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Tritium Compatibility of Alumina and Fosterite

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TRITIUM COMPATIBILITY OF ALUMINA AND FOSTERITE

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ABSTRACT

Many pressure measurements are required to control processing of the fuel gases associated with fusion power reactors. Since most pressure transducers respond to changes in pressure sensitive electrical parameters, insulators will be required to withstand chronic exposures to concentrated tritium. For this investigation samples of alumina and Fosterite were exposed to concentrated tritium gas for 11 weeks. Gas phase impurities were then analyzed for clues that would indicate decomposition of the exposed materials. The only gaseous impurity resulting from these tritium exposures was tritio-methane, which is always produced when tritium is stored in stainless steel containers. There was no evidence that either alumina or Fosterite decomposed in the presence of tritium.

1. INTRODUCTION

Many pressure and vacuum transducers are required to monitor tritium gas processes at the Los Alamos Tritium Systems Test Assembly (TSTA). For several TSTA subsystems capacitance manometers have been chosen because of their wide range, their insensitivity to gas composition, and other desirable features. One manufacturer* of these gauges uses Fosterite**, a naturally occurring ceramic composed of $2\text{MnO} \cdot \text{SiO}_2$, as an electrical insulator. Since both sides of the capacitance diaphragm must be exposed to tritium in differential pressure applications, there is some concern about long-term tritium compatibility

*NKS Instruments, Inc., 22 Third Ave., Burlington, MA 01803

**Alsimag 243, Technical Ceramic Prod. Div., 3M Products, Chatanooga, TN 37415

of the ceramic insulators used in these instruments. Because alumina (Al_2O_3) is a common insulator, already evaluated for fusion applications¹⁻³, a sample of this material was also included in the current experiment.

This compatibility study was limited to the observation of gaseous products due to radiation-induced decomposition. Since these insulators are refractory materials, no changes in physical properties are expected over reasonable periods of tritium exposure. Such changes would occur only in organic materials, which contain weakly bound elements that can react with tritium under the stimulation of beta radiation.

II. THE EXPERIMENT

MKS Instruments provided sample disks of alumina and Fosterite, identical to the insulators in their Baratron capacitance manometers. Each disk was 5 cm in diameter and 6 mm thick, yielding a surface exposure of about 50 cm^2 . Each sample was cut in half with a diamond saw and was enclosed in a 125-cm^3 stainless steel cylinder described previously⁵. Three cylinders, each attached to an isolation valve, were manifolded to a common fill/recovery valve and to a viscous leak for transmitting samples to a mass analyzer, as shown in Fig. 1. One cylinder each enclosed alumina (A) and Fosterite (C) samples, and a third empty cylinder (B) served as a control.

The impurity analyses of previous compatibility studies were difficult because water preadsorbed on the test specimens gave rise to tritiated water peaks that overlaid the range of interest (17-22 amu). This time these effects were minimized by heating the cylinders to 200°C under vacuum for 18 h. Cylinders A, B, and C were then simultaneously backfilled to 67 kPa at 23°C with tritium gas of the original composition as given in Table II. The samples and the control were stored under these conditions for 77 days.

III. THE RESULTS

At the end of the exposure period the viscous sampling manifold was connected to the ion chamber of an Inficon* IQ 200 quadrupole mass analyzer. Samples from each cylinder were run consecutively on this instrument, sweeping

*Inficon, Leybold-Heraeus, 6500 Fly road, East Syracuse, NY 13057

Fig. 1. Tritium exposure experiment.

from 2 to 40 amu. Sample pressures were approximately 60 kPa, whereas the viscous capillary leak reduced the pressure in the analyzer tube to approximately 0.13 mPa by limiting the sample flow rate to less than $1 \text{ Pa}\cdot\text{cm}^3/\text{s}$. The sample volume and analyzer tube were evacuated to high vacuum between successive samples, as confirmed by the complete disappearance of the tritium peak (6 amu) during evacuation. Mass spectra of samples A, B, and C, corrected for background, are presented in bar graph form in Fig. 2.

Although these data present a rather confused picture, it is evident that the mass spectra of exposure samples A and C are essentially identical to that of the control B. Previous experiments have established that tritium stored in stainless steel synthesizes methane^{4,5}. Since the largest peaks appear at 21 and 24 amu, tritio-methane is the principal impurity.

Arbitrary
Scale

Fig. 2. High mass spectra of exposure samples.

TABLE I
STANDARD FRAGMENT PATTERNS OF TYPICAL VACUUM CONTAMINANTS

Amu	Percent of Principal Peak ^a		
	Nitrogen	Methane	Water
12	...	2.4	...
13	...	2.7	...
14	7.2	15.6	...
15	...	85.8	...
16	...	100.0	1.1
17	...	1.2	23.0
18	...	...	100.0
28	100.0	...	...
29	0.8	...	...

^aData from Ref. 6.

The following unverified assumptions are made in order to complete the analysis:

1. The mass sensitivity of the quadrupole analyzer is uniform over the range of 12-28 amu.
2. Standard fragment patterns⁶ for ordinary methane are applicable to all isotopic forms and are correct for our particular mass analyzer. (Standard fragment patterns for methane, nitrogen and water are given in Table I.)
3. Only the methane fractions composed of all tritium atoms and those containing one atom of either hydrogen or deuterium are synthesized in significant quantity. For example, CT_4 , CHT_3 and CDT_3 are assumed to be present, but none of the other possible isotopic forms of methane ($CHDT_2$, CH_2T_2 , etc.) are considered. This assumption is valid because the hydrogen and deuterium impurities in the original tritium are limited to a few per cent.
4. Isotopic composition of the original tritium gas directly determines isotopic distribution in the synthesized methane fractions. Isotopic effects on the kinetics and equilibria are ignored.

The peaks appearing at 14, 28, and 29 amu are due to nitrogen from air inleakage and outgassing during the exposure period. A proportionate quantity of oxygen can also be assumed at 22 amu (T_2O), 19 amu (OT), and 16 amu (O). Free oxygen (O_2 at 32 amu) is never detected in tritium gas because it converts rapidly to tritiated water.

The spectra for methane are complex because the presence of all three hydrogen isotopes insures that every peak between 12 and 27 amu (C , CH , CD , CT , CHT , CDT , CT_2 , CHT_2 , CDT_2 , CT_3 , CHT_3 , CDT_3 , CT_4 , CHT_4 , CDT_4 , and CT_5) is represented. Assumption 3 eliminates consideration of compound peaks due to overlapping isotopic forms (for example, $CHDT_2$, which would add to the CT_3 peak at 21 amu). Of the allowable methane fractions the major peak at 24 amu can only be the principal molecule tritio-methane (CT_4), from which the other methane fractions and isotopic varieties can be derived using the original isotopic analysis (Table II) and the fragment pattern of methane (Table I).

Fig. 3. Residual spectra after subtracting air and methane.

When the nitrogen, water, and methane contributions are subtracted from the original spectra, the residuals shown in Fig. 3 result. About 95% of the impurity spectrum has been accounted for by air and methane, and the residual peaks at 12, 15, 16, and 21 amu are an order of magnitude smaller than those already identified. These residuals, which together account for less than 0.1% of the gas composition, probably represent experimental error. These peaks are quite characteristic of the methane spectra already discussed, and like them, are as evident in the control as in the exposure samples.

The final analysis is given in the last column of Table II. Since all three analyses were consistent within experimental error, a composite analysis is listed. The low mass compositions (helium and hydrogen isotopes) were determined on a cycloidal mass spectrometer* shortly after backfilling the samples, and again a few days before the impurity samples were run on the quadrupole machine. Again the isotopic ratios in all three samples were the same within experimental error. The quadrupole analyzer was not calibrated for mass sensitivity, so the stated high mass composition is approximate.

IV. CONCLUSIONS

During a 77-day exposure of alumina and Fosterite to concentrated tritium no gaseous products evolved that indicated any decomposition of these insulators. Previous observations that tritium stored in stainless steel

*Consolidated Electrodynamics Corporation, Model 21-620

TABLE II
COMPOSITION OF TEST GAS

<u>Component</u>	<u>Mole Per Cent</u>	
	<u>Original</u>	<u>Final</u>
Tritium	89.0	87.38
Deuterium	6.6	6.45
Hydrogen	0.8	0.77
Helium-3	3.6	4.00
Methane (CT ₄)		1.28 ^a
Water (T ₂ O)		0.02 ^a
Nitrogen		0.09 ^a

^aUnit sensitivity factor assumed for all atomic masses.

evolves tritio-methane were confirmed, and the synthesis rate was within the range of previous measurements. Vacuum bakeout of the samples before exposing them to tritium eliminated ambiguous impurity peaks from the post-exposure analysis.

There is no evidence from this experiment that would discourage the use of either alumina or Fosterite in instruments chronically exposed to tritium. Although no attempt was made to measure changes in electrical properties due to tritium exposure, refractory insulators have been judged suitable⁷ for fusion chambers. The tritium beta radiation constitutes a much milder environment than does the radiation field in the plasma chamber itself.

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