

- 17 D. Astruc, *Angew. Chem. Int. Ed. Eng.*, 1988, **27**, 643
- 18 J. Hawecker, J.-M. Lehn and R. Ziessel, *J. Chem. Soc., Chem. Commun.*, 1985, 56
- 19 H. D. Abruna, *Coord. Chem. Rev.*, 1988, 156
- 20 J.-F. Andre and M. S. Wrighton, *Inorg. Chem.*, 1985, **24**, 4288
- 21 J. V. Caspar, B. P. Sullivan and T. J. Meyer, *Inorg. Chem.*, 1987, **26**, 4145
- 22 M. Bakir, K. Wilbourn, R. J. Murray, T. J. Meyer and B. P. Sullivan, manuscript in preparation
- 23 M. Bakir, T. J. Meyer and B. P. Sullivan, manuscript in preparation
- 24 T. R. O'Toole, T. J. Meyer and B. P. Sullivan, manuscript in preparation
- 25 P. Braunstein, D. Matt and D. Nobel, *Chem. Rev.*, 1988, **88**, 747

## The Clean-Up of Fusion Reactor Waste Gases

### A STUDY OF THE EFFECTS OF IMPURITIES ON SILVER-PALLADIUM

The viability of nuclear fusion reactors will depend, in part, upon the effectiveness of the fuel clean-up system. One of the purposes of this system is to separate hydrogen isotopes from impurities in the exhaust emanating from the plasma. Such treatment can be achieved by cryogenic techniques, but these are made more effective if a preliminary removal of impurities can be performed. The technique favoured for this first stage is diffusion through palladium alloy membranes, which has been used commercially for many years to produce high purity hydrogen for use in a wide range of industrial applications including the electronics industry.

The alloy most commonly used for hydrogen purification is silver-palladium, with silver contents in the range 20 to 25 weight per cent. The operating regime is predominantly hydrogen rich, at pressures between 1 and 2 MPa and temperatures between 570 and 720K. Under these conditions no loss of activity of the alloy membrane surface due to carbon monoxide, carbon dioxide or water present as impurities has been reported.

In the potential fusion reactor fuel application the aim is to separate the hydrogen isotopes from impurities such as  $C(H,D,T)_4$ ,  $(H,D,T)_2O$  and  $N(H,D,T)_3$ , as well as from carbon monoxide, carbon dioxide, oxygen and nitrogen. Thus it is desirable to restrict operating temperatures and pressures as far as possible, in order to limit diffusion through the structural components of the system. Under such conditions, however, loss of membrane activity due to poisoning by impurities is possible; the most likely cause being related to chemisorption onto active alloy sites of impurity molecules, with a subsequent loss of these sites for the promotion of through-membrane diffusion. A reduction in the operating temperature will promote chemisorption, but at the expense of desorption, and there has been a lack of information about the effect of large amounts of impurities upon

membrane behaviour. However, a recent paper from the Centre d'Etudes Nucléaires de Saclay gives experimental data for the diffusion of hydrogen through palladium-silver in the presence of the impurities expected to occur in fusion reactor waste gases (J. Chabot, J. Lecomte, C. Grumet and J. Sannier, *Fusion Technol.*, 1988, **14**, (2), part 2A, 614-618).

The French team investigated palladium-23 silver under partial pressures of hydrogen of 14 kPa, and temperatures between 420 and 725K. Their findings indicate that methane and carbon dioxide have a depressing effect upon the hydrogen permeability at temperatures below 425K, when present in concentrations of 9 volume per cent in a carrier of helium. However, carbon monoxide at 0.2 volume per cent was found to severely degrade hydrogen permeability, under the same conditions. The detrimental effect of carbon monoxide was found to extend up to 570K, as the concentration was increased to 9.5 volume per cent. No synergism between the various impurity gases was observed, and the effect of carbon monoxide in mixtures was primarily that equivalent to the concentration of carbon monoxide alone. Regeneration of the membrane was found to be possible by thermal treatment, either in vacuo at 523K or by surface oxidation/reduction treatments.

A second valuable finding from this work is that a significant carbon monoxide/hydrogen reaction was observed at temperatures in excess of 650K, with the formation of methane, carbon dioxide and water, probably as a result of the catalytic activity of the alloy membrane surface. Thus this work has defined limits within which the utilisation of palladium alloy membranes for the purification of fusion reactor feed gas appears to be feasible, that is within the temperature range 475 to 650K.

The same group are currently considering the influence of other gaseous contaminants on this process.

D.R.C.