calculated critical condensation flux density at the collector for condensation of cadmium is indicated to be between 0.81×10^{-9} and 1.14×10^{-9} g/cm^2-sec for collector temperatures of 23° and 68°F.

At (relatively) high cadmium flux densities (e.g., $G = 10^{-8}$ g/cm^2-sec), cadmium condensed on aluminum (placed in the system for shielding purposes) at something above room temperature. Condensation was indicated on the copper (e.g., $G = 5 \times 10^{-8}$ g/cm^2-sec) at about 100°F, with strong condensation at 130°F. Figure 3 is a picture showing the manner of cadmium condensation when maximum temperatures of about 158°F were attained on one-half of the collectors.

Collectors cleaned in various manners, or deliberately soiled, were tried. In general, little difference was found in uniformity, or lack of uniformity of condensation, except for silicone grease and finger grease, both of which prevented cadmium condensation as illustrated by the two half-coated collectors in Fig. 3.

Conclusions

Electroplated cadmium has an apparent rate of evaporation (G) in high vacuum that is about 0.02 times that of the calculated rate for pure bulk cadmium (G_T); by study of the plating conditions, surface, and/or post-plating treatment, this factor might be decreased to 0.001.

The results obtained in this work tend to support the view of Sennett et al.[13] that a certain critical flux density is necessary for condensation, not only for glass, but for other materials as well. Extrapolation of the data presented by Palatnik and Gladkikh[16] indicates that the mechanism and condensation anomaly presented by them very probably explain the so-called anomalous results obtained in this work and by Sennett et al.[13]

No final conclusions can be drawn as to the applicability of electroplated cadmium in the space environment, but the case for use of cadmium under space environmental condition appears somewhat more favorable than some have thought. The following areas need further study: 1) Why does electroplated cadmium have a lower G than pure bulk cadmium? 2) What conditions affect its G? 3) What are the effects of surface upon the condensation? 4) How much does the surface and its cleanliness affect the critical condensation rate? 5) Is the critical condensation rate explained by the mechanism presented by Palatnik and Gladkikh?[16]

References

[1] Gatzek, L. E., "Some corrosion problems of missiles in silo storage," Corrosion **17,** 28–29 (1961).

[2] Jaffe, L. D. and Rittenhouse, J. B., "Behavior of materials in space environments," Jet Propulsion Lab. TR 32-150, p. 68 (November 1961).

[3] Dushman, S., *Scientific Foundations of Vacuum Technique* (John Wiley and Sons, Inc., New York, 1949), pp. 18–24.

[4] Gloria, H. R., Stewart, W. J., and Savin, R. C., "Initial weight loss of plastics in a vacuum at temperatures from 80° to 500°F," NASA TN D-1329 (December 1962).

[5] Lange, N. A., *Handbook of Chemistry* (McGraw-Hill Book Co., Inc., New York, 1961), 10th ed., p. 1426.

[6] Honig, R. E., "Vapor pressure data for the more common elements," RCA Rev. **18,** 195–204 (1957).

[7] Nesmeyanov, A. N., Khandamirova, N. E., Vilenskii, V. D., Birin, E. A., and Borisov, A. E., "Effect of oxide films on the rate of evaporation," Russ. J. Phys. Chem. **34,** 681–684 (1960).

[8] Frauenfelder, H., "Radioactive investigation of surface processes," Helv. Phys. Acta **23,** 347–80 (1950).

[9] Devienne, F. M., "Variation of the conductivity factor of molecular jets of antimony and cadmium," J. Phys. Radium **14,** 257–259 (1953).

[10] Knudsen, M., "Die Verdichtung von Metalldämpfen an abgekühlten Körpern," Ann. Physik **50,** 472–489 (1916).

[11] Rapp, R. A., Hirth, J. P., and Pound, G. M., "Condensation coefficients in the growth of cadmium and zinc from the vapor," J. Chem. Phys. **34,** 184–188 (1961).

[12] Holland, L., *Vacuum Deposition of Thin Films* (John Wiley and Sons, Inc., New York, 1956), p. 200.

[13] Sennett, R. S., McLanchlan, F. A., and Scott, G. D., "Electron microscope studies of evaporated cadmium and zinc," Can. J. Phys. **30,** 370–372 (1952).

[14] Bueche, F., "The optical properties of Ni, Co, Fe, Mn, and Cd," J. Opt. Soc. Am. **38,** 806–810 (1948).

[15] Fraser, R. G. J., *Molecular Rays* (The University Press, Cambridge, Mass., 1931).

[16] Palatnik, L. S. and Gladkikh, N. T., "On the microheterogeneous condensation of metals in vacuum," Soviet Phys.—Doklady **6,** 804–806 (March 1962).

[17] Palatnik, L. S. and Gladkikh, N. T., "Zinc and cadmium condensation conditions in the second (lower) cut-off temperature region," Soviet Phys.—Solid State **4,** 307–310 (1962).

[18] Gloria, H. R., personal communication (April 1964).

Titanium, Zirconium, and Yttrium Hydrides as Space Shielding Materials

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The neutron and proton attenuation properties of hydrogen are well established and documented. Water is an ideal neutron shielding material with regard to cost and hydrogen content, but it is not ideal with regard to physical and mechanical properties and containment considerations. Organic hydrogenous materials are subject to large volume changes associated with the liquid-solid phase change, and materials such as polyethylene frequently exhibit excessive distortion and swelling under high-temperature radiation.

Metal hydrides have numerous merits; LiH is an excellent neutron-proton shield material, and its physical chemistry, fabrication, and postfabrication properties have been investigated extensively. Hydrided titanium, zirconium, and yttrium have some physical and mechanical property advantages, which make them superior to LiH for certain applications. Some general information on metal hydrides and other hydrogenous shield materials is presented in Table 1, which confirms the high-temperature advantages of Ti, Y, and Zr hydrides. Titanium and zirconium have hydrogen contents equivalent to or greater than water and most organics, are more effective as gamma shielding materials than water and organics, are metallic in nature, are fabricable, and retain enough strength to be considered as self-supporting structures in simple geometric shapes (e.g., slabs, right circular cylinders, or hexagons) at elevated temperatures (> 500°C). In addition, certain shield configurations, e.g., the spherical shield for a point source, amplify the importance of high N_H values [(H atoms/cm^3) $\times 10^{-22}$], which can result in distinct weight savings. This note reviews recent, previously unpublished data for several metal hydrides of Ti, Y, and Zr as shield materials for space nuclear powerplants. Fabrication techniques and engineering design data are included.

Titanium Hydride

Titanium hydride has the highest useful hydrogen content per unit volume (to 0.16 g H/cm^3) of any known, thermally

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Presented as Preprint 64-362 at the 1st AIAA Annual Meeting, Washington, D. C., June 29–July 2, 1964; revision received November 9, 1964. This paper originated from work sponsored by the Fuels and Materials Development Branch, Atomic Energy Commission, under Contract No. AT(40-1)-2847.

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Table 1 High-temperature hydrogen-containing shield materials

Material	Density, g/cm³	H_2, wt %	$N_H \times 10^{22}$, atoms/cm³	Melting point, °C	Max. useful temp., °C
YH_x($x \approx 2$)[a, b]	4.3 (R.T.)	2.0–2.1	5–5.3	...	1200
ZrH_x($x = 0 \rightarrow 2$)[a, c]	6.5–5.62 (R.T.)	0–2.1	0–6.7	...	800
TiH_x($x = 0 \rightarrow 2$)[a, c]	4.5–3.8 (R.T.)	0–3.9	0–9.0	...	600
CaH_x[d]	1.6–2.0 (R.T.)	0–5.4	0–5.4	(816)	700
LiH[b, e]	0.75–0.78 (R.T.)	~12.6	5.9[f]	683.5	550
...	0.55[f] (700°C)	...	4.1	...	800
Polyethylene (CH_2)n	0.93 (R.T.)	14.	8.02	...	...
Eicosyl benzene (M.W. 350)[e]	0.7[f] (280°C)	12.8	5.1 (280°C)	−40	350[f]
Terphenyl (Santowax R)[e]	0.88[f] (280°C)	6.1	3.6 (280°C)	145	300[f]
Water[e]	1.0[f] (4°C)	11.1	6.7 (0°C)	0	150[f]

[a] Self-supporting.
[b] Normally requires surface protection in air.
[c] Needs no surface protection in air.
[d] Normally requires surface protection at all of the temperatures.
[e] Requires containment and structural support.
[f] Liquid phase.

stable (300°C and above), massive solid material. (This includes all of the known organic hydrocarbons.) This high hydrogen content and its low density (~3.8 g/cm³) make TiH_x attractive for neutron-proton shielding applications.

Prehydride processing

A series of laboratory experiments was conducted on arc-cast buttons of titanium and titanium alloys to determine the amount of metallurgical processing required to refine the normal cast structure and improve the hydriding characteristics in order to allow high N_H (7.0–8.9) massive hydride bodies to be obtained. Hot-forging at 810°–1000°C (30–40% reduction in area) and cold-rolling (20–30% reduction in area) operations were found to give fine grain structures. A hydriding process using these specimens was developed which yielded high N_H (8.0–8.5) crack-free pieces. The hydriding process normally results in grain growth caused by long time exposure at the high temperatures. Certain grain growth inhibitors and special hydriding techniques can minimize grain growth. Proper hydride processing control may employ all or part of the following operations: composition control (grain refining operations), prehydride metallurgical processing, recrystallization, and precise control of the hydride cycle. The latter is more critical as the N_H and size (cross section) of the hydrided body increase. (Complete details cannot be released at this time, however.)

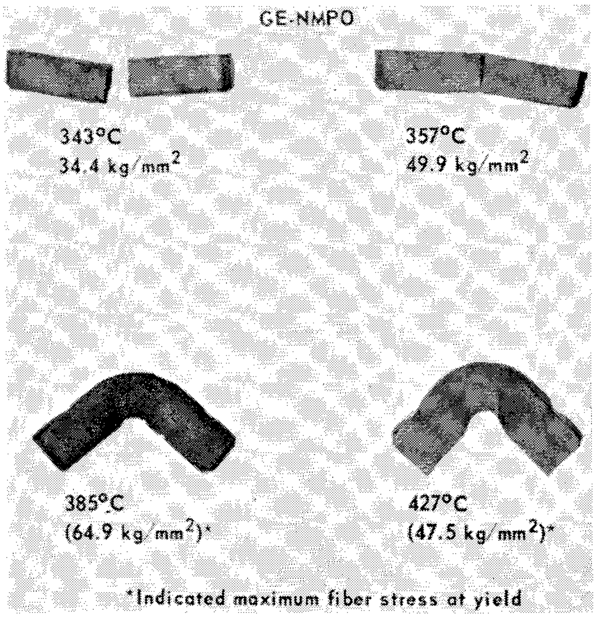

Fig. 1 Modulus-of-rupture specimens of hydrided titanium showing relative degrees of ductility from 343° to 427°C. Note slight deformation before breaking at 357°C (neg. P63-12-7B).

Several small samples of commercially pure, wrought 99.2 Ti-0.1% carbon-0.2% Fe were hot-press-forged (1000°C, 40% reduction) and then hydrided to an N_H of 8.9. This experiment demonstrated that metal hydrides can be fabricated with as much as 30% more hydrogen than water at STP. Large (14.0-kg) Ti ingots (commercial grade), 9-cm thick × 21 cm in diameter, were successfully hydrided to an N_H of 7.2. With the exception of small surface cracks, these specimens were essentially crack-free.

Thermal stability

A section measuring 3 × 9 × 21 cm was cut from one of the 14-kg hydrided slabs and was sawed into blocks of approximately 2 × 2 × 3 cm. These blocks were held at 540°C in static furnace air to determine hydrogen retention and oxidation resistance. They were rapidly removed into a room temperature environment at 100- to 200-hr intervals for weighing, dimensioning, and inspection. Weight gain curves are indicative of a parabolic oxidation rate with little or no induction period. The average total weight gain for 24 specimens after testing for 2500 hr at 540°C was 0.145%. The average parabolic rate constant K derived from the weight change data is 3×10^{-11} g²/cm⁴-min and is very similar to that for unalloyed Ti at this temperature.[1]

Modulus of rupture

Modulus-of-rupture results from specimens machined from the thermal stability samples are presented in Table 2. These tests indicate: 1) outstanding strength and ductility at temperatures above 350°C, and 2) capability of retaining useful engineering properties after prolonged exposure under normal atmospheric operation conditions. The post-test appearance of several samples of this material are shown in Fig. 1.

Hydrogen distribution and retention

The hydrogen distribution through a center section of a 14.0-kg slab was determined. Representative photomicrographs are shown in Fig. 2. Chemical analysis for H was performed on the broken modulus-of-rupture samples, which had been exposed in air for 456, 1072, and 2600 hr at 540°C. No significant hydrogen loss was detected (Table 2). These data indicate the effectiveness of an adherent oxide film on the surfaces of these metal hydrides in preventing hydrogen loss by diffusion.

Linear thermal expansion and heat capacity

The differential (relative to Al_2O_3) and total expansion data for a 0.625-cm-diam × 6-cm-long TiH_x are plotted in Fig. 3.

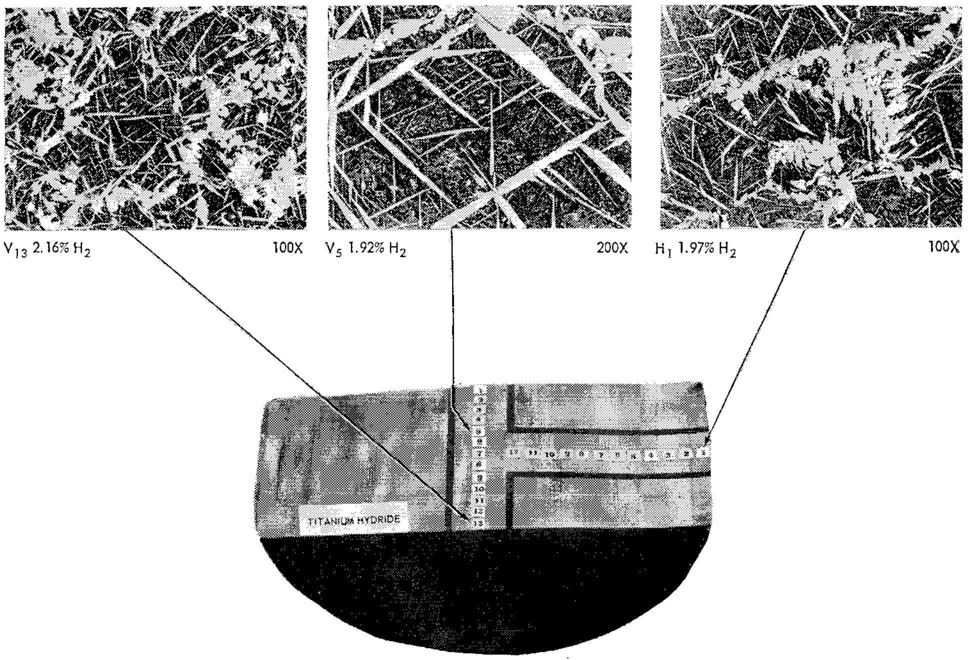

Fig. 2 Cross sections of cylindrical 14-kg titanium hydride specimen. This specimen was completely sound. Note the smooth-cut face and the excellent uniformity of hydrogen concentration.

The data show instantaneous linear expansion coefficients of 5 × 10^{-6} cm/cm-°C at 30°C and 13 × 10^{-6} cm/cm-°C at 500°C. The accuracy of the measuring technique as determined by calibration of the Al_2O_3 standard is ±0.001%. A specific heat curve is presented in the lower part of Fig. 3.

Hydrided Yttrium Alloys

The equilibrium hydrogen pressures of yttrium hydride at high temperatures are significantly lower than those of any other known hydride and offer the promise of at least a 200°C higher allowable operating temperature (see Fig. 4). However, the long-term (>1000 hr) thermal stability, clad stability, and performance characteristics of this material were not previously established. The present study was conducted with a Y-5Cr alloy, flame-sprayed with 0.007-cm chrome, hydrided to an N_H of 5.1–5.3 (~2.0 wt % hydrogen), clad with 0.04 cm of Fe-20Cr-4.51Al (wt %), and gas-autoclave-bonded at 980°C and 5000 psi. The chromium addition increases the mechanical stability of the hydrided yttrium, and the cladding provides oxidation resistance for the hydrided Y-5Cr alloy.

Table 2 Modulus of rupture or maximum fiber stress at yield point of titanium hydride[a]

Test temp., °C	Modulus of rupture (max. fiber stress), kg/mm²	Hr at 540°C	Nature of fracture	Hydrogen analysis after test, wt %
20	44.6	456	Brittle	1.92
343	34.4	1072	Brittle	...
350	36.6	2600	Bend, then break	...
357	49.9	1072	Green stick-type fracture (some evidence of ductility)	
357	40.9	2600	Break	...
357	55.6[b]	2600	None (ductile)	2.01
360	49.2	2600	Bend, then break	1.88
365	41.2[b]	2600	None (ductile)	...
427	47.5[b]	1072	None (ductile)	...
540	21.2[b]	456	None (ductile)	1.91
540	22.1[b]	456	None (ductile)	1.92

[a] 3-point loading (specimen 0.6 × 0.6 × 3.0 cm, 2.3 cm on span); nominal hydrogen content 1.93 ± 0.02%; specimens cut from 14.0-kg ingot; specimens tested after oxidation testing; and $MR = (MC/I) = (3/2) \times (Pl/W(H)^2)$.

[b] Ductile bend obtained in these samples; therefore not a true modulus of rupture (data shown for comparative purposes only).

Thermal stability

A series of specimens was tested in air for periods of 10,000–11,000 hr at temperatures of 950°–1040°C. The specimens were subjected to numerous, relatively severe thermal cycles during the course of the test. No core cracks were apparent in the first 5000-hr test period. Limited core cracking was observed in all of the specimens after 10,000 hr and is attributed in part to the severity of the thermal cycles. The cracking had no measurable effect on the dimensional stability of the specimens. Clad blisters were encountered with 3 of 4 specimens at 1040°C. Chemical analysis shows that the clad blisters had little effect on hydrogen retention. All of the test specimens retained substantially 100% of the original hydrogen content.

Radiation stability

Three high-temperature, air-cooled reactor tests have been completed. The longest was in air at 900°C for 5100 hr for an estimated thermal neutron exposure of 1.3 × 10^{20} nvt. The early phases of a hot laboratory examination have been completed, and no evidence of distortion, dimensional or weight instability, or clad blisters was found. The clad-to-core bond of all of the specimens was intact, as determined by ultrasonic inspection, except for one end of one specimen. No core cracking was noted by x rays taken through lead with a Van de Graaff electrostatic accelerator. However, resolution, as determined with cracked standards, was marginal. Tests at higher exposures are in progress. The excellent dimensional stability characteristics coupled with the hydrogen retention properties of this material over a 10,000 hr period represent the first successful long-time operating life demonstration of metal hydrides at temperatures in the 1000°C range.

Zirconium Hydride

A considerable amount of research and development on this material has been conducted as part of the General Dynamics Training Research Isotope (-production) General Atomic (TRIGA) reactor program,[3] the Aircraft Nuclear Propulsion (ANP) program,[4] and more recently as part of the Atomic Energy Commission Systems for Nuclear Auxiliary

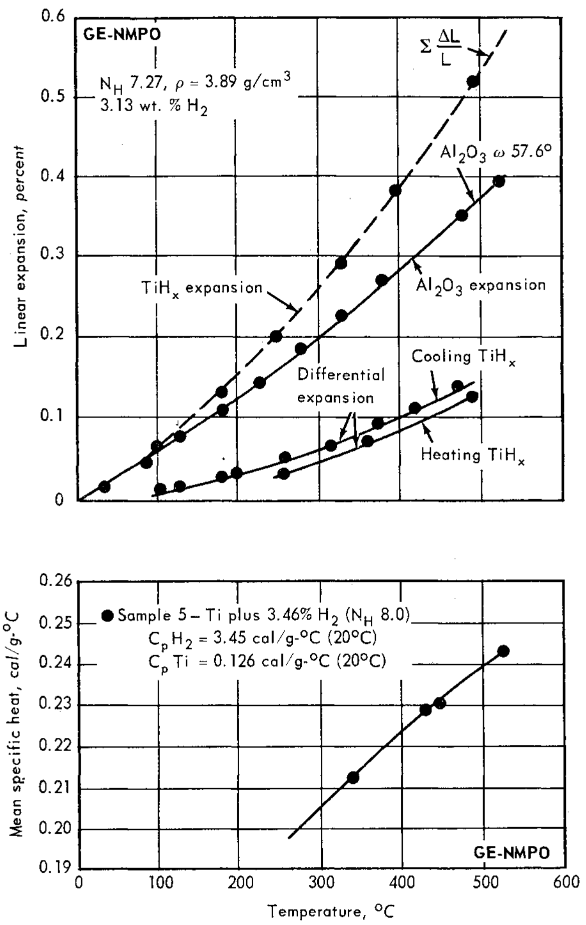

Fig. 3 Linear thermal expansion and specific heat of hydrided titanium.

Power (AEC SNAP) 2, 8, and 10A reactor programs. Among the latest publications is one by Beck.[5]

Table 1 shows that the density of this material (5.6 g/cm^3) is the highest of the metal hydride materials listed and that it has an 800°C temperature limitation. By appropriate additions and the subsequent metallurgical working of the structure, a fine prehydrided grain size can be produced. The fine grain size combined with the proper hydriding cycle will produce sound, small-grained Zr bodies of N_H 7.0, although N_H 6.0 bodies are more practical for engineering applications.

Thermal stability

Unclad samples of ZrH_x (N_H 6.0) retained their dimensional stability and hydrogen (>95%) after >1000 hr in Mach 0.2 air at 650°C.

Seven samples (~1.5 cm in diameter × 2.0 cm in length–N_H 6.6) were included in the 540°C thermal stability experiment on titanium hydride previously described. The weight gain rates and the average parabolic oxidation rate constant for the 2522-hr test period are approximately three and ten times greater, respectively, than the values for hydrided titanium and are very similar to the values for unalloyed zirconium at this temperature.[2]

Concluding Remarks

Of the three metal hydrides discussed in this paper, acceptance of zirconium is by far the most advanced relative to nuclear applications. As early as the late 1950's, complete reactor tests were run, employing hydrided zirconium[4] and hydrided zirconium-uranium.[3] Since these tests, SNAP 2, 8, and 10A programs have announced the use of a hydrided Zr-U alloy as a reactor fuel element. At present there are no known reactors in operation or under construction employing yttrium or titanium hydride. However, it is believed that, as more engineering and physical property data on massive metal hydrides are obtained, these materials will be accepted by design engineers for application as nuclear shield and moderator materials in advanced powerplants.[2]

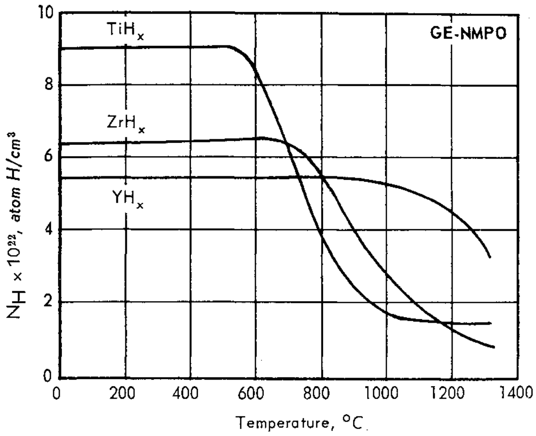

Fig. 4 Content of absorbed hydrogen in metallic zirconium, titanium, and yttrium in equilibrium with 15 psia of hydrogen at various temperatures.

References

[1] McQuillan, A. D. and McQuillan, M. K., *Titanium* (Butterworths Scientific Publications Ltd., London, 1956), p. 418.

[2] Lustman, B. and Kerze, F., *Metallurgy of Zirconium* (McGraw-Hill Book Co., Inc., New York, 1955), pp. 584, 589.

[3] Wallace, W. P., Simnad, M. T., and Turovlin, B., "Fabrication and properties of uranium-zirconium hydride fuel elements for Triga reactors," Nucl. Met. **5**, 49 (1958).

[4] Linn, F. C., "Heat transfer reactor experiment no. 3," APEX 906, General Electric Co. GE-NMPO, p. 84 (June 1962).

[5] Beck, R. L., "Thermophysical properties of zirconium hydride," Am. Soc. Metals Trans. Quart. **55**, 556 (1962).

Atmospheric Density Variations with Latitude and Season

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VARIATIONS in atmospheric density are of increasing importance in the design of aircraft, missiles, and space boosters. This note presents a diagram and a table, which relate average density and density variation for January and July at latitudes over the Northern Hemisphere to the

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Received October 6, 1964; revision received November 2, 1964. Most of the compilation of material in this report was done as part of the Polaris program, under Contract No. NOW 63-0050. The interest of Carl E. Grant, Earl A. Harris, and Charles A. Louis, of the Re-Entry Systems Office, Lockheed Missile and Space Company, is appreciated. Helpful comments and advanced copies of papers and tabulations were provided by R. S. Quiroz of the Air Weather Service Climatic Center, Allen E. Cole and Norman Sissenwine of the Air Force Cambridge Research Laboratories, and Oskar Essenwanger of the Army Missile Command.

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