This research was supported by the Deutsche Forschungsgemeinschaft. We thank Dr Sperling and Dr Sondermann for preparing the europium complex I.

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Received May 11, 1970.

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Effect of Low Magnetic Fields on the Catalysed Parahydrogen Conversion Rate over Certain Rare Earths

THE purpose of this communication is to report a change in rate of the parahydrogen conversion if the catalyst is placed in a magnetic field of a few oersteds. The change is a decrease of several per cent, and it has been observed over Pr_2O_3, Nd_2O_3, Sm_2O_3 and Yb_2O_3. It has not so far been observed over Eu_2O_3, Gd_2O_3, Tb_2O_3, Dy_2O_3, Ho_2O_3, Er_2O_3 or Tm_2O_3. But Tb_2O_3 supported at low surface concentration on high-area Al_2O_3 shows the effect.

In a typical experiment high purity rare earths are calcined in air at 800° C overnight. From 10 to 100 mg is then placed in the flow reactor and heated for 1 h in purified hydrogen at 550° C. Praseodymia and terbia require special treatment and are heated in hydrogen at 800° C for 12 h. A 1 : 1 *ortho-para* hydrogen mixture is then passed over the catalyst at from 25 to 50 ml. per min at 1 atmosphere pressure and at 25° C. Zero extrinsic field conversions average about 20 per cent of equilibrium and are monitored by differential thermal conductivity. The effect of placing the catalyst in a magnetic field is measured by assembling the reactor in the gap of a large electromagnet or, for fields of less than 40 oersteds (3,000 A m^{-1}), in a coreless solenoid of 5,000 turns. Fields are monitored by a commercial Hall effect instrument sensitive to 0·1 oersted (7·95 A m^{-1}).

In the conditions of the experiment no catalyst (except possibly the alumina-supported Tb_2O_3) shows any measurable hydrogen–deuterium equilibration; the applied field causes no change of hydrogen flow rate, no temperature effect and no shift of equilibrium. The changes in conversion rates appear to occur instantaneously, or almost so, on application of the field, and they are completely reversible. No such effects have been observed with the diamagnetic rare earths.

Fig. 1 shows the rate changes observed over supported Tb_2O_3 on so-called gamma-Al_2O_3 as a function of extrinsic field applied to the catalyst. This sample, prepared by impregnation of alumina with an aqueous solution of terbium nitrate, contained 0·14 atom per cent of Tb_2O_3 on a specific surface of 160 m^2. The rate changes are expressed as the fraction $(k_H-k_0)/k_0$ where k_H is the specific rate in a field H and k_0 that in zero extrinsic field. The value of k_0 was about 1×10^{-8} mole o-H_2 formed per second per cm^2 (of catalyst surface). This is probably accurate within a factor of two, a major source of uncertainty lying in the specific surface of the Tb_2O_3. The rate changes are quickly and accurately determined by turning the magnet current on or off. The determination is thus essentially differential and, consequently, the precision is high. The minimum change of rate detectable was about ±0·5 per cent of the zero field conversion rate.

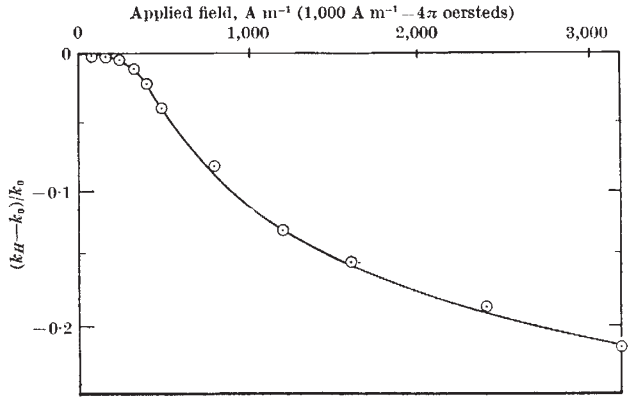

Fig. 1. Fractional change in the parahydrogen conversion rate over Tb_2O_3/γ-Al_2O_3 when the catalyst is placed in a magnetic field.

Fig. 1 shows the rate changes observed up to a field of 3,180 A m^{-1}. At much higher fields the rate rises as previously described for Cr_2O_3 above the Néel point[1,2], although the rise is much less for the supported Tb_2O_3 than for the pure rare earths[2]. It appears that the negative low-field effect described is not related to the much larger positive effects observed at high fields.

It will be noted in Fig. 1 that the effect of the extrinsic field changes rapidly, with field, in the region of 400–800 A m^{-1}. This suggests that, in this region, the Earth's magnetic field may be sufficient to influence the conversion rate. That this is indeed the case is readily shown by reversing the current in the solenoid. With the solenoid vertical and the angle of declination about 60°, reversal of the current gives a net resultant field change of about 80 A m^{-1}. It was found that, at 429 A m^{-1}, reversal of the current gave a readily perceptible change of rate, the greater change being found when the field produced by the solenoid was in the same direction (or nearly so) as the Earth's field. Field reversal gave no effect below 160 A m^{-1} and little or none above 8,000 A m^{-1}.

If the interaction of a molecule and a surface may thus be influenced by the Earth's magnetic field it would appear that a re-examination might be warranted of the possible role of terrestrial magnetism in such diverse areas as the navigational instincts of birds and the exclusive occurrence of L-amino-acids in natural proteins.

This work was supported under grant from the Army Research Office (Durham).

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Received May 21; revised June 29, 1970.

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BIOLOGICAL SCIENCES

Evidence for a New Haemoglobin Chain (ζ-Chain)

THE possibility of a human embryonic haemoglobin was first indicated by the work of Drescher and Kunzer[1]. Two such haemoglobins, Hb-Gower 1 and Hb-Gower 2, have been discovered[2,3] and their structures are ε_4 and $\alpha_2\varepsilon_2$, respectively. The ε-chain is a distinct haemoglobin chain synthesized during embryonic development. It has therefore been suggested[4] that there is a separate genetic locus determining the structure of the ε-chain, just as there