

ionic radius. They found that the Sc_2S_3 type structure, which they labeled ζ , was stable for ratios above 2.10 and the Th_3P_4 structure existed for ratios below 1.95, with a transition structure in the ratio range 1.95 to 2.10. The Th_3P_4 structure is stable for the sulfides of La to Gd and for the selenides of La to Sm and the Sc_2S_3 structure is stable for Sc_2S_3 and for the selenides of Y, Er, Yb, Lu, and Sc. These limits of stability, based on radius ratios, are calculated using the ionic radii of 1.82 and 1.93 Å. for sulfur and selenium, respectively. This analysis will not hold for the tellurides based on the Te^{-2} ionic radius of 2.12 Å., however a corrected radius of 2.03 Å. for the ion will fit the above conclusions. Thus the stability of the structure to be expected in the 2:3 compositions is strongly dependent on the ratio of rare earth to metalloid ionic radius and appears to have distinct limits.

The Th_3P_4 structure is ordered with the 12 rare earth and 16 metalloid crystallographic sites filled at the 3:4 composition and becomes progressively more disordered toward the 2:3 composition as the rare earth concentration decreases. The disorder in the Th_3P_4 structure is a result of the statistical occupancy of the 12 rare earth sites by vacancies. The limits of stability of the structure are due to the ionic character of the 2:3 composition and the complete filling of all the rare earth sites at the 3:4 composition. The basic skeleton of the Th_3P_4 structure is the tellurium network, as the rare

earth sites are statistically variable with the composition. The erbium solid solution structure, however, has as its basic skeleton the 2:3 composition of the erbium and metalloids. Richer erbium concentrations are obtained by the statistical filling of vacant sites which are crystallographically different from the erbium sites of the 2:3 composition. We believe that the limits of solubility observed for solid solutions having the Sc_2S_3 -type structure are due to slight distortions of the idealized framework.⁷ The strong X-ray diffraction lines obtained from the solid solution compositions in this system are sharp even in the back-reflection region and can be indexed unequivocally on a cubic subcell. It is therefore possible to add erbium statistically to the 16 vacancies of the 2:3 skeleton without distorting it, and the structure is stable over the composition Er_2Te_3 to the composition ErTe .

It is reported that Dy_2Te_3 and Y_2Te_3 have the Sc_2S_3 structure and an undistorted cubic subcell.⁵ From the above discussion it is therefore expected that these systems should also exhibit a solid solution range from the 2:3 to 1:1 compositions. The tellurides Dy, Y, and Er have radius ratios of 2.28 or larger. The Sc_2S_3 structure and the extended solid solubility range are also expected for the tellurides of Yb, Lu, and Sc, as well as for the Sc-Se system, because their radius ratios also exceed 2.28.

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Systems of Lithium Hydride with Alkaline Earth and Rare Earth Hydrides

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Solid-liquid equilibrium has been investigated by thermal analysis of systems of lithium hydride with the dihydrides of strontium, barium, europium, ytterbium, scandium, lanthanum, cerium, and samarium. The compound LiSrH_3 , with congruent melting point 745°, was found. Eutectic temperatures and compositions found, in mole % MH_2 , were: SrH_2 644.6°, 11.2%; BaH_2 670.8°, 5.0%; EuH_2 664.1°, 6.8%; YbH_2 668.7°, 6.8%. The dihydrides of the other rare earth metals, and also LaH_2 , did not affect the freezing point of LiH within experimental error. The soluble hydrides are the saline ones; the insoluble ones have metallic conduction electrons which contribute an interaction unfavorable to solubility in the saline molten LiH . The apparatus is described, and the purity and melting point of the lithium hydride are discussed.

Introduction

Earlier investigations in this laboratory, by the method of thermal analysis, on systems of LiH with Li metal,² LiF ,³ CaH_2 ,⁴ and Li_2O ⁴ showed behavior closely resembling that to be expected from the ionic

nature of LiH and its physical similarity to LiF . X-Ray crystallographic determinations of the probable inverse perovskite structures of LiBaH_3 ,⁵ LiSrH_3 ,⁵ and LiEuH_3 ,⁶ further substantiated the crystallographic similarity between LiH and LiF . No novel phases were found in the LiH-CaH_2 and LiH-YbH_2 systems.

However, it was also found that TiH_2 ⁴ and ZrH_2 ,⁴ which are considerably metallic in nature, failed to

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depress the freezing point of LiH, as the ionic CaH₂ did. Thus, these studies were extended to include several of the rare earth hydrides. These are presumably intermediate in nature between TiH₂-ZrH₂ and the alkaline earth hydrides, although mostly showing metallic characteristics.

Lanthanum, cerium, europium, ytterbium, samarium, and scandium were selected. Europium and ytterbium form stable orthorhombic dihydrides of the CaH₂ structure type,⁷ in contrast with the fluorite structure of the dihydride phases of the other rare earths. Samarium was selected because it is known⁸ to form compounds of the +2 oxidation state more readily than any of the rare earths other than Eu and Yb. Scandium was included to test the effect of its smaller ionic size. The definitely ionic SrH₂ and BaH₂ were also included.

Experimental

Apparatus.—The thermal analysis apparatus described in the earlier paper¹ was redesigned with a view to higher precision and more automatic controls.

The sample bomb (Figure 1), of Type 316 stainless steel, was constructed with two concentric chambers. The inner or sample chamber is identical with that of the earlier apparatus. The vacuum seal between cap and chamber is a replaceable lead gasket set on a copper base.

The outer chamber was provided to reduce diffusion losses of hydrogen from the sample chamber and to act as a radiation shield to reduce thermal gradients in the sample. The hydrogen pressure in the outer chamber can be adjusted separately from that in the sample chamber, the two normally being kept approximately equal.

The furnace is a Hevi-Duty three-element hinged tube-type, with the longer central heater supplying 890 watts and each end heater 380 watts, at 115 v. a.c.

The sample temperature is determined by two independent Honeywell Megopak chromel-alumel thermocouples, both run up from below with their tips against the top of the sample chamber well (E, Figure 1). One was connected to a Brown Elektronik recorder for continuous temperature recording, the other to a Rubicon thermocouple potentiometer to check the calibration of the recorder periodically. The normal range of the recorder was 550–750° in 1° divisions; this could be changed for occasional use outside this range by an auxiliary potentiometer. The thermocouples were calibrated against the freezing points of National Bureau of Standards samples of tin, zinc, and aluminum, in an assembly designed to fit into the furnace in place of the sample bomb.

One of the temperature sensors in the sample well (E), and one of the sensors at the same level in the shield (D), were 25-ohm three-lead platinum resistance thermometers of 1/8-in. o.d., encased in alumina powder and sheathed with Type 316 stainless steel. The currents from these resistors were fed to either of two bridges: one to control constant temperature and one to maintain a constant temperature difference between sample and shield. The bridge outputs were amplified to control a servomotor which actuated the variable transformer used to control the center element of the furnace.

The other temperature sensors were chromel-alumel Megopak thermocouple junctions, used to control the currents through the upper and lower furnace elements, limiting vertical temperature gradients to ±2°.

Materials.—The hydrogen gas was prepared by decomposition of uranium hydride. The lithium metal was Foote Mineral Co.

99.98% assay, received in large ingots and cut and trimmed of its thin oxide-nitride coating in an argon drybox.

Strontium and barium were obtained from Dominion Magnesium, Ltd., Toronto, Canada. Lanthanum, cerium, and europium were obtained from the Cerium Metals Division of Ronson Metals, Inc., Newark, N. J. Scandium, samarium, and ytterbium were obtained from United Mineral and Chemical Co., New York, N. Y. All were assayed by the suppliers as 99% or better. All samples were bulk metal and were cut or filed in the argon glove box or under mineral oil. In all cases the final weighed sample was lustrous in appearance.

Procedure.—All hydrides were synthesized as needed in the sample bomb from metal plus hydrogen. The metal samples, freshly cut and trimmed, were weighed to ±1 mg. in the argon glove box and placed in the sample bomb, which in turn was transferred and connected anaerobically to the vacuum-hydrogen line. Sample size was about 7 g. of Li, or the equivalent in sample volume.

The final temperature of hydriding was about 25° above the suspected phase change of highest temperature (usually about 650–750°), and the final pressure close to 1 atm.

The amount of hydrogen taken up was determined by *P-V-T* measurements. For eleven Li samples, H/Li was 1.012 ± 0.015. For the added metals, usually small in amount, the hydrogen absorbed was 85–100% of the theoretical amount based on MH₂.

In most cases the lithium was first hydrided, and one or more freezing runs were made on the resulting LiH. The other metal was then added in weighed increments in the drybox and hydrided, and the mixture run. For those mixtures with high percentages of the second metal, Li and the other metal were hydrided together.

Each sample of a given composition was annealed for 3–18 hr. before an initial cooling run, and for 0.5–3 hr. before each subsequent cooling or warming run. The cooling or warming rate was adjusted to about 1°/min. just before the onset of the first phase change.

Because the change of slope at the phase transition was sharper, the liquidus temperatures from the cooling curves were considered more reliable than those from warming curves.

Melting Point and Purity of LiH.—The average freezing point of eight samples of LiH, as evaluated directly from the temperature traces, was 690.8 ± 0.2° (95% confidence interval). If two anomalous samples were included, this average would be 690.2 ± 0.7°. The reproducibility on any one sample was 0.0–0.3°.

The purity of the LiH was evaluated from detailed analysis of one melting run and one freezing run (on different samples), which had each been extended over the entire process. Graphs of temperature *vs.* reciprocal of fraction in liquid state gave $T_0 = 691.24^\circ$ and 0.09 ± 0.01 mole % liquid-soluble, solid-insoluble impurity for the melting run, and 691.33° and 0.20 ± 0.03 mole % for the freezing run. These results are within the limits of purity of 99.6–99.9% normally reported for LiH.^{9,9} The cryoscopic constant of 354° (mole fraction)⁻¹ is obtained from the reported latent heat of fusion of 5210 cal./mole,¹⁰ which is close to the cryogenic value of 5095 cal./mole⁴ from this laboratory.

The indicated freezing point, extrapolated to 100% purity, is about 691.3°. This is 3° higher than the original published value,² the difference being attributed to better temperature measurement and control in the new apparatus and possibly to higher purity of the new samples.

The freezing point was observed to decrease slightly with increasing cooling rate, the extrapolation to zero cooling rate indicating a freezing point $1.5 \pm 0.5^\circ$ higher than that at 1°/min. Hence, the *absolute* freezing point of pure LiH cannot be better represented than $692 \pm 2^\circ$.

Results

The phase diagrams of the systems of LiH with SrH₂, BaH₂, EuH₂, and YbH₂ are shown in Figures 2, 3, and 4.

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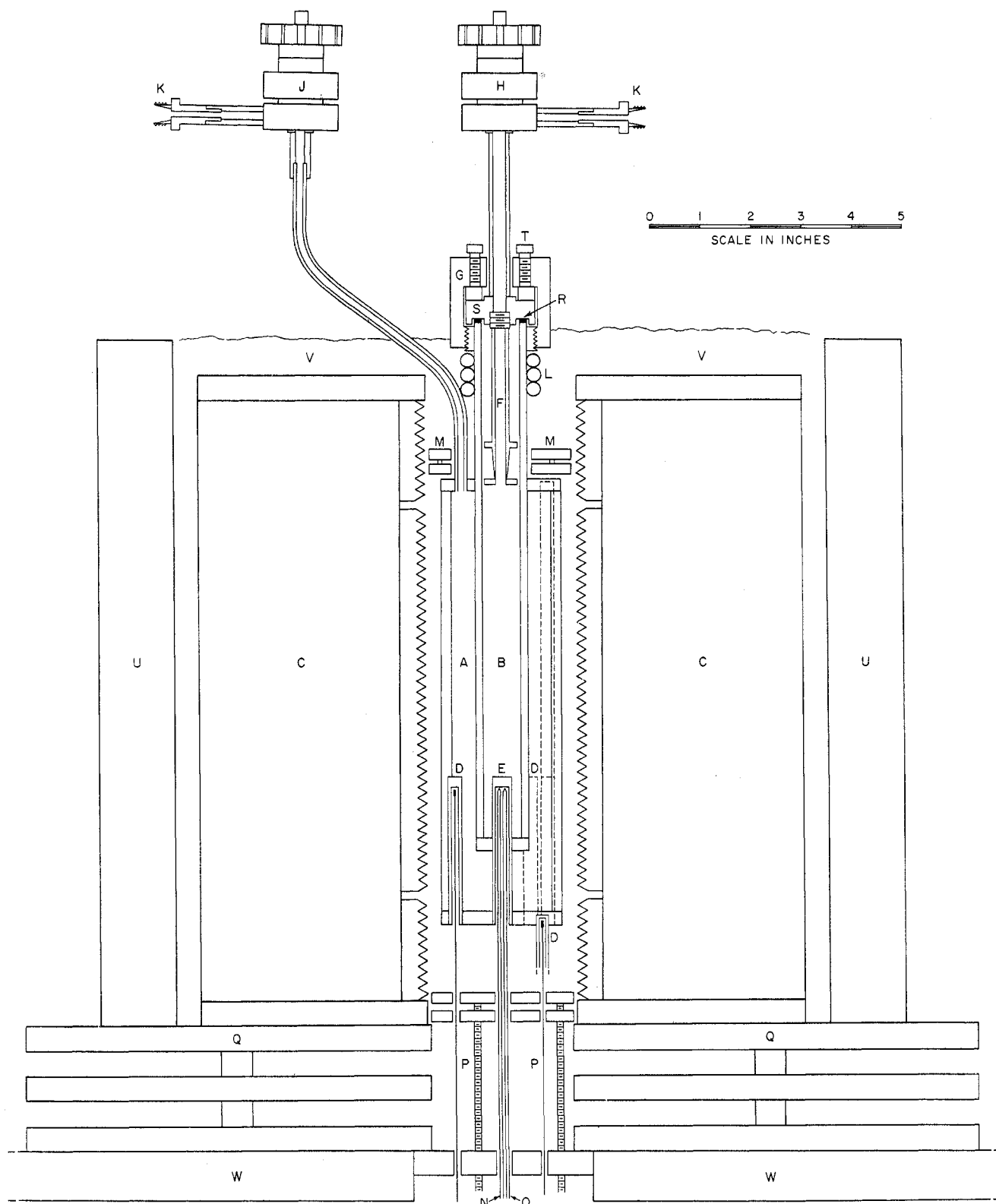


Figure 1.—Sample bomb and furnace: A, outer chamber; B, inner (sample) chamber; C, furnace; D, shield temperature sensor wells; E, sample temperature sensor well; F, baffle plates (internal); G, removable cap; H, J, diaphragm valves; K, fittings to vacuum line; L, cooling coil; M, P, baffle plates (external); Q, transit slabs; R, lead gasket; S, gasket ring; T, set screws; U, magnesia insulation; V, rock wool insulation.

The system LiH-SrH_2 was shown to contain the compound LiSrH_3 , of congruent melting point 745° , confirming the X-ray evidence of the perovskite structure.

The system LiH-BaH_2 could not be followed above 800° because of corrosion of the stainless steel container. The decrease in length of the eutectic pause with increasing BaH_2 content and the disappearance

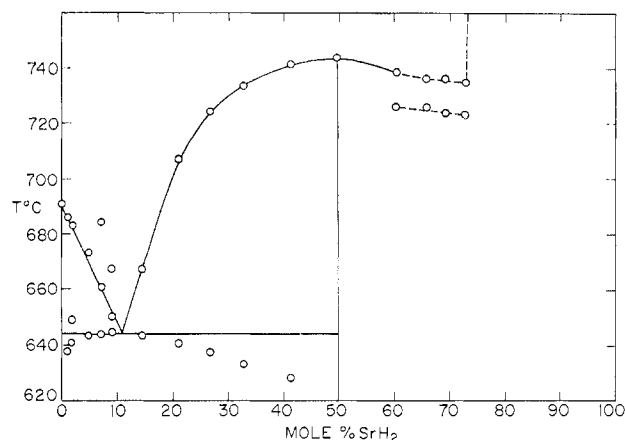


Figure 2.—The system lithium hydride–strontium hydride: eutectic 645°, 11.2 mole % SrH₂; congruent m.p. of LiSrH₃ 745°.

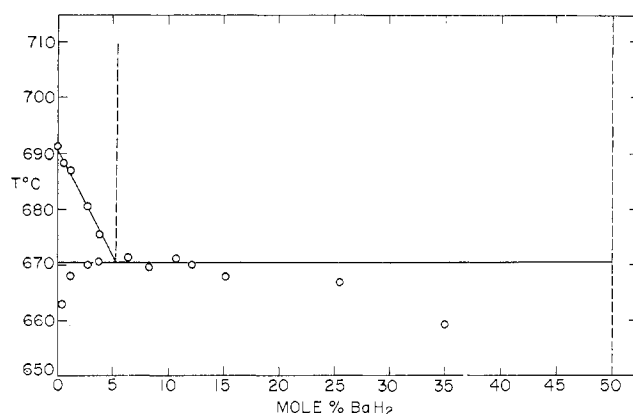


Figure 3.—The system lithium hydride–barium hydride: eutectic 671°, 5.0 mole % BaH₂.

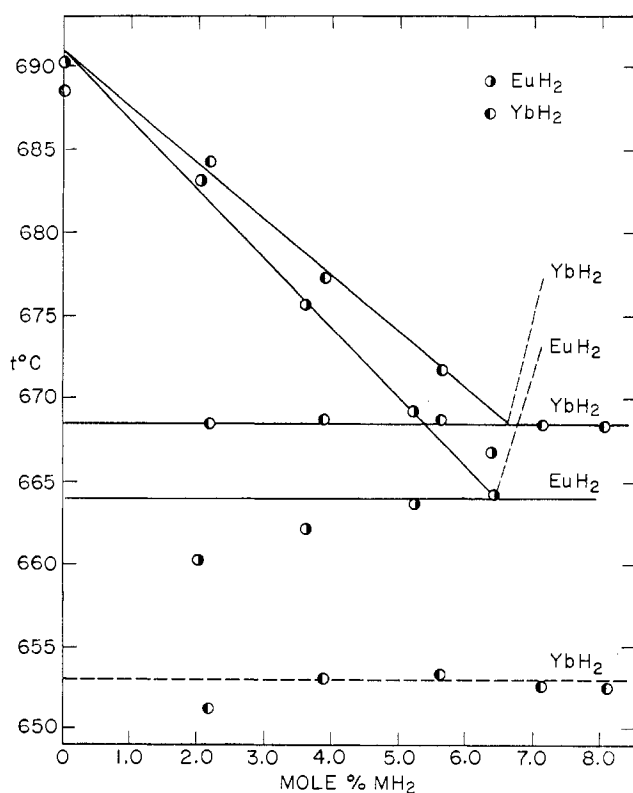


Figure 4.—The systems lithium hydride–europium hydride and lithium hydride–ytterbium hydride: eutectics: 664° and 6.8 mole % EuH₂; 669° and 6.8 mole % YbH₂.

of the eutectic at 45 mole % indicate LiBaH₃ as the composition of the second solid phase at the eutectic. The liquidus points in this system could not be detected beyond the eutectic composition, presumably due to very rapid rise of freezing point with composition in this region.

The high cost of europium unfortunately precluded the extension of the compositions investigated to include the range near 50% EuH₂ where evidence of the perovskite compound LiEuH₃ might have been found.

In all cases except YbH₂, the highest eutectic temperatures were obtained at the eutectic compositions, the temperatures tending to become increasingly lower as the composition LiH or LiMH₃ was approached. This may be attributed to liquid-soluble, solid-insoluble impurity, to increasingly poor heat transfer with decreasing amount of liquid phase present at the eutectic, or to both.

The addition of small amounts (2–4 mole %) of the hydrides of Sc, La, Ce, and Sm to lithium hydride was shown in each case to leave the freezing point of lithium hydride unchanged, within 0.7° or less. In all of these cases the rare earth hydride retained the form of the original metal, except for fissures due to the expansion on hydriding, and there was no visible evidence of solubility.

From known dissociation pressure data¹¹ the compositions of the rare earth hydride phases at 700° and 1 atm. are estimated to be ScH_{2.0}, LaH_{2.2}, CeH_{2.2}, and SmH_{2.2} (a higher hydride of Sc is not known).

To determine if increasing hydrogen content might lead to increasing solubility, a high pressure line was constructed to enable thermal analysis runs to be made up to 20 atm. hydrogen pressure. One of the Li–La–H samples was run under these conditions, again with no effect on the freezing point of LiH. The composition of lanthanum hydride at 700° and 20 atm. is about LaH_{2.5}.

Certain anomalous thermal arrests were found on certain samples and are shown in the figures. These cannot be unequivocally interpreted. They are presumably due to the formation of a small amount of solid phase before or after the principal arrest, possibly in a region not at equilibrium with the main portion of the sample. They are of two types.

(1) Two points each in the LiH–SrH₂ and LiH–EuH₂ systems at temperatures above the liquidus and at compositions just on the LiH side of the eutectic. The magnitudes of these arrests were 10–20% of the eutectic pause for the strontium system, and 5–10% for the europium system. The temperatures of the arrests were reproducible to ±1° or better, except in the case of the sample of 6.38 mole % EuH₂ where the reproducibility was to ±4%. Conceivably, these points might be due to the presence of the higher melting perovskite phase.

(2) A series of points in the LiH–SrH₂ system beyond 50 mole % SrH₂ was found at 724 ± 2°, 12° below the eutectic. A series in the LiH–YbH₂

system was found at $653.0 \pm 0.5^\circ$, 15° below the eutectic. The magnitudes of the heat effects were 25–50% and 5–25%, respectively, of those at the eutectic pauses. In the SrH_2 case it was definitely noted that the samples beyond 72% SrH_2 could not be completely melted, presumably because of high SrH_2 content.

A crystalline transition of the type known to exist for CaH_2 ,¹² SrH_2 ,¹³ and BaH_2 ,¹⁴ is also suggested by the constancy of the points. However, the known crystalline transition of SrH_2 is at 855° . Presumably YbH_2 would also have such a transition, but it is not yet known.

Figure 5 shows the departure of the saturated liquid phases from the ideal solution. The ideal line¹⁵ is

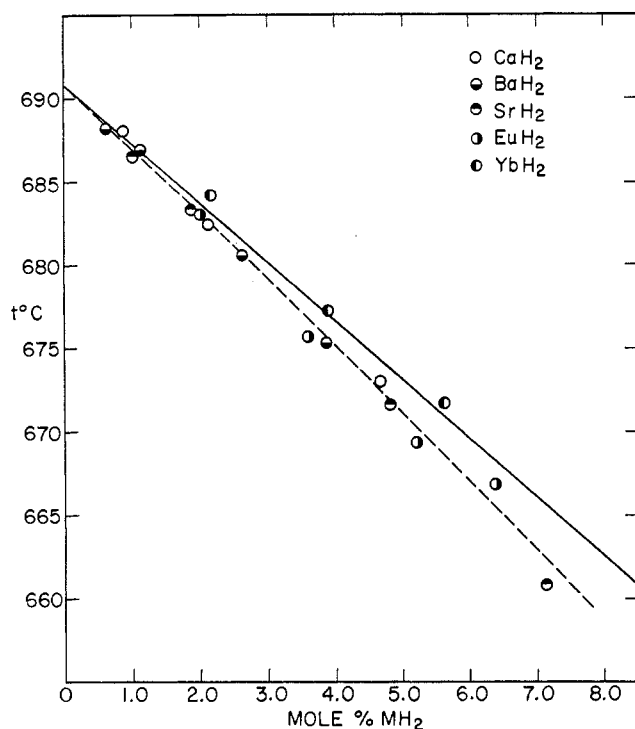


Figure 5.—Departure of equilibrium solutions of MH_2 in LiH from ideal behavior: —, ideal (Raoult's Law); ---, actual general trend (except YbH_2).

calculated using the values¹⁰ of $\Delta H_f^\circ = 5210$ cal./mole and $\Delta C_p^\circ = -2.6$ cal./mole deg. for the fusion of LiH at 690.8° . The temperatures for the CaH_2 points, from earlier research⁴ in a different apparatus, are raised by 2.5° , the difference between the freezing point values of LiH in the two researches. For all of the other points, from this research, the experimental values are used directly. Direct experimental liquidus points are used even though the freezing points of the corresponding LiH samples varied over several tenths of a degree, the assumption being made that errors in the freezing point lowering are random rather than

systematic, even for the case of the YbH_2 samples where the melting point of the original LiH was 687.3° .

With the exception of YbH_2 , the deviation is seen to be small and negative, the single dotted line representing the trend for four of the five solutes within experimental error.

Discussion

Libowitz and Gibb¹⁶ and Gibb and Schumacher¹⁷ have rationalized the ionic model of transition metal and rare earth hydrides on the basis of internuclear distances. Pebler and Wallace¹⁸ have discussed the electrical conductance and magnetic behavior of the rare earth hydrides on the basis of an ionic model. Similar behavior is also indicated in magnetic and electrical studies of the compounds of the rare earths with the elements of groups V and VI.¹⁹ On hydrogenation the electrons of the conduction band of the metal—two for Eu and Yb and three for the other lanthanides—are used in forming hydride ions, the +3 charge (+2 for Eu and Yb) of the rare earth ion being essentially unchanged.

Stalinski²⁰ has specifically shown, on the basis of magnetic and electrical conductivity studies, that lanthanum hydride is metallic in character with increasing hydrogen content up to about $\text{LaH}_{2.7}$, but at higher hydrogen concentrations becomes diamagnetic and temperature-dependent in electrical conductivity. It is to be noted that these changes occur at compositions higher than $\text{LaH}_{2.5}$, the highest achievable under the conditions of this research.

Europium and ytterbium dihydrides are essentially saline in nature and have the same orthorhombic structure as the dihydrides of Ca , Sr , and Ba . They lower the freezing point of LiH much as the alkaline earth hydrides do.

The other dihydrides, with the fluorite structure and one remaining electron per metal atom in the conduction band, show metallic conduction. They did not lower the freezing point of LiH , within experimental error. TiH_2 and ZrH_2 resemble these dihydrides in properties and structure.

The trihydrides seem to be ionic in nature and it would be predicted that they should dissolve in molten LiH . However, at 700° with lanthanum it was not possible to add hydrogen beyond $\text{LaH}_{2.5}$ at a pressure of 20 atm., the highest available in this research. At this composition, no solubility in LiH was detected.

Thus solubility in molten lithium hydride seems in general to be a satisfactory test for salinity of hydrides, although the fact that ytterbium and europium dihydrides have some metallic characteristics may indicate a limit to the general applicability of the test.

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The solubility of a hydride in liquid LiH should be dependent upon the difference between its ordinary lattice energy as a pure solid and its effective partial molar energy in the dissolved state in molten LiH at low concentrations. For solutes of the same structural type and bonding (here MH_2), the energy in the dissolved state at low concentrations might be nearly constant.

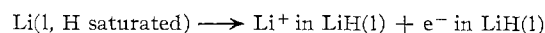
Gibb²¹ has evaluated the lattice energies of most of the known hydrides. The following are the values, calculated by the Born-Haber cycle, for several of the hydrides of this research (values in kcal./mole): CaH_2 , 570; SrH_2 , 540; BaH_2 , 520; LaH_2 , 610; TiH_2 , 700; and ZrH_2 , 700.

The soluble hydrides as a group do have lower lattice energies than the insoluble hydrides. However, the lattice energy of Li_2O is 693 kcal./mole,²² and Li_2O is soluble in molten LiH. The lattice energies of the difluorides of the transition metals are of the order of magnitude of 700 kcal.²¹ Cerium and plutonium trifluorides,²³ zirconium and uranium tetrafluorides,²³ and manganous fluoride²⁴ lower the freezing points of lithium and sodium fluorides down to more or less normal eutectics. Ferrous and nickelous fluorides form eutectics with NaF.²³ The lattice energy relationships for the fluoride systems resemble those for the hydride systems enough so that lattice energy per se cannot be the factor primarily restricting the solubility of the transition metal hydrides in molten LiH.

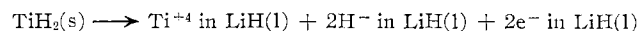
The lattice energies of the hydrides were calculated by Gibb assuming Ti^{+2} ions in TiH_2 and La^{+2} ions in LaH_2 . On the basis of the ionic model, TiH_2 would consist of Ti^{+4} ions, H^- ions, and conduction electrons. Lanthanum dihydride would similarly consist of La^{+3} ,

H^- , and conduction electrons. If such a hydride could be dissolved in molten LiH, the conduction electrons would have to be highly attenuated, and any delocalization energy due to metal-metal bonds would be lost.

The dissociation pressure data of Heumann and Salmon²⁵ for lithium hydride at 700° indicate that about 4% of the hydrogen may be removed from molten LiH before the metal phase appears. Thus, one might consider that mole fraction 0.04 represents the tendency of the following reaction to go at 700°.



The rare earth and transition metal dihydrides would have to undergo a similar process to release their conduction electrons to molten LiH.



These hydrides would require much more energy to give up their conduction electrons than would Li metal, thus would be much less soluble—enough less, apparently, so that solubility is undetectable by freezing point lowering.

A localization of the conduction electrons on the Ti or La, to form La^{+2} or Ti^{+2} ions, might conceivably occur as the hydride dissolved in molten LiH. Such a hydride might be expected to dissolve in molten LiH, as the transition metal difluorides would dissolve in molten LiF. The fact that the metallic dihydrides do not appear to dissolve in molten LiH indicates that such localization does not occur.

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