CAPABILITIES AND RESOURCES
FOR TRITIUM RESEARCH
AND DEVELOPMENT

February 7, 1974

MOUND LABORATORY
Miamisburg, Ohio
operated by
MONSANTO RESEARCH CORPORATION
a subsidiary of Monsanto Company
for the
U. S. ATOMIC ENERGY COMMISSION
U. S. Government Contract No. AT-33-1-GEN-53
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
Capabilities and Resources for Tritium Research and Development

Excerpts from a Presentation by Mound Laboratory at AEC Headquarters

February 7, 1974

AGENDA

Introduction ................ Warren H. Smith
Metal-Hydrogen Systems .......... Richard J. DeSando
Gas Dynamics and Cryogenic Separations .... William L. Taylor
Materials Research ............... Japnell D. Braun
Helium Diffusion and Tritium Effluent Control . Carl J. Kershner

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
F - CENTER CONCENTRATION AS INDICATED BY EPR

CORRECTED EPR SIGNAL STRENGTH, ARBITRARY UNITS

U. V. IRRADIATION TIME, MIN

4.2°K

N_F^- t^{1/2} I^{1/2}

1.4°K

78°K
ISOTHERMS, 200°C
Temperatures in °C

Pressure, atm

Composition, T/U
\[ \log P_{\text{sto}} UT_3 = \frac{-4038.2}{T} + 6.074 \]

- UD$_3$ - Carlson
- UD$_3$ - Wicke and Otto
- UH$_3$ - Northrup

Temperature: 763°C

Pressure, atm

1000/T, °K

1000°C 500°C
THERMODYNAMIC PROPERTIES OF URANIUM HYDRIDES

<table>
<thead>
<tr>
<th></th>
<th>$\Delta H_s$ (kcal/mole)</th>
<th>$\Delta S_s$ (cal/°K/mole)</th>
<th>$\Delta G_s$ (kcal/mole)</th>
</tr>
</thead>
<tbody>
<tr>
<td>UH$_3$</td>
<td>-30.5</td>
<td>-43.4</td>
<td>-17.65</td>
</tr>
<tr>
<td>UD$_3$</td>
<td>-30.8</td>
<td>-44.8</td>
<td>-17.5</td>
</tr>
<tr>
<td>UD$_{2.66}$</td>
<td>-28.4</td>
<td>-42.1</td>
<td>-15.9</td>
</tr>
<tr>
<td>UT$_{2.90}$</td>
<td>-27.7</td>
<td>-41.7</td>
<td>-15.3</td>
</tr>
</tbody>
</table>
METAL-HYDROGEN SYSTEMS

Selected Bibliography


C. M. Love and L. J. Nielson, "Physical Properties of Irradiated Lithium Hydride," (U), MLM-CF-70-9-63 (SRD); MLM-CF-70-12-43 (SRD) and MLM-CF-71-6-349 (SRD).


Papers to be published:


R. C. Bowman, Jr., and A. Attalla, "Pulsed NMR Study of Heavily Irradiated Lithium Hydrides," (U), Mound Laboratory Report (SRD).
CRYOGENICS

DISTILLATION
$H_2/D_2/T_2\ He^3/He^4$

ISOTOPIC VAPOR PRESSURES

ADSORPTION ISOTHERMS $T_2/He$

PRESENT STAFF
1 PhD PHYSICAL CHEMISTRY
2 PhD PHYSICS
2 B.S. PHYSICS
1 TECHNICIAN

GAS DYNAMICS

MOLECULAR BEAM

THERMAL DIFFUSION

ORDINARY DIFFUSION

THEORETICAL AND PROGRAM LIBRARY
BLOCK FLOW DIAGRAM OF HYDROGEN ISOTOPE DISTILLATION SYSTEM

FROM STORAGE

PRESSURE REGULATOR

FLOW METER

LIQUID NITROGEN TRAP

H₂ + HD + D₂

TO STORAGE

COMPRESSOR

FLOW METER

NEEDLE VALVE

H₂

TO STORAGE

COMPRESSOR

FLOW METER

HD + D₂

NEEDLE VALVE

TO STORAGE

REFRIGERATOR

77 K

DISTILLATION COLUMN

22 K
SCHEMATIC OF 2 COLUMN DISTILLATION APPARATUS FOR THE SEPARATION OF PROTIUM FROM DEUTERIUM

H₂, HD, D₂

Catalyst

25% H₂
25% D₂
50% HD

COLUMN 1

~100% HD

HD

COLUMN 2

D₂
DIAGRAM OF VAPOR PRESSURE MEASUREMENT SYSTEM

- BARATRON PRESSURE SENSORS
- PRESSURE METERS
- CHARCOAL TRAP
- STORAGE CONTAINER
- LIQUID HELIUM DEWAR
SIMPLE SINGLE COMPONENT ADSORPTION ISOTHERM APPARATUS

CALIBRATED VOLUME

DIFFERENTIAL PRESSURE GAGE

TO VACUUM PUMPS

FROM GAS SUPPLY

COLD CELL WITH ADSORBANT

LIQUID NITROGEN DEWAR
Adsorption isotherms at 77.2 K for hydrogen on a coconut charcoal and two molecular sieves.

- Coconut charcoal
- Linde 4A mol. sieve
- Linde 5A mol. sieve
TOTAL SCATTERING CROSS SECTIONS

SCATTERING CHAMBER

ARM A (not used)

SIGNAL AVERAGER $\Phi$

DETECTOR

ATTENUATED INTENSITY $I'$

TARGET GAS FEED

ARM B

COLLIMATING CHAMBER

SOURCE CHAMBER

TARGET GAS CELL WITH GAS DENSITY $n$

VELOCITY SELECTOR

CHOPPER $\Phi$

SKIMMER

NOZZLE SOURCE $B$

METHOD

FEED $B$
THERMAL DIFFUSION FACTORS FOR Kr

\[ \alpha_t \times 10^2 \]

- DERIVED FROM SELF-DIFFUSION COEFFICIENTS AND VISCOSITY DATA
- DIRECTLY MEASURED VALUES, PAUL AND WATSON

TEMPERATURE, °K
PROGRAM LIBRARY

- MULTICOMPONENT DISTILLATION
- THERMOMOLECULAR PRESSURE DIFFERENCE SOLUTION
- QUANTUM PHASE SHIFTS
- TRANSPORT COLLISION INTEGRALS
- TRANSPORT PROPERTIES
- SCATTERING CROSS SECTIONS
GAS DYNAMICS AND CRYOGENIC SEPARATIONS

Selected Bibliography


G. T. McConville, Derivation of Modified Weber-Schmidt Equa for Thermal Transpiration, MLM-1627 (June 1, 1969), 18 pp.


B. L. van der Waerden, Theory of the Trennschaukel, MLM-1181 (TR) (October 14, 1963), 62 pp. [translated by W. L. Taylor from Zeitschrift fur Naturforschung, 12a, 583-598 (1957)].


Papers to be Submitted


W. L. Taylor and Stanley Weissman, "Experimental Thermal Diffusion Factors for $^{20}$Ne-$^{22}$Ne and Their Applications as a Test of the Neon Interatomic Potential," (accepted by J. Chem. Phys.).
Patent Application


In Preparation


W. L. Taylor, "Thermal Diffusion Factors for Xenon" (to be submitted to J. Chem. Phys.).
SPECIALIZED MECHANICAL TESTING EQUIPMENT

- HIGH-TEMPERATURE, HIGH-VACUUM CREEP TESTERS
- HIGH-TEMPERATURE, HIGH-VACUUM FATIGUE TESTER
- HIGH-TEMPERATURE, HIGH-VACUUM, LARGE (6 x 8 in.) ANNEALING FURNACE
- MTS TEST SYSTEM LOCATED IN TRITIUM-CONTROLLED GLOVE BOX LINE
- HIGH-TEMPERATURE IMPACT FACILITY
RESEARCH ANALYTICAL EQUIPMENT

- TRANSMISSION ELECTRON MICROSCOPES
- SCANNING ELECTRON MICROSCOPES
- X-RAY EQUIPMENT
- ELECTRON MICROPROBE
TRITIUM CONCENTRATION (ppm by wt.)

DIFFUSION COEFFICIENT (cm²/sec)

\[ D = 1.24 \times 10^{-9} \exp \left( \frac{-41.17}{301.9 - C} \right) \text{ cm}^2/\text{sec} \]
AUTORADIOGRAPHS

A.
ANNEALED
STAINLESS STEEL
1000 X

B.
COLD-WORKED
STAINLESS STEEL
1000 X

C.
ENHANCED PENETRATION
IN WELD METAL
100 X
<table>
<thead>
<tr>
<th>MATERIAL</th>
<th>SPECIMEN THICKNESS (cm)</th>
<th>ISOPOE</th>
<th>MEASURED PERMEABILITY CONSTANT Q (cm²/sec·atm)</th>
<th>LITERATURE VALUE (cm²/sec·atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Latex</td>
<td>0.018</td>
<td>H₂</td>
<td>3.5 x 10⁻⁷</td>
<td>3.9 x 10⁻⁷</td>
</tr>
<tr>
<td>Latex</td>
<td>0.018</td>
<td>D₂</td>
<td>3.0 x 10⁻⁷</td>
<td></td>
</tr>
<tr>
<td>Mylar</td>
<td>0.005</td>
<td>H₂</td>
<td>4.5 x 10⁻⁹</td>
<td>4.4 x 10⁻⁹</td>
</tr>
<tr>
<td>Mylar</td>
<td>0.005</td>
<td>D₂</td>
<td>4.1 x 10⁻⁹</td>
<td></td>
</tr>
<tr>
<td>Teflon</td>
<td>0.007</td>
<td>H₂</td>
<td>6.1 x 10⁻⁸</td>
<td></td>
</tr>
<tr>
<td>Teflon</td>
<td>0.007</td>
<td>D₂</td>
<td>5.7 x 10⁻⁸</td>
<td></td>
</tr>
<tr>
<td>Saran</td>
<td>0.001</td>
<td>H₂</td>
<td>3.6 x 10⁻⁸</td>
<td>3.1 x 10⁻⁷</td>
</tr>
<tr>
<td>Saran</td>
<td>0.001</td>
<td>D₂</td>
<td>2.3 x 10⁻⁸</td>
<td></td>
</tr>
</tbody>
</table>
HELIUM DIFFUSION AND PERMEABILITY STUDIES

- PuO₂ PLASMA-FIRED MICROSPHERES
- PuO₂ SOL-GEL MICROSPHERES
- Pu METAL
- PuO₂-Mo CERMET
- REFRACTORY METALS

TRITIUM DIFFUSION IN STAINLESS STEELS
MECHANICAL TESTING OF DOPED METALS AND ALLOYS

- TENSILE TESTS
- HOT IMPACT STUDIES

CREEP STUDIES OF REFRACTORY METALS AND ALLOYS

- TENSION vs COMPRESSION
- SUBSTRUCTURE CORRELATIONS

MECHANICAL TESTING OF IRIDIUM
MECHANICAL TESTING OF NEUTRON IRRADIATED MATERIALS

- COMPRESSION CREEP
- DYNAMIC ELECTRON MICROSCOPE STUDIES
- SUBSTRUCTURE CORRELATIONS


Helium Diffusion and Tritium Effluent Control
AN ADVANCED TECHNOLOGY PROJECT WAS INITIATED IN 1972 TO ACHIEVE LONG RANGE GOALS FOR REDUCING TRITIUM EMISSIONS

- Facility Evaluation
- Basic Decontamination & Concentration Technology
- Complete Processes & Systems
- High Sensitivity Monitoring Instrumentation
THERE IS A WIDE VARIATION IN QUANTITY AND CONTAMINATION LEVEL OF MOUND’S GASEOUS EFFLUENTS

<table>
<thead>
<tr>
<th>CLASS</th>
<th>TYPICAL SOURCE</th>
<th>CONCENTRATION (Ci/m³)</th>
<th>HTO (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-g</td>
<td>G. B.</td>
<td>1-100</td>
<td>1-10</td>
</tr>
<tr>
<td>II-g</td>
<td>Room</td>
<td>~4x10⁻⁵</td>
<td>—</td>
</tr>
<tr>
<td>III-g</td>
<td>Stack</td>
<td>~1x10⁻⁵</td>
<td>10-90</td>
</tr>
</tbody>
</table>
There is a wide variation in quantity and contamination level of Mound's liquid waste.

<table>
<thead>
<tr>
<th>CLASS</th>
<th>TYPICAL SOURCE</th>
<th>CONCENTRATION (μCi/ml)</th>
<th>QUANTITY (l/yr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-ℓ</td>
<td>ERS (aqueous)</td>
<td>≥11-ℓ</td>
<td>&gt;11-ℓ</td>
</tr>
<tr>
<td>II-ℓ</td>
<td>Pump Oil</td>
<td>4×10⁴</td>
<td>~250</td>
</tr>
<tr>
<td>III-ℓ</td>
<td>Decon. H₂O</td>
<td>1</td>
<td>60,000</td>
</tr>
</tbody>
</table>
EXPERIMENTAL WORK IS IN PROGRESS ON DECONTAMINATION TECHNOLOGY BASED ON:

- Cryogenic Adsorption (Class I-g)
- Getters (Class I-g)
- Extractive Distillation (Class III-ℓ)
- Molecular Excitation (Class III-ℓ)
- Catalytic Oxidation/Adsorption (ERS, ECS, Class I & II-g)
CRYOGENIC PROCESS SCHEMATIC FOR TRITIUM CONTROL IN GLOVEBOX ATMOSPHERE SYSTEMS

GLOVEBOX LINE → SUCTION VOLUME → BLOWER

HEAT EXCHANGERS

G D R

SMALL ADSORPTION SEPARATION

CRYOGENIC STILL → RECOVERED TRITIUM

MOLECULAR SIEVE DRYERS

D R

WASTE AIR

TRITIATED WATER
SYSTEM SCHEMATIC FOR EMERGENCY CONTAINMENT OF TRITIUM

1. RECYCLE AIR
2. VENT TO STACK
EXPERIMENTAL WORK IS IN PROGRESS ON RECOVERY TECHNOLOGY BASED ON:

- Cryogenic Distillation (Gas)
- Pd Column Separation (Gas)
- Fuel Cell/Electrolysis (Liquid)
- Chemical Decomposition (Liquid)
THE "TECL" DESIGN IS BASED ON A CLOSED $^3$H CYCLE CONCEPT

< 10% RCG AT STACK

GAS

EXPERIMENTS

WATER

~ZERO AQUEOUS WASTES
TRITIUM EFFLUENTS HAVE BEEN REDUCED BY MORE THAN AN ORDER OF MAGNITUDE

![Bar chart showing tritium effluent from 1969 to 1980.](chart.png)
A VARIETY OF HELIUM DEPOSITION & MIGRATION STUDIES HAVE BEEN CARRIED OUT AT MOUND

- $^4\text{He}$ IN $^{238}\text{PuO}_2$ ($\alpha$ DECAY IMPLANTED)
- $^4\text{He}$ PERMEATION THROUGH TANTALUM
- $^3\text{He}$ IN TANTALUM ($T_2$ DECAY IMPLANTED)
A HIGH SENSITIVITY QUADRUPOLE MASS SPECTROMETER SYSTEM HAS BEEN DEVELOPED FOR He DIFFUSION STUDIES
PERMEABILITY OF $^3$He THROUGH TANTALUM METAL HAS BEEN DEMONSTRATED AND A MECHANISM POSTULATED

<table>
<thead>
<tr>
<th>VACANCY</th>
<th>INT.</th>
<th>MEAS.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Q_p$</td>
<td>202</td>
<td>110</td>
</tr>
<tr>
<td>D</td>
<td>$6 \times 10^{-11}$</td>
<td>$1 \times 10^{-6}$</td>
</tr>
<tr>
<td>S</td>
<td>$1 \times 10^6$</td>
<td>$1 \times 10^{10}$</td>
</tr>
<tr>
<td>P</td>
<td>$9 \times 10^{-5}$</td>
<td>$1 \times 10^4$</td>
</tr>
</tbody>
</table>
HELIUM DIFFUSION AND TRITIUM EFFLUENT CONTROL

Selected Bibliography


