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THERMOPHYSICAL PROPERTY DATA -
WHO NEEDS THEM?

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THERMOPHYSICAL PROPERTY DATA - WHO NEEDS THEM?

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ABSTRACT

Specific examples are cited herein to illustrate the universal needs and demands for thermophysical property data. Applications of the principle of similarity in fluid mechanics and heat transfer and extensions of the principle to fluid mixtures are discussed. It becomes quite clear that no matter how eloquent theories (or experiments) in fluid mechanics or heat transfer are, results of their application can be no more accurate than the thermophysical properties required to transform these theories into practice - or in the case of an experiment, to reduce the data. Present day projects take place on such a scale that the need for international standards groups and mutual cooperation is evident.

INTRODUCTION

Who needs thermophysical property data? Ask any company manager with a deadline to meet and no way to proceed until thermophysical properties data become available. When this happens, management will permit its engineers to go to extremes to acquire the needed information quickly. The number of times this occurs makes it clear that data, or methods for their prediction, are needed on the shelf in anticipation of need. Furthermore, present day applications take place on such a scale with materials and energy so costly that it is no longer acceptable to simply use "just any old properties" to describe a fluid, but rather more faithful reproduction of the fluid properties at the desired conditions is now required.

The need for accurate, consistent, universally acceptable thermophysical property data and representative computer programs are requirements of industry, government and universities alike throughout the world in advance of specific applications.

In a treatise on thermodynamics, Zeleznik (ref. 1) wrote that thermodynamics perhaps more than any other physical theory pervades the fabric of physical science; as a corollary, I would suggest that thermophysical properties which heuristically relate the physical nature of matter, pervade our daily lives. Applied and basic disciplines alike in such areas as engineering, medicine, physics, chem-

istry, biology to cite only a few, rely heavily on adequate thermophysical properties bases. Yet thermophysical property data usually arouse little interest or concern on the part of the engineers and management until such information is needed and found to be unavailable; an even more detrimental situation arises when management thinks that the data are available, but the information - which has not been critically evaluated - is wrong. Somehow it is always assumed that such properties either will be available or predictable when needed. Due to this prevailing philosophy, many of the advances in thermophysical property measurements, techniques of representation and computer programs for rapid production of this information are developed in response to an urgent request. The Aerospace Industry found a need for new property data sets with nearly every major project and more recently a similar need has arisen in the Natural Gas Industry. On some rare occasions, and through the diligent efforts of a very few, new property sets have been available when required. Organizations as NBS with CODATA, NASA, IUPAC at Imperial College, TPRC at Purdue, AGA in conjunction with the AIChE, the ASME and the Electric Power Utility Industry have sponsored such programs.

How can engineers and scientists benefit from systematic studies of accurate thermophysical property information? Subsequent sections will (1) discuss some project needs related to the development of data bases using (i) para-hydrogen as an example of a well defined program and data set, and (ii) RP1 (JP etc.) as examples of sets where effort is required; (2) cite a few property related projects in the areas of (i) fluid mechanics, using property similarity principles to correlate data, and (ii) metallic, ceramic and plastic materials; and (3) look into the van der Waals one fluid model to relate thermodynamic and transport properties of some pure component fluids and mixtures, and indicate some moves toward generalization with the application of corresponding states.

SOME DEVELOPMENTS IN THERMOPHYSICAL PROPERTIES

INSTIGATED BY THE AEROSPACE INDUSTRY

The following section will (1) illustrate some projects and related development of one of the world's finest data bases, para-hydrogen, as a specific example, and (2) indicate some current activities including multicomponent mixtures and cite some work with RP (kerosene).

Some Developments Related to Para-Hydrogen

Even before this country had a space program, mission analyses codes clearly demonstrated the advantages of propellants with high specific impulse. From the combustion analysis program of Huff, Gordon and

Morrell (ref. 2), two of the most promising propellant combinations, H_2-O_2 and H_2-F_2 , were identified. Early literature searches for properties of these propellants (H_2 (ref. 3)), (O_2 (ref. 4)), (F_2 (ref. 5)) revealed a dearth of information. The need for such data became urgent.

With hydrogen designated as the engine coolant and NACA transformed into NASA, a substantial effort was launched with NBS-Boulder to determine the properties of, among others, hydrogen, oxygen, and fluorine. We will look specifically at the development of the hydrogen data base where the classic work of Woolley, Scott, and Brickwidde (ref. 6) was our initial guide, the limitations of which had become transparent.

One of the early problems we faced was the long term storage of liquefied hydrogen. Over 75 percent of the liquefied hydrogen just vanished after a few days, clearly an intolerable situation for any space mission. While the tank insulations were poor, they were not that poor; returning to the thermophysical properties, we found that at ambient temperatures, hydrogen was not really hydrogen at all, but a mixture of 75 percent ortho and 25 percent para hydrogen. Further the equilibrium composition varied with temperature, with substantial property changes, notably in enthalpy, specific heat and thermal conductivity, and at 20 K it was 99 percent para (see figs. 1 and 2). Thus the ortho-hydrogen would simply boil-off leaving the smaller quantity of para-hydrogen. Ortho-para conversion units were researched and installed (ref. 7) and long term storage of hydrogen became a reality, but only after the additional development of the thermophysical properties of insulations and superinsulations over a period of years (ref. 8) (fig. 3).

Concurrent with the thermophysical properties work at NBS-Boulder, heat transfer and fluid dynamics studies were undertaken, with results that appeared completely contradictory to known theories and experimental results. For example, in fully developed turbulent flows, the wall temperature increases in a monotone manner over the length of the heated tube; however, with hydrogen, the profile increased to a maximum, then decreased in a monotone manner, sometimes to a minimum followed by a monotone rise (refs. 9 to 11) (fig. 4). The need for thermophysical properties data of para-hydrogen and for understanding catalytic conversion then became even more acute. Goodwin's data compilation (ref. 12) and Goodwin's simplified equation of state (ref. 13) along with a computer program by Harry (ref. 14) and the generalized property charts of Thodos (refs. 15 and 16) proved adequate to resolve a few of the early heat transfer and fluid dynamics problems. But, as in any field where accurate, consistent property data are lacking, we found several different researchers, manufacturers, and the nuclear people in disagreement as to which computer codes and properties to use. On many occasions, we would sit around a table, peek at our computer out-

put and ask, "What number did you get?" as if there existed some magical set of proprietary properties to explain the differences in the calculated results for that particular application. Recognizing this problem, NASA launched an early effort toward unification. For example, early computer codes by Roder (ref. 17) and Farmer (ref. 18) though fast, lacked uniformity, and suffered from the lack of adequate quantitative data.

With the classical data of Goodwin (ref. 19) and the key Strobbridge-Stewart extension of the BWR (Benedict-Webb-Rubin) technique (ref. 20), Roder and Goodwin (ref. 21) produced the first accurate representation of the PVT surface for para-hydrogen to 300 K. With this equation of state, based on accurate, consistent, and extensive data and the measurement of transport properties by Diller and Roder (refs. 22 and 23), the entire industry began to settle down. Experimental heat transfer and fluid dynamics results could now be placed in proper perspective even though many could not yet be correlated - but now the analysts, experimentalists, and engineers stopped trying to assess the impact of differences between poor properties and the fact that our understanding of the dynamics of the application was totally inadequate (ref. 24).

The loss of many man-years of effort, duplication, and unnecessary proof of concept projects could perhaps have been avoided if adequate thermophysical property information had been available when needed. Since that time, NASA-Lewis has supported an ongoing effort at NBS and an effort at the University of Maryland in order to minimize the recurrence of such events; to date we have been moderately successful in assessing the state of the art in the areas of high pressures, fluid mixtures, transport properties, and the near critical region anomalies. We have not yet been successful in the attempt at unification, but it is an area which must be confronted and overcome both on a local and international scale. I might add that the hydrogen and oxygen data bases represent the most extensive, accurate, and consistent sets in existence.

Current efforts are toward exploring (1) the thermophysical properties of hydrogen and oxygen to 100 MPa (~15 000 psia) for future mission applications, (2) the basic concepts needed for predicting the thermophysical properties of fluid mixtures, and (3) the nature of thermal conductivity and viscosity including the anomalous region near the thermodynamic critical point.

Why are we still interested in property data? It is obvious that high pressure applications will increase payload and decrease the costs while the spinoffs to landbased power systems appear substantial in terms of increased efficiency. As for mixtures, if we could determine how to predict the PVT_x properties of a simple mixture of an organic-inorganic fluid, with substantially different critical points and vapor

pressure curves, from the extensive, yet smaller data sets associated with the pure components we could project a generalized method for predicting the PVT_x surface of any simple mixture. The near critical region provides a key to understanding thermophysical property surfaces, and while thermodynamics of the critical region have received much attention, the transport properties have not. Sengers et al. (ref. 25) have been able to assess the anomalous thermal conductivity in the critical region for a number of fluids. The anomalous thermal conductivity is strong and can be related to the divergence of compressibility. Recently Basu and Sengers (ref. 26) have formulated an equation which predicts a very weak anomaly in viscosity. Efforts in these areas are making remarkable advances.

Some Developments Related to RP1

Concurrent with the effort to develop high performance propellants, a large effort was launched to improve the propellant kerosene or RP1 as it was designated. In this case, a sound, systematic program as completed for hydrogen, was not undertaken, and unfortunately I must report that little has been added to our fundamental understanding since that time. Neither a uniform PVT computer code or data base is available to high pressures and transport properties are of questionable value. The field could benefit from a mixtures program and pure component hydrocarbon data bases. Even though the data base is of questionable value, we are currently seeking to understand the effects of coking both in the fluid and combustion gas states.

Early RP rocket engines were not adequately cooled, theoretically, to prevent burnout of the coolant passages. A careful inspection revealed that a thin layer of coke formed on the thruster side of the coolant passage which protected it against the extreme temperatures and thermal fluxes. Coking has been studied by the petrol-producers (e.g., refs. 27 to 29) and found to be quite sensitive to oxygen and sulfur compounds in the fuel. These compounds also markedly changed the heated tube data of (ref. 30), where coking was found. No systematic effort has been undertaken to determine the nature or limitations of coking in coolant passages (thermostability), even though hydrocarbons are the mainstay of our transportation industry in the form of fuels and lubricants. The problem is compounded when shale derived fuels are considered.

We should assess the availability of data for RP, incorporating results of the mixtures and pure component data base programs, and provide the industry with a representation of the PVT and transport properties data to 100 MPa (15 000 psia). We should then investigate the limits of heat transfer to 80 MN/m^2 ($50 \text{ Btu/in.}^2\text{-sec}$), with wall temperatures to perhaps as high as 2000 K (3600° R).

While these examples serve to illustrate the developments related

to aerospace, the results have been extensively used by industry in general. Data bases for oxygen (ref. 31), fluorine (ref. 32), FLOX (ref. 33), nitrogen (ref. 34), helium (ref. 35), and selected hydrocarbons (refs. 36 to 38) have been established for NASA at NBS Boulder.

Some Other Property Bases and Related Applications

In this section, we cite some other data bases of interest to NASA and industry while citing a few applications. Some organizations where such data can be found are covered in the papers of Neil A. Olein and Peter E. Liley, presented at this meeting.

NASA has or has had properties related projects in fields other than fluid properties. They are or have been establishing data bases in areas such as superconductors, slushes and gels, ultra-high pressures, combustion, and materials as metals, refractories and plastics. We will only briefly mention some of these fields.

In the area of superconducting magnet technology and low temperature physics NASA has produced several magnets, developments in magnet wire and coatings such as formvar. Currently there is some interest in MHD.

In the area of ultra high pressures and transition points, the potential of an ambient temperature superconductor initiated a study on the possibility of compressing hydrogen to the metallic state which may require a pressure of 4 Mbars. Many semiconductor to metal transitions have also been studied (refs. 39 and 40). More recently, the production of metallic neon has been achieved (ref. 41). A spinoff of this work is the gadolinium heat pump (refs. 42 to 44) which has the potential of achieving near Carnot efficiency.

Free radical fuels offer very high impulse potentials and have stimulated work in producing atomic hydrogen propellant; however, free radicals are difficult to stabilize and produce, and little effort is currently given to it.

Slush propellants have increased bulk densities, increased coolant capacity and reduced boiloff losses. A slush is readily produced in small quantities through evacuation of the container, but NBS produced p-H₂ slush using a novel refrigeration scheme (ref. 45). NBS has also determined the properties of some slushes.

Gels have been considered for thermal stability of fuels and fire inhibitors, and in some cases, for providing increased performance (ref. 46). The problem with gels and slushes is in transferring them; they tend to clog up pumps and adhere to the containers, and for RP mixtures, the tendency is toward formation of waxes.

In the area of plastics, the need for high temperature (to 600° F) polymer matrix composites lead Serafini (refs. 47 and 48) to develop PMR (in situ polymerization of monomer reactants) polyimides. In the PMR approach, the reinforcing fibers are impregnated with a solution containing a mixture of monomers dissolved in a low boiling point alkyl alcohol solvent. The monomers are essentially unreactive at room temperature, but react in situ at elevated temperatures to form a thermo-oxidatively stable polyimide matrix. These highly processable addition-type polyimides can be processed by either compression or autoclave molding techniques. PMR offers superior processing versatility, high temperature properties, and find application in advanced engine and airframe concepts. Refractories are used in many engine components where complexity precludes adequate cooling for conventional materials (ref. 49). For example, a two-layer system with a bond coating of nickel-chromium-aluminum-yttrium (Ni-16Cr-6Al-0.6Y) and a ceramic coating of yttria-stabilized zirconia (ZrO₂-12Y₂O₃) has been tested in several applications. Similar refractory coatings are being applied to turbine blades and rubbing surfaces (refs. 50 to 52), to reduce gas path sealing losses and increase engine efficiency (ref. 53).

SOME MOVES TOWARD BETTER PROPERTY REPRESENTATION

In this section we will (1) discuss the application of the principle of similarity to thermophysical properties and fluid mechanics - a move toward generalized transforms, and (2) suggest a merger of the extended corresponding states concepts with the thermophysical properties function of Zeleznik (ref. 54) - a move toward generalized thermophysical property representations.

Toward a Generalized Transform

The strongest impetus toward generalization is the insight gained by comparing experimental results for fluids α and β ; the implication that experimental data taken with a cryogen are applicable to, say water, and vice versa, is of significant value.

Based on the engineering needs for properties in heat transfer and fluid flow computations and sparked by Goodwin's enthusiasm, we advocated earlier that simple fluids could be represented accurately, consistently and extensively by a form similar equation, but with different coefficients. Based on Bender's equation for five fluids (ref. 55), we were able to produce a uniform properties computer code GASP for eight fluids (currently 12) and subsequently the code WASP for water. Armed with these codes and the principle of similarity we began to explore the utility of this approach. We found that we could (ref. 56)

(1) Extend fluid ranges in PVT and determine the character of the surface derivatives

(2) Produce reduced Joule-Thomson coefficients and the reduced Joule-Thomson locus for many fluids

(3) Produce anomalous thermal conductivity representations for most of these fluids

(4) Correlate two-phase choked flow data and predict these results analytically

(5) Group experimental heat transfer results over a relative wide range of fluid conditions, and

(6) Produce a wide variety of normalized property parameters for several fluids which could then be represented in reduced form on a single chart.

Currently order for many of these fluid property related projects and concepts can now be found using the work of NBS and Mollerup (refs. 57 to 59), where the properties of two pure substances can be related by the equations:*

$$Z_m[V, T, x] = Z_0[V/h, T/f]$$

$$G_m[V, T, x] = fG_0[V/h, T/f] + RT \left(\sum_{\alpha} x_{\alpha} \ln x_{\alpha} - \ln h \right)$$

where 0 denotes the reference fluid and the properties are termed configurational because they have similar molecular potentials, e.g., both are adequately described by two parameter potentials. For $x = 1$, the equations relate the properties of two pure components while for $x \neq 1$ the relation expresses the properties of a multicomponent mixture. The functions f and h are dependent on the ratio of the critical temperature and volume, and for pure components,

$$f = \left(\frac{T_{\alpha}^c}{T_0^c} \right) \theta \quad h = \left(\frac{V_{\alpha}^c}{V_0^c} \right) \phi$$

the parameters θ and ϕ being weakly dependent functions of temperature and volume (see Leach et al. refs. 60 and 61).

For mixtures, we need to account for the energy of mixing, introduce some "mixing rules," and redefine f and h as

$$h = \sum_{\alpha} \sum_{\beta} x_{\alpha} x_{\beta} h_{\alpha\beta}$$

*The so-called van der Waals or one fluid model.

$$fh = \sum_{\alpha} \sum_{\beta} x_{\alpha} x_{\beta} f_{\alpha\beta} h_{\alpha\beta}$$

where

$$f_{\alpha\beta} = \xi_{\alpha\beta} (f_{\alpha\alpha} \cdot f_{\beta\beta})^{1/2}$$

$$h_{\alpha\beta} = \phi_{\alpha\beta} \left(\frac{h_{\alpha\alpha}^{1/3} + h_{\beta\beta}^{1/3}}{2} \right)^3$$

and where $\xi_{\alpha\beta}$ and $\phi_{\alpha\beta}$ are the binary interaction parameters.

Hanley et al. (refs. 62 and 63) applied these configuration functions and proposed that thermal conductivity and viscosity of mixtures could be related in a similar manner. For pure components

$$\lambda[\rho, T] = \lambda_0[\rho_h, T/f] F_{\lambda}$$

$$\eta[\rho, T] = \eta_0[\rho_h, T/f] F_{\eta}$$

$$F_{\lambda} = \left(\frac{m_0}{m_{\alpha}} \right)^{1/2} / h^{2/3} \quad F_{\eta} = \left(\frac{m_{\alpha}}{m_0} \right) F_{\lambda}$$

For mixtures, the functions f and h are as defined previously and F_{λ} , F_{η} are redefined using the Enskog theories.

The use of configurational functions f and h along with an accurate reference equation of state greatly enhances the users capability to accurately relate thermophysical properties. Its greatest utility comes when a dearth of information exists for pure component α , or a multicomponent mixture, and in gaining mechanistic insights when analyzing the data from transport processes with different fluids, or mixtures. It does, however, have the disadvantage of being more complex, requires a very accurate pure component representation, similar molecular potentials and then one needs to determine the configuration factors and, for the case of mixtures, the interaction parameters. While fluid mixtures represent a very difficult problem, some progress is being made. Mollerup has determined some interaction parameters of interest for LNG and has written a computer program to predict properties of a few simple fluid mixtures whose configurational properties are closely associated with methane (the major constituent of LNG).

In reference 30, it was noted that since the conservation equations and the transport relations for heat, momentum, and diffusion are tensors, and the coefficients of proportionality ($\lambda, \eta, \mathcal{D}$) can be transformed nearly independent of the fluid - it was proposed that the conservation equations can be coordinate and fluid invariant. In support of such a proposal, several examples of applications where the data sets for different geometries and fluids could be related through the use of similarity were cited in reference 56, including

(1) Two phase choked flows could be grouped for several geometries and multiple fluids (refs. 64 to 66), free jet flows through tubes with Borda and sharp edge inlets (refs. 67 and 68) circular, square, rectangular, and triangular inlet geometries (ref. 69) and for choked flows of gases a general relation was developed for tubes, annular and tapered convergent annular flows (ref. 70).

(2) Heat transfer data for developed turbulent flows through tubes, from several investigators, could be grouped for multiple gases, liquids and near critical fluids using volumetric expansion parameters and a density fluctuations model (ref. 71).

(3) Application of the generalized transformation to calculating two-phase-choked flows, using the various reference fluids methane, nitrogen, and oxygen, and compared to the experimental data of reference 66 in figures 5 and 6. The theoretical calculations are for the most part, in good agreement independent of the working fluid and the reference fluid used to calculate the results.

The point here is that many insights have been gained through analysis of the properties and application of the similarity principle. Even more insights could be gained if a uniform method of representing properties were available.

Other Thermodynamic Properties Representations

Zeleznik (ref. 54) has elegantly proposed a linearized relation to represent thermodynamic data which conforms to all the principles of thermodynamics (see also refs. 1 and 72). The relation is

$$G = \sum_{\ell=1}^N \phi^{\ell} \sum_{i=1}^N \sum_{j=1}^N \left(\mu_{ij}^{\ell} + v_{ij}^{\ell} \ln x_j \right) x_i x_j$$

G is the thermodynamic chemical potential, x_i are the mole fractions, ϕ^{ℓ} are the elementary symmetric functions, μ_{ij}^{ℓ} the symmetric contribution to G and the $v_{ij}^{\ell} \ln x_j$ introduces asymmetry.

In Zeleznik's space, points represent the pure components, edges the binary interactions and so on to higher order spaces. Although the theory is quite complex, currently requiring very large accurate data sets and accurate numerical representation procedures, the range and potential of this formulation are documented in reference 72 where data for mixtures as ethanol and n-heptane and ionic solutions such as NaCl + H₂O are accurately represented.

As a logical next step in this approach, it would appear that one should (1) combine the work of Zeleznik, Mollerup, and NBS, (2) attempt to integrate the principles of fluid similarity, and (3) return to mixtures of simple fluids.

In the very restricted framework of fluids whose properties are configurational with methane, we can:

(1) Usually predict pure component properties which represent end points in Zeleznik's space whether or not such properties in fact exist.

(2) Often predict properties for binary mixtures which represent the edges or lines joining the points in Zeleznik's space, again whether or not in fact such properties exist.

Once the points and edges of such a space are generated (requiring a lot of computation), one can then use Zeleznik's representation to give properties on and within the surface.

Applying these concepts, the work of Zeleznik (refs. 54 and 72) can probably be extended from a method of representing thermodynamic data to a technique for predicting thermophysical properties. But, such a technique would not be as accurate as a formal representation of an extensive data set, is very computational time intensive, and one would probably seek a more simplistic resolution of the problem.

SUMMARY

In summary, we have looked at some problems and development of fluid data bases along with other data bases and some related applications; discussed mechanistic (flow modeling) insights gained from application of the principle of similarity to the conservation and thermophysical property equations; discussed moves toward generalization of thermophysical property representations; and suggested a technique to extend mixture methods.

We have also cited the need for a continuous effort to produce accurate, consistent, extensive thermophysical property data bases which are placed in a form readily accessible to the user and acceptable at the international level.

It should follow that maintenance of an ongoing program in thermophysical properties and international standardization are vital not only to our aerospace and related industrial growth, but our scientific growth as well.

It would seem that too often we operate on the principle that the squeaky wheel gets the grease, but I think that a well lubricated, properly maintained machine is the most efficient in terms of cost, maintenance, down time, longevity, and conservation of basic resources. So who needs properties? - we all do! - and we must continue to support the accurate consistent determination of thermophysical properties.

SYMBOLS

| | |
|--------------------------|--|
| D | diffusion coefficient |
| F_λ | scale function for thermal conductivity |
| F_η | scale function for viscosity |
| f | scale function, fluid α to reference fluid |
| G | gibbs free energy |
| G_R | reduced mass flow rate, $G_R = \rho u / G^*$ |
| G^* | mass flow normalizing parameter, $G^* = \sqrt{\frac{P_c \rho_c}{Z_c}}$ |
| h | scale function, fluid α to reference fluid |
| m | molecular weight |
| P | pressure |
| P_R | reduced pressure, P/P_c |
| R | gas constant |
| T | temperature |
| T_R | reduced temperature, T/T_c |
| u | velocity |
| V | specific volume |
| x_α | mole fraction |
| Z | compressibility factor, PV/RT |
| η | viscosity |
| θ | shape factor |
| λ | thermal conductivity |
| μ | symmetric function |
| ν | asymmetric function |
| $\epsilon_{\alpha\beta}$ | binary interaction parameter |
| ρ | density |

- ϕ elementary asymmetric function
- ϕ shape factor
- $\phi_{\alpha\beta}$ binary interaction parameter

Subscripts:

- c critical
- m mixture
- 0 reference fluid
- α computer fluid
- $\alpha\beta$ binary interaction
- $\alpha\alpha, \beta\beta$ pure component interaction

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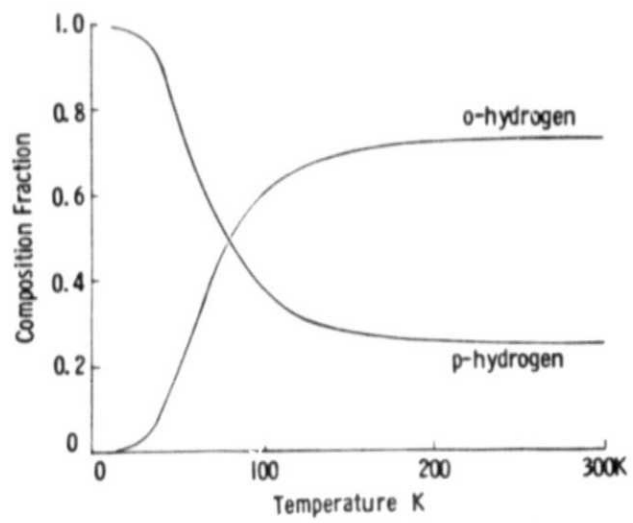


Figure 1. - Composition dependence of hydrogen.

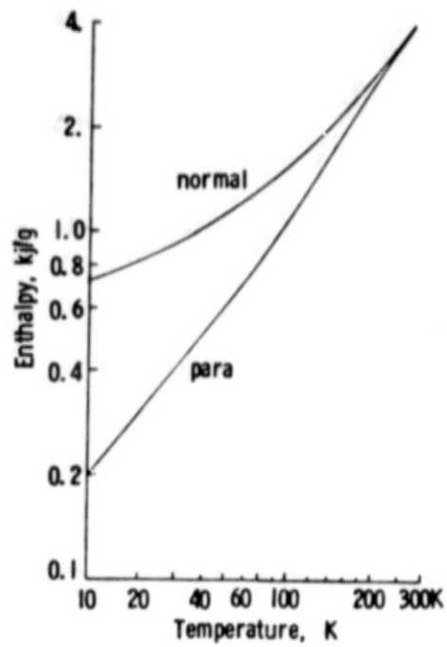


Figure 2a. - Enthalpy of normal and para hydrogen at one atmosphere.

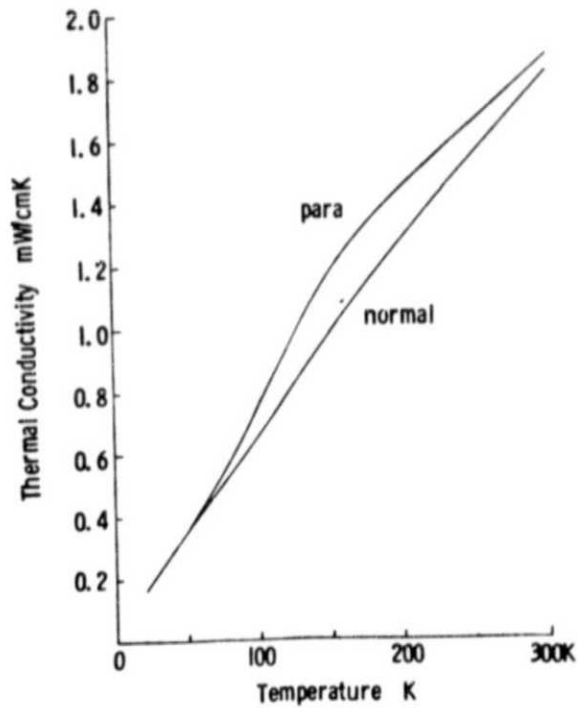


Figure 2b. - Thermal Conductivity of Gaseous Hydrogen at One Atmosphere.

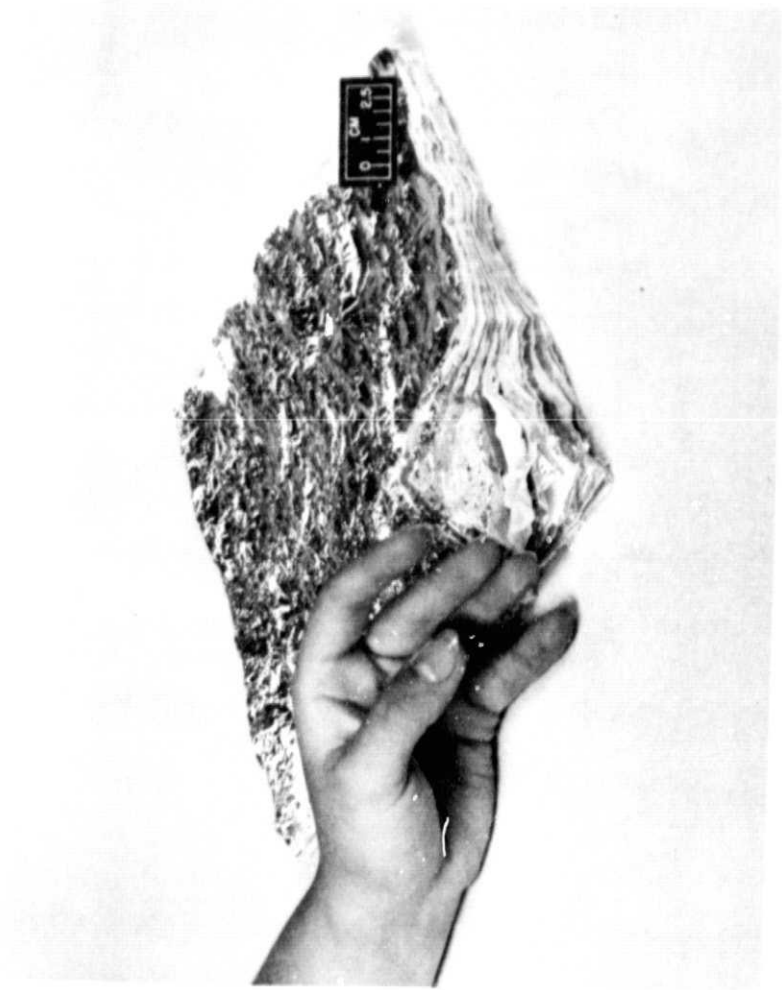


Figure 3. - High-efficiency insulation (MLI)

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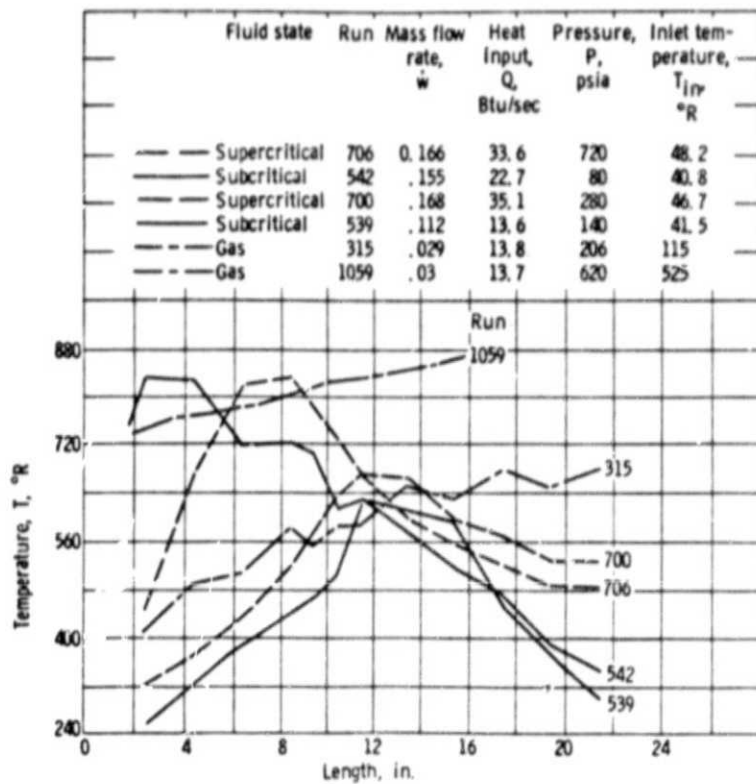


Figure 4. Surface temperature distribution with axial position for near-critical and gaseous hydrogen. Tube inside diameter, 0.335 inch.

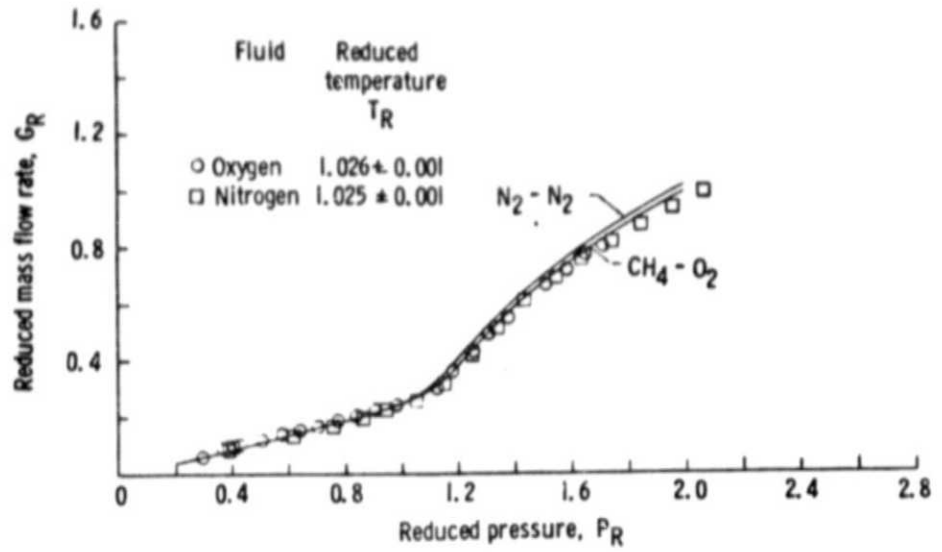


Figure 5. - Choked flow rate for reduced temperature of ≈ 1.025 .

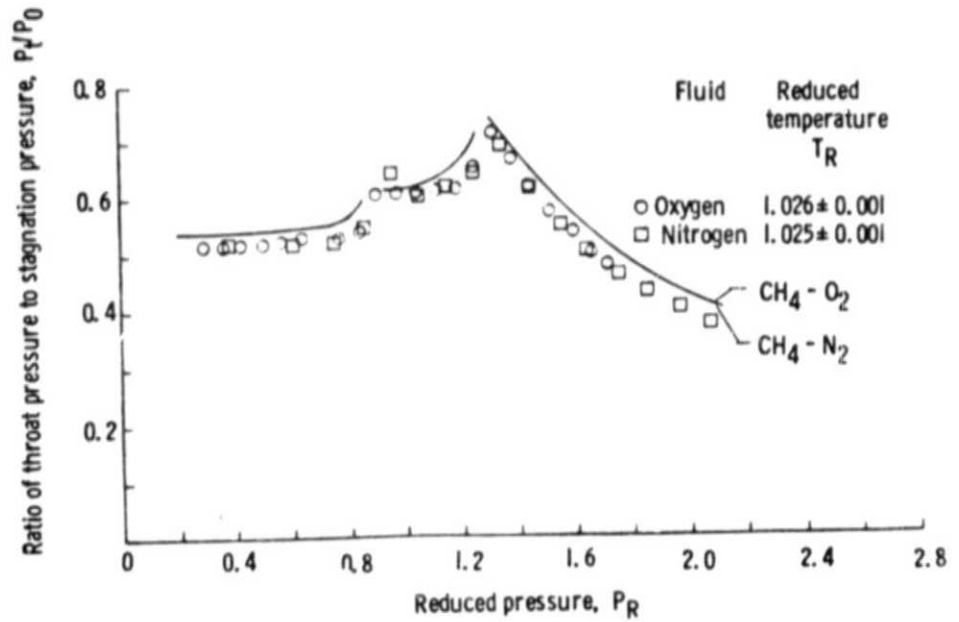


Figure 6. - Pressure ratio for reduced temperature of ≈ 1.025 .