) have been prepared by coprecipitation. The structures of the catalysts were characterized by DTA, X-ray diffraction, surface area and electrical conductivity measurements. The catalytic decomposition of isopropanol was carried out at 325°C using a flow system. The results obtained revealed that the activity and selectivity of the catalysts are strongly affected by the structure of the ferrite spinel formed as a result of the solid - solid interaction between Fe_2O_3 and Co_3O_4 . The normal spinel formed at ($x = 1$) gives low activity and low selectivity in comparison with the spinels deviated from the stoichiometry. Additionally, a little change in catalytic behaviour observed on increasing the calcination temperature from 400° to 600°C. Correlations between the composition, electronic and catalytic properties of the catalysts are reported.

1. INTRODUCTION

In an increasing number of areas of technology, industrial progress is related to the understanding of the factors influencing the reactivity of the solids. The co-existence of different oxidation states of transition metal ions occurs naturally in minerals and is frequently induced during the preparation of solid state catalysts [1]. The distribution of cations of ferrite spinel among available tetrahedral (t) and octahedral (o) sites are of considerable industrial interest due to their catalytic activity [2-5]. However, it was already reported that the wide variation in both catalytic and electrical properties of ferrite spinels is related mainly to their stoichiometry [3,4]. The aim of this work is to correlate the catalytic activity and electrical conductivity of ferrite spinel to their structure changes.

2. Experimental

Analard grade chemicals were used throughout. Mixed samples of iron and cobalt hydroxides (x the mole contents of Co^{2+} are 0.6, 0.8, 1.0, 1.5, 2.0, 2.2 and 2.4) were prepared according to the method reported elsewhere [6]. Differential thermal analysis (DTA) was carried out using an automatically recording thermobalance, type 160 KS (Germany). X-ray diffraction (XRD) of the thermal products were recorded using a Philips diffractometer (Model PW2103). Surface area measurements were determined at -196°C with the help of conventional volumetric apparatus. IR spectra of solid catalysts were recorded in 1600-200 cm^{-1} region with the spectrophotometer Perkin-Elmer Model 599B, using KBr technique. The electrical conductivity measurements were made as described previously [7]. The catalytic activity determination was carried out in a conventional fixed - bed flow type reactor.

3. Results and discussion

The prepared catalysts are characterized by DTA, XRD, IR and SBET measurements. The results showed that cobalt ferrite spinels were formed at a calcination temperature of 400°C and their compositions depend on the x ratio and the calcination temperature. The electrical conductivity (σ) of these spinels was measured in air and in presence of IPA and cited in Table 1.

Table 1. Variation of $\log \sigma$ with x ratio of Co-Fe mixtures calcined at 500°C and measured at 300°C.

x ratio	0.6	0.8	1.0	1.5	2.0	2.2	2.4
$\log \sigma$ (air)	-4.4	-4.6	-4.3	-4.1	-3.8	-3.6	-3.4
$\log \sigma$ (air + IPA)	-3.5	-3.3	-4.5	-4.3	-4.4	-4.6	-4.5

It appears that the conductance in the rich region with normal spinel is slightly changed in the presence and absence of IPA. The catalytic activity of spinel catalysts calcined at 500°C was tested at a reaction temperature of 325°C using the decomposition of IPA. Figure 1 shows that the inverse spinel catalysts are active and selective towards the formation of acetone. The formation of inverse spinel is accompanied with the occurrence of redox couples within the octahedral sites which are responsible for the chemisorption of alcohol and eventually also its decomposition. On the other hand, the sharp decrease of acetone yield for the ratio $x = 1$ may be attributed to the formation of normal spinel which is accompanied with a small electron exchange within its structure [4].

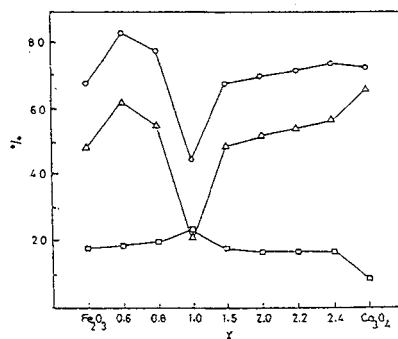


Figure 1: Variation of IPA conversion (o), acetone yield (Δ) and propylene yield (\square) (all in %) with the catalyst composition calcined at 500°C for 3 h.

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