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Tritiated Water on Molecular Sieve: Water Dynamics and Pressure Observations

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Introduction

The production of fusion energy in a Tokamak using deuterium and tritium requires the safe handling and processing of exhaust gases that contain various amounts of tritium. Initial operation of the Tokamak Fusion Test Reactor (TFTR), Princeton Plasma Physics Laboratory, oxidized exhaust gases for tritium recovery or long-term storage. One of the most efficient and safest ways to contain tritiated water is to sorb it onto a pelletized 4A molecular sieve.[1] A Disposable Molecular Sieve Bed (DMSB) was designed as a pressure vessel[2] because of the possibility of pressure generation from the radiolysis of tritiated water on molecular sieve.[3] Hydrogen production contributes to the complexity of the containers used to transport and store tritiated water, and increases the fabrication costs.

Two months after removing a DMSB from the process at TFTR, a pressure in excess of that predicted from self-radiolysis was observed. Interestingly, pressure measurements at longer times (up to 2.5 years) showed less pressure than expected. Pressure was not being generated in the DMSBs at the predicted rate. This was unexpected and prompted an investigation into the mechanism responsible for the anomalous pressure measurements.

Description of Work

A mechanism for gas production from self-radiolysis of tritium contaminated water on molecular sieve has been discussed.[4] This report describes the exceptional observation that the over pressure in several DMSBs charged with tritium contaminated water was not the expected pressure. This report also describes a small pressure increase in a sealed DMSB not related to tritium decay. For these pressure measurements, the crucial observation was that the water was loaded on the DMSBs in a non-uniform way and continued to equilibrate to drier parts of the molecular sieve during storage. This mechanism was verified through bench scale experiments.

Results

Over 30 DMSBs were loaded with tritiated water during the high powered D-T experimental program at TFTR and returned to Westinghouse Savannah River Company. Pressure and composition of the gas phase were measured prior to regeneration. The Figure displays the pressure data from ten DMSBs. The beds contained from 0.2 to 2 grams of tritium and from 1 to 2.5 kilograms of water. After removal from the system, 2 months to 2.5 years elapsed prior to data collection. The calculated pressures from tritium self-radiolysis models are also displayed on the graph. The pressure data are plotted as a function of the amount of tritium that decayed for each bed. The average observed pressure in these DMSBs was approximately 0.131 MPa (980 Torr), and it was nearly independent of the loading and storage conditions.

Insignificant amounts of gaseous hydrogen isotopes (<0.2 mole percent) were observed before regeneration of the DMSBs, where several mole percent was calculated to have been produced. This is consistent with the lack of a pressure as a function of tritium decay. Analysis of the gas sample showed mainly nitrogen, helium-3 from tritium decay, and argon, with small amounts of oxygen. The lack of hydrogen gas in these analyses was exceptional. The small amount of oxygen observed probably came from bed exposure to the laboratory atmosphere during its removal from the system. After regeneration, the recovered tritium and water showed that tritium was present in the water, and that the amount of recovered tritium was approximately the amount loaded onto the beds, less the amount that decayed. But the pressure measurements showed that there was a small pressure increase (0.03 MPa, about 220 Torr) over atmospheric pressure with no hydrogen observed. A mechanism that would cause the pressure to be less than expected is the scavenging of hydrogen atoms, and/or their precursors, by the molecular sieve/binder combination before they can combine to form hydrogen gas. However, the anomalous, small pressure measurements with no hydrogen production is explained by the equilibration of non-uniformly distributed water, with concomitant displacement of nitrogen from the drier molecular sieve pellets. This mechanism is not related to tritium decay, and contributes nitrogen to the gas phase at a rate dependent on the degree of saturation and non-uniformity of water distribution. For these DMSBs, hydrogen production from radiolysis appears to be almost totally quenched, and small pressure increases over the initial storage pressure result from water migration to drier sections of the molecular sieve bed that displaces adsorbed nitrogen gas.

Should a molecular sieve/binder pellet combination be found that greatly depresses the rate of hydrogen production from self-radiolysis, a much safer and less expensive waste container for tritiated water could be obtained. Investigations into the material properties of molecular sieve/binder clays with respect to tritium self-radiolysis mechanisms are continuing.

References

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