

# Y-12

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LOCKHEED MARTIN ENERGY SYSTEMS, INC.  
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UCN-13672 (26 6-95)

CONF-960855--1

Y/DZ-1185/R1

### SOLUBILITY OF HYDROGEN AND DEUTERIUM IN bcc-URANIUM-TITANIUM ALLOYS

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RECEIVED  
DEC 20 1996

OSTI

Date of Issue: August 5, 1996

International Symposium on Metal Hydrogen Systems,  
Fundamentals and Applications  
Les Diablerets, Switzerland  
August 25-30, 1996

MASTER

Prepared by the  
Oak Ridge Y-12 Plant  
Oak Ridge, Tennessee 37831  
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LOCKHEED MARTIN ENERGY SYSTEMS, INC.  
for the  
U. S. DEPARTMENT OF ENERGY  
under contract DE-AC05-84OR21400

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## SOLUBILITY OF HYDROGEN AND DEUTERIUM IN bcc-URANIUM-TITANIUM ALLOYS

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### ABSTRACT

For the bcc-U-Ti alloy system, H and D solubility measurements have been made on 12 alloy specimens ranging in composition from pure U to pure Ti and temperature range bounded by 900 K to 1500 K. The results are described by a model within a standard error of 3%.

### INTRODUCTION

The U-Ti alloy system has a high temperature (~900°C) region in which the bcc phase extends unbroken across the composition range from pure U to pure Ti/1/. Over this range, the solubility of hydrogen in these alloys represented as the equilibrium constant (k), the ratio of the H concentration to the square-root of the H<sub>2</sub> pressure, varies by almost three orders of magnitude. It can be measured near infinite dilution without serious experimental interference from factors such as hydride formation, slow surface and diffusion kinetics, or trapping effects. In the context of the bound proton model/2-5/, k is described as

$$\ln(k) = \ln(N) + \ln(1 + Ae^{-BT}) - 3\ln(1 - e^{-CT}) + (E - 3C/2)/T - 0.5\ln(LT^{7/2}/(1 - e^{-J/T})) - M/2T \quad (1)$$

where N = 3 or 6 is the number of H sites per bcc metal atoms, C ( $C_D = C_H / 2^{0.5}$ ) is the Einstein temperature, E is the ground state energy relative to atomic hydrogen at rest, the last two terms

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together is the free energy function for H<sub>2</sub> (or, with different parameters, the free energy function for D<sub>2</sub>)/4/, and the term containing A and B is an approximation for Boltzman states not explicitly included in the harmonic oscillator model for H in its site.

## EXPERIMENTAL

H solubility measurements/2-4/ were made at infinite dilution with 1% accuracy on 12 alloy specimens representing Ti compositions in mole fraction of 0.000, 0.018, 0.036, 0.060, 0.080, 0.147, 0.185, 0.242, 0.335, 0.597, 0.850, and 1.000. Similar measurements were made for D except for the Ti compositions 0.060 and 0.185. The temperature range for these solubility measurements extended from the lesser of the liquidus temperature (1406 K for U) or 1473 K down to the solvus line for phase separation that ranged from 1173 K for the composition of U<sub>2</sub>Ti to temperatures as low as 900 K for Ti composition 0.850.

## RESULTS

Preliminary analyses indicated that eq. I would describe each alloy with a precision comparable to the measurement precision for N = 3, B = 3600 K, and C = 1680 K, with A and E optimized for a particular alloy. Difficulty was encountered in fitting high Ti composition with N = 6 since the A,B term was very small at the low temperature extremes, and the desired slope and magnitude of eq. I could not be simultaneously achieved. To describe the alloy effect, a more global model was employed for which N=3, B=3600 K, C<sub>H</sub>=1680 K. A and E were optimized using all the ln(k<sub>H</sub>) data with the following form Q(x) to describe the alloy (x) dependence of A and E.

$$Q(x) = a\{x-1/g \ln(\cosh[g(x-b)])\}/2 + c\{1/g \ln(\cosh[g(x-b)]) - 1/g\{\ln(\cosh[g(x-d)])\}\}/2 + e\{1/g \ln(\cosh[g(x-d)])+x\}/2 + f \quad (\text{II})$$

Eq. II describes 3 linear segments that are smoothly connected. For A, the parameters were a=-1131.1221, b=-0.031043, c=17.2527, d=0.296235, e=0.37877, f=9.969, g=17.5788, fit standard

error=0.595. For E,  $a=28957.2$ ,  $b=0.02406$ ,  $c=8179.18$ ,  $d=0.28692$ ,  $e=4833.22$ ,  $f=29358.8$ ,  $g=15.4827$ , fit standard error=32.1. Using the above parameters, the data for D was fit with C as the only variable. The Einstein temperature was found to be 1680.9 K with a fit standard error of 7.9 K. The overall fit standard error was 3%.

### CONCLUSIONS

The Einstein temperature ( C ) is not a function of alloy composition. The ground state energy ( E ) varies from 28908 K ( $A=16.73$ ) for U to 35462 K ( $A=2.71$ ) for Ti, a change of -0.56 eV. Inflections occur at U-0.19Ti ( $E=31189$ ,  $A=4.27$ ) and U0.29Ti ( $E=31927$  K,  $A=2.81$ ). The contribution from thermally excited states also grew markedly as the concentration of titanium approached zero. This large increase in hydrogen solubility in U resulting from small additions of Ti is consistent with the observation that there is total transfer of valence electrons from Ti to U at concentrations below that of U<sub>2</sub>Ti /6/. The A, B term is apparently due to electronic factors.

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**International Symposium on Metal Hydrogen Systems, Fundamentals and Applications**

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