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A Comment on "Catalysis by Transition Metal Sulfides" by Harris and Chianelli

Important advances have been made in understanding the structure of hydrodesulfurization (HDS) catalysts. The classical work was the relationship between the amount of Co in the Co–Mo–S phase, as measured by *in situ* Mössbauer emission spectroscopy, and the HDS reaction rate constant, (*1*). Fe–Mo–S has been found (*2*) to have the same structure as Co–Mo–S. However, Fe does not promote HSD with Mo–S catalysts as much as Co and Ni.

Harris and Chianelli (*3*) have successfully explained this HDS promotional effect for the first row transition metal sulfides (including Fe, Co, and Ni) using electronic concepts. They found a good correlation between HDS of dibenzothiophene and an activity parameter calculated using the SCF-Xα scattered wave method.

The purpose of this communication is to compare the activity parameter calculated by Harris and Chianelli with experimental data (*4*) obtained at higher pressures (13.9 MPa, 2000 psig) using a real feedstock (heavy gas oil) and catalysts calcined at 500°C (Fig. 1). The results for the Mo-Al_2O_3 unpromoted catalyst (in the units of Fig. 1) were 0.163 for HDS, 0.0207 for hydrodenitrogenation (HDN), and 1.71×10^{-4} for hydrogenation. With the exception of the Cr-promoted catalyst, the correlation is reasonably good. In particular, the experimental data show that Cu is a poison, in complete agreement with the calculations of Harris and Chianelli (*3*).

Figure 1 shows that the calculated activity parameter of Harris and Chianelli, A_2, also correlates with HDN and with hydrogenation. A_2 only depends on the properties of the solid catalyst and is independent of any particular reaction or mechanism. Therefore, these correlations indicate that the same catalyst properties that influence the promotion of HDS reactions also influence the promotion of HDN and hydrogenation reactions.

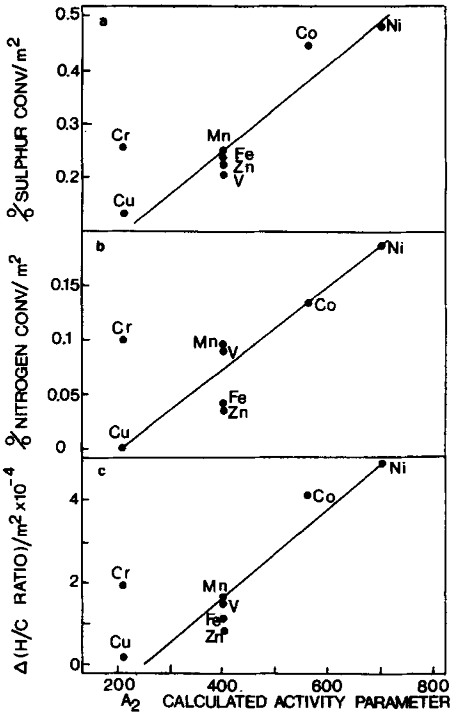

FIG. 1. Percentage sulfur conversion per m² (a), percentage nitrogen conversion per m² (b), and change (liquid product minus feedstock) in atomic hydrogen to carbon ratio per m² (from Ref. (*4*)) versus the calculated activity parameter A_2 (from Ref. (*3*)) (c).

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