

109

## Effect of Iodine on Platinum-Catalyzed Combustion of Residual Hydrogen in Nuclear Power Plants

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### INTRODUCTION

In the secondary cycle of a nuclear power plant, hydrogen is produced. Its concentration is low, about 2% in air, but nevertheless it must be eliminated because of the danger of explosion. Two main methods are currently used for this process: (i) its catalytic combustion, or (ii) its burning in flame. In the former process, a Pt/Al<sub>2</sub>O<sub>3</sub> catalyst is applied. During the operation of the plant, several elements and compounds are formed, e.g. iodine, which may behave as catalyst poisons. In this paper we report on results concerning the poisoning effect of iodine on the transformation of hydrogen in air over a Pt/Al<sub>2</sub>O<sub>3</sub> catalyst.

The reaction conditions do not exactly mimic the real situation in the plant, as the iodine concentration in this work is much higher than can be expected in the plant and the Pt/Al<sub>2</sub>O<sub>3</sub> is not the industrial catalyst (it was made in our laboratory); nonetheless, it seemed worthwhile to carry out the investigation because, at least to our knowledge, very little information is available about the poisoning effect of iodine on any kind of metal-catalyzed transformation (1, 2).

### EXPERIMENTAL

A 2.5% hydrogen-in-air mixture was prepared by mixing H<sub>2</sub> and N<sub>2</sub> with O<sub>2</sub> in a calibrated rotameter. This mixture was passed through a flow reactor equipped with a cold trap after the reactor and with a sampling device. The reaction was run over a 0.43% Pt/Al<sub>2</sub>O<sub>3</sub> catalyst, prepared by impregnation. The samples were analyzed by gas chromatography. The signal of hydrogen was monitored.

The poisoning experiments were performed with I<sub>2</sub>. Iodine was placed in a saturator and its vapor was washed by the gas stream onto the catalyst. The iodine stream corresponded approximately to a 15 Pa gas flow. Experiments without poison were performed in the temperature range 323–723 K, and the poisoning measurements at 523–723 K.

### RESULTS AND DISCUSSION

The activity of the catalyst was appreciable (conversion: 83%) even at 323 K. This activity increased with elevation of temperature, but full conversion required 723 K. It was surprising though that the temperature range 423–723 K commonly applied in the plant is not enough for the complete removal of residual hydrogen. This sort of self-poisoning is quite probably due to the blocking effect of adsorbed water, which desorbs completely only at 723 K.

Treatment with iodine exerted a dramatic effect on the catalyst. A short (5 min) treatment under an iodine stream caused complete deactivation at both 523 K and 623 K. Switching off the iodine stream and purging the catalyst with the reactants for some time resulted in partial reactivation of the catalyst (523 K: a 3-hour purge, 26% activity regain; raising the temperature to 623 K: a 1-hour purge restores 93% of the original activity observed at this temperature). The completely poisoned catalyst at 623 K could be partially revitalized at 723 K and on switching back the iodine stream only partial deactivation occurred (65% activity loss after 10 min on stream). At this temperature 87% of the original activity was restored by only a 10-min purge with the pure reactants. The experimental results are summarized in Tables 1 and 2.

Table 1 Temperature dependence of unpoisoned reaction

temp.(K)	conversion(%)
323	83
523	86
623	89
723	100

Table 2 Effect of iodine treatment and reaction procedures through (a) purging with reactant stream, (b) raising temperature

temp. (K)	I <sub>2</sub> treatment (min)	purge with reactant (h)	conversion (%)
523	5	-	0
		1(a)	5
		2(a)	11
		3(a)	23
623	5	-	0
		1(a)	80
723(b)	10	-	35
		1/6(a)	87

At lower temperatures, the iodine stream results in complete coverage of the surface via multilayer adsorption with both physically and chemically adsorbed species. The pure reactant flow removes the excess iodine and the chemisorbed layer does not cover the entire surface (3): partial reactivation occurs. At the highest temperature applied, there is extensive iodine desorption; complete deactivation does therefore not occur, even under an iodine stream, and in its absence a short purge with the pure reactants provides an easy reactivation route.

#### CONCLUSIONS

The above set-up and results, in spite of the differences from the real conditions, indicate the following findings: (i) in the operating range of the plant, self-poisoning of the catalyst occurs, (ii) iodine poisoning is detrimental to the catalyst (the described set-up can be regarded as an accelerated poisoning procedure). Upon exposure to a  $10^{-4}$  Pa iodine flow, 0.37 surface iodine coverage develops over Pt(111) under UHV conditions (3). Consequently, the catalytic combustion of residual hydrogen can be safely applied in nuclear power plants only under rigorous checking conditions.

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