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THE JET HYDROGEN-OXYGEN RECOMBINATION SENSOR - A SAFETY DEVICE FOR HYDROGEN ISOTOPE PROCESSING SYSTEMS

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ABSTRACT

Fusion vacuum systems for processing of deuterium and tritium pose safety hazards by formation of potentially explosive mixtures in case of air leakage. Early detection of such hazards is essential for the safety of process equipment for fuel purification and recycling. Having established the fact that conventional oxygen monitors do not work well in hydrogen atmospheres at low pressures, we developed a sensor compatible with this duty. It is based on the detection of the oxygen-hydrogen reaction heat in a differential thermoelectric calorimeter, using a proprietary AECL wetproof catalyst for promoting the recombination reaction and semi-conductor thermopiles for detection. Operating at room temperature, the sensor provides signals in excess of 1mV for explosive hydrogen-air mixtures at pressures as low as 1mbar. The differential principle employed suppresses erroneous signals originating from hydrogen sorption/desorption heat on the catalyst.

The design and performance of these sensors will be described in detail.

Two of the sensors have been installed on the main forevacuum lines of the JET device to provide unambiguous alarms in case of air leakage. A previously developed (batch processing) oxygen detector reacting a sample in a vessel at 77K by means of a hot platinum filament (-900K) will then be used for exact quantitative assessment of the combustible mixture. Depending on the result, a suitable safe process route will be selected.

INTRODUCTION

The development of controlled thermonuclear fusion to a source of energy involves the handling of the hydrogen isotopes deuterium and tritium at low pressures (10^{-4} to 10 Pa) in the actual reactor and at higher pressures (up to approximately 100 kPa) in auxiliary gas purification and isotope separation systems. Such processes carry the risk of formation of explosive hydrogen/air mixtures, eg by leakage of air into the low pressure section of the process loop and by subsequent compression to atmospheric pressure. This risk would be aggravated by the release of the radioactive isotope tritium following explosive destruction of a process loop. It is therefore desirable to detect the presence of oxygen in hydrogen at sufficiently low pressures and concentrations as a warning for dangerous process leaks. Once identified, hazardous mixtures can then be safely purified, eg by selective absorption of O_2 on molecular sieve at cryogenic temperatures. Such procedures, being uneconomic and time

consuming, are not normally employed in the process main stream.

EXISTING OXYGEN MONITORING DEVICES

1) *Commercial oxygen detectors:*

A range of commercially available electrolytic O_2 detectors were tested on mixtures of air (1 to 5%) with hydrogen in the pressure range of 0.1 to 1 kPa. In this pressure range, the signals were found to be too low and irreproducible for reliable detection. Furthermore, we obtained information [1], that conventional electrolytic oxygen detectors operating at atmospheric pressure would exhibit erroneous signals when exposed to tritium, requiring frequent sensor replacement.

2) *JET batch recombination sensor:*

This device, described in detail earlier[2], measures the pressure of a gas sample in a sample flask at 77K before and after recombination by means of a platinum filament heated to 600°C. The pressure change observed corresponds to three times the oxygen partial pressure, since H_2O , the reaction product of $2H_2 + O_2$ exhibits negligible vapour pressure at 77K. The main disadvantage of this batch sampling method is the low repetition rate of ~ 5 minutes.

H_2-O_2 RECOMBINATION SENSOR

To avoid the disadvantages of the above described detection devices, a new sensor[3] was developed which is tritium compatible, stable and operates continuously at room temperature. Its principle is shown in Fig 1. It uses a proprietary wetproof AECL catalyst(3) originally developed for tritium sampling devices. The reaction heat is detected by a semiconductor thermopile(2). The thermopile is arranged in a pocket on a vacuum flange(1), the catalyst is retained by wire mesh(4). The signal(10) of the thermopile is a direct measure of reaction heat in the catalyst. A vacuum-tight envelope(5) and pressure gauges, hydrogen and air admission valves and a vacuum pump (6, 7, 8, 9) are used to adjust test mixture composition and flow rate. This device showed good sensitivity to pressures well below 1 kPa for hydrogen with 1 to 4% air, but still suffered from undesirable side effects:

- sensitivity to ambient temperature fluctuations;
- a strong (but quickly decaying) positive (= heat generation) output signal upon admission of pure H_2 or pure air due to the heat of adsorption of gas molecules on the catalyst surface;

- a strong (but quickly decaying) negative (= heat consumption) signal during pumpdown after tests due to the heat of desorption of gas molecules from the catalyst surface.

OD Conflat® flange, the mask on the reference sensor (nickel foil with 3mm DIA hole) is clearly visible.

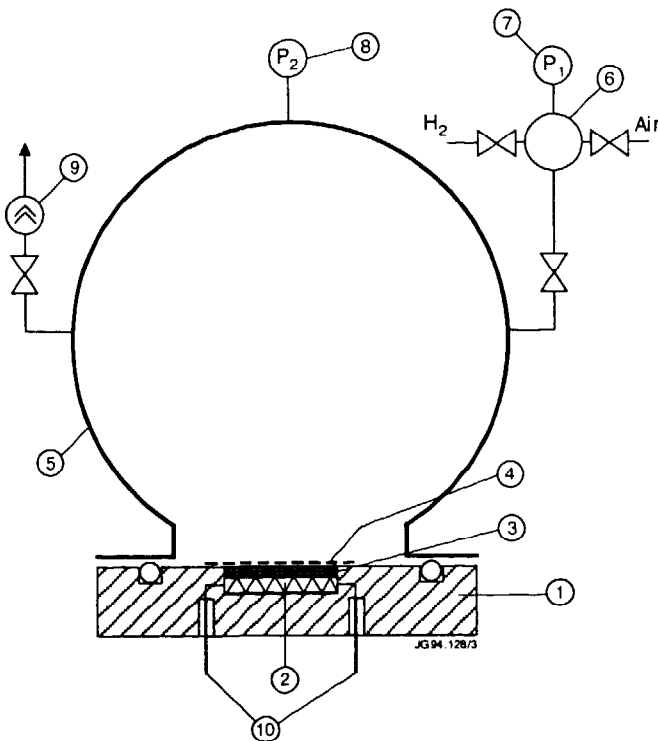


Fig 1 Schematic of single thermoelectric recombination heat detector

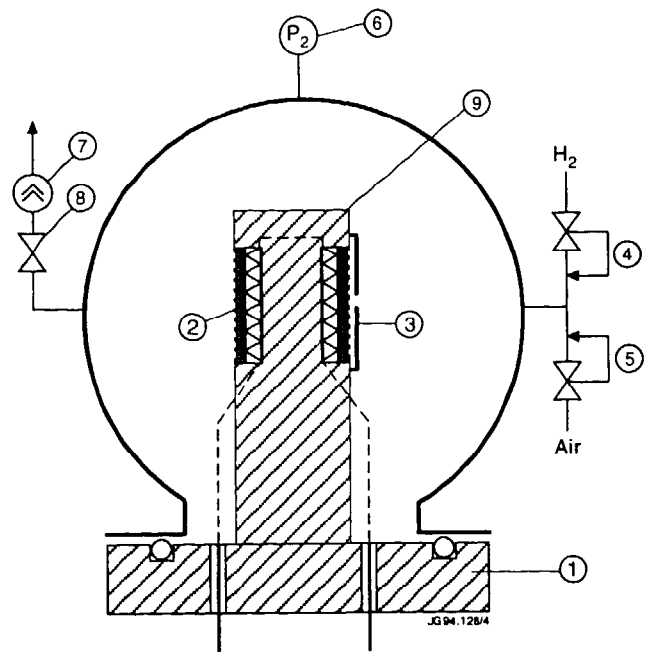


Fig 2 Schematic of differential sensor

DIFFERENTIAL H₂-O₂ RECOMBINATION SENSOR

These disadvantages were overcome by using a differential sensor shown in Fig 2: two identical sensors (2 and 3) are used, their signals being electrically subtracted. The main sensor (2) permits access for the reactant gas mixture over the full surface area of the catalyst, the reference sensor (3) is covered with a mask which reduces the exchange area between reactant gases and catalyst to about 1% of the sensor area. This device exhibits the expected improvements:

- insensitivity to temperature changes;
- in case of pressure changes (gas admission or pumpdown) the pressures of the majority gas in both catalyst pockets and therefore adsorption or desorption heat are virtually identical and hence cancel each other;
- the main sensor admits the reactant minority over the full area, whereas the masked reference sensor signal is two orders of magnitude lower. Hence, this differential sensor shows virtually the same sensitivity as the single sensor but simultaneously suppresses error signals due to temperature fluctuation and gas pressure changes.

Fig 3 shows the final version of the differential sensor: both main and reference sensor are arranged side by side on a 6"

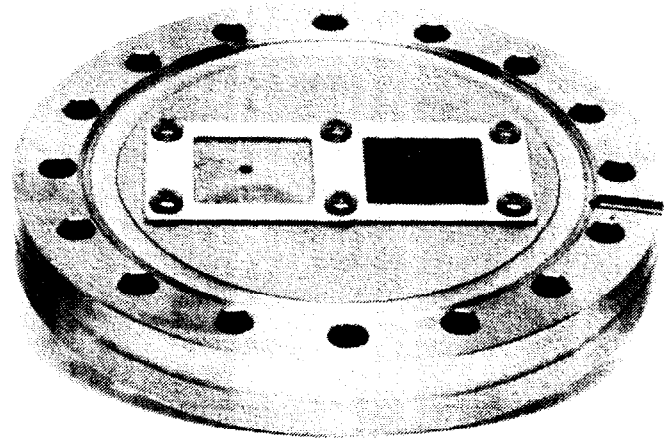


Fig 3 Differential sensor embedded in a vacuum flange

SAFETY CONSIDERATIONS

The prototype device was based on a catalyst already available at JET and used for oxidation of HT in atmospheric tritium samplers. As this catalyst was not necessarily appropriate for exposure to flammable mixtures, a functional test in a fire

test facility was carried out by puffing hydrogen gas onto the sensor area in air. This raised the catalyst temperature to ignition and showed that in this form the sensor did not appear acceptable for safety reasons. A minor modification remedied this. A triple layer of low transparency copper wire mesh covering both main and reference sensor fully suppressed ignition on hydrogen exposure in air. An alternative solution would be a catalyst specifically designed for safe recombination of flammable mixtures. (Note: It has subsequently been pointed out by AECL that the catalyst used, type 86-35, uses organic polymer support and was designed for use with low concentrations (<1 ppm to 1%) of H₂ (or HT) in humid air. AECL does not recommend its use at high H₂ concentrations where, as with any highly active catalyst, ignition may result.)

The addition of the copper screens reduced the sensor signal by approximately 50%. However, the signal is still sufficiently strong for unambiguous detection of flammable mixtures. This sensor is shown in Fig 4. Calibration tests were performed with 1 to 8% air in H₂ and 0.5 to 4% H₂ in air. These results are shown in Figs 5 and 6.

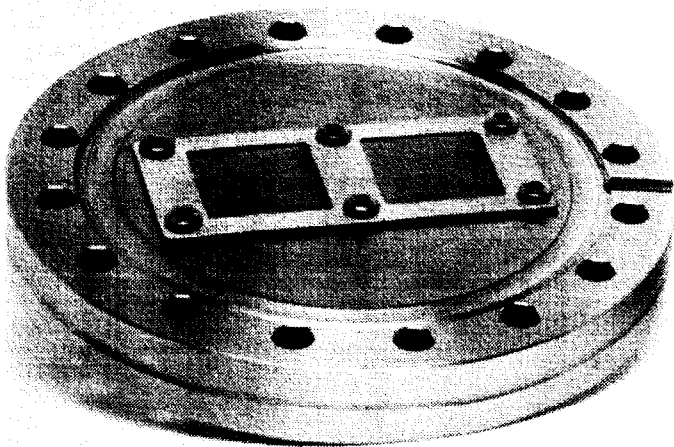


Fig 4 Differential sensor protected with copper mesh

Two sensors have been connected to the gas transfer lines between the JET Torus and the Active Gas Handling Systems[4]. The signals are connected to trip amplifiers raising alarms for signals exceeding 1 millivolt. In case of alarms indicating the presence of combustible gas mixtures, the composition of these mixtures is further analysed by use of the previously described batch recombination sensor and, as required, by mass spectrometry and gas chromatography. Depending on composition a safe process route can then be selected.

CONCLUSION

The new differential recombination sensor is suitable for on-line detection of flammable mixtures in hydrogen isotope processing systems in the pressure range from 0.1 to 100kPa. Due to their high sensitivity at atmospheric pressure, such

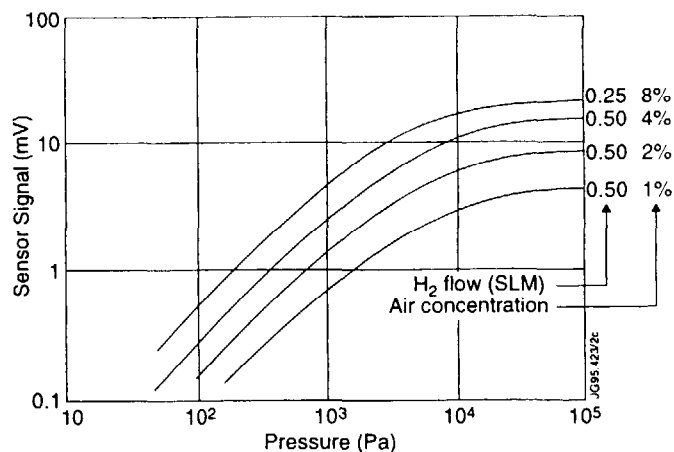


Fig 5 Sensor signal vs pressure for 1 to 8% air in hydrogen flowing across sensor array

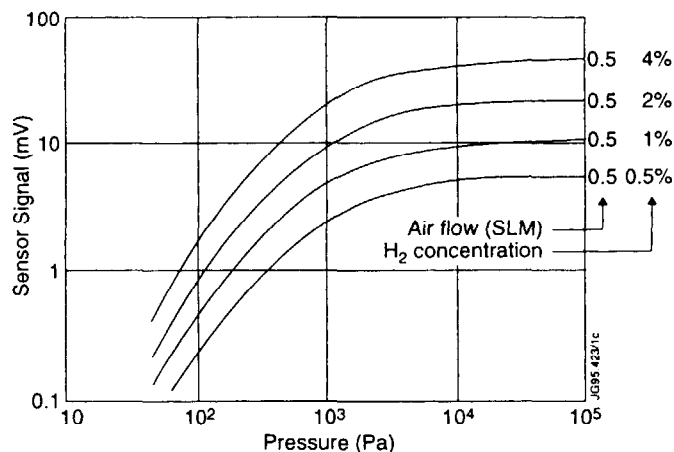


Fig 6 Sensor signal vs pressure for 0.5 to 4% hydrogen in air flowing across sensor array

sensors can also be used to detect traces of hydrogen in air (signal ~ 1 μ V/ppm H₂). Due to their intrinsic safety feature (no electrical supply voltages required) they can be installed in hazardous areas.

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