Requisite Temperatures for the Stabilization of Atomic H in Solid H₂

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Introduction

VALUES for the theoretical specific impulse in the neighborhood of and above 750 s are predicted if it is possible to employ an atomic hydrogen/molecular hydrogen propellant that contains at least 15% free H atoms by weight. Of current experimental interest among the methods 1 for the manufacture of such an H/H2 propellant is the tritiumimpregnation concept.² In this manufacturing scheme, primary and secondary energetic electrons from the β decay of tritium produce atomic H in solid H, at temperatures below 0.8 K by electron-impact dissociations of H₂ molecules. A phenomenological rate process theory has been developed for the matrix-isolation storage^{3,4} and equilibrium stability⁵ of atomic H produced at such ultralow temperatures in tritiumimpregnated H₂. Primary beta electrons emitted in tritium decay have a mean energy of 5.7 keV, and original theoretical estimates 1-5 predicted that about 30% of the tritium-decay energy would be storable under favorable conditions via H₂-2H dissociations involving matrix-isolated atomic hydrogen atoms in the final state. However, the most recent experiments⁶ suggest that an energy storage efficiency η greater than 0,30 is in fact attainable at temperatures below 100 mK. In order for the storage of atomic H to be stable with respect to arbitrary small perturbations,5 the surface temperature must be less than a critical value which depends on the energy storage efficiency η , the tritium weight fraction w, and the volume of the sample V. A concise derivation of the formula for this critical surface temperature is presented in the following section, with inclusion of the general dependence on η as a disposable parameter to be fixed by experiment.

Theory

If the fraction η of tritium-decay energy is stored in H_2-2H dissociations, then the fraction $(1-\eta)$ is dissipated in uniform volumetric heating. Let us assume here that no heat is removed locally from the sample volume (e.g., by fine copper wires through the sample) and that all heat is removed by thermal conduction to the surface of the sample, with the maximum temperature occurring at the center of the volume. From empirical expressions for thermal conductivity and specific heat of solid H_2 at temperatures below 0.80 K, we find the approximate formula for the temperature at the center of the sample [see Eq. (36) in Ref. 5]:

$$T_{\text{max}} = [T_s^4 + (0.589) (I - \eta) w V^{2/3}]^{1/4}$$
 (1)

in which T_s is the surface temperature in degrees Kelvin, w is the weight fraction of tritium in the sample, and V is the volume of the sample in cubic centimeters. For stability it is necessary and sufficient that the temperature be less than a certain critical value T_c at all points through the sample volume, where the critical temperature is defined implicitly by the formula [case of equality for first part of Eq. (20) in Ref. 5

Table 1 Values of T_{cr} , the critical temperature below which the equilibrium atomic hydrogen concentration is locally stable (units mK)

| | | W | |
|------|--------------------|-------|-------|
| η | 3.0×10^{-4} | 0.012 | 0.048 |
| 0.30 | 134 | 165 | 181 |
| 0.50 | 138 | 171 | 189 |
| 0.70 | 140 | 174 | 192 |

Table 2 Values of $(T_s)_{cr}$, the critical surface temperature of a sample volume with $V=0.10~{\rm cm}^3$, below which the equilibrium atomic hydrogen concentration is stable throughout the sample (mK, with three dots denoting instability for all surface temperatures)

| η | | w · | ν . | |
|------|----------------------|-------|-------|--|
| | 3.0×10^{-4} | 0.012 | 0.048 | |
| 0.30 | 131 | | | |
| 0.50 | 136 | 98 | | |
| 0.70 | 139 | 146 | | |

with $T_0 \ll T_e$]

$$T_{cr}^{-1}[\exp(-2.8/T_{cr})] = 7.23 \times 10^{-5} \eta w$$
 (2)

Table 1 shows the critical temperatures given by Eq. (2) for selected values of η and w of current experimental interest, with $9 \times 10^{-5} \le \eta w \le 3.36 \times 10^{-2}$. Over the latter range of values for ηw , the solution to Eq. (2) is expressed explicitly as

$$T_{cr} \cong 2.8[\zeta + \ln(\zeta + \ln\zeta)]^{-1} K \tag{3}$$

$$\zeta \equiv ln(4.94 \times 10^3 / \eta w)$$

to an accuracy better than 0.5% (i.e., to within about 1 mK). By substituting Eqs. (1) and (3) into the stability condition $T_{\rm max} < T_{cr}$, one obtains $T_{s} < (T_{s})_{cr}$, where the critical surface temperature is

$$(T_s)_{cr} = [61.5[\zeta + \ln(\zeta + \ln\zeta)]^{-4} - (0.589)(1 - \eta) w V^{2/3}]^{\frac{1}{6}} K$$
(4)

Table 2 shows the critical surface temperatures given by Eq. (4) with $V=0.10~{\rm cm}^3$ and for the selected values of η and w that appear in Table 1. In the case of the currently attainable surface temperature $T_s=50~{\rm mK}$, energy storage efficiencies $\eta>0.45$ and 0.77 would have to be manifest for stabilization with w=0.012 and 0.048, respectively.

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