

Diffusivity and solubility of protium and deuterium in tungsten derived from a combined experimental and modelling approach

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Robust prediction of tritium transport and retention in tungsten plasma-facing materials for future fusion devices depends on reliable data for solute diffusion and solubility. Therefore, extensive resources are invested for modelling. The basic data for these modelling efforts rely on measurements for protium, originating from the late 60's [1]. Lacking measured data for deuterium and tritium, a classical mass-ratio-based extrapolation is used to extrapolate to deuterium and tritium. However, other experimentally derived values show a substantial scatter, likewise recent simulations, e.g., based on DFT. Thus, the parameters of solute diffusion and solubility appear not to be very reliable [2]. Therefore, a state-of-the-art approach, combining experiment and simulation, was undertaken to establish these values for protium and deuterium. The experimental set-up is similar to the set-up of [1] but with several improvements. A cylindrical tungsten specimen of 30 mm in diameter was used to conduct gas loading in 500 mbar protium or deuterium at elevated temperature. After a cooling-cycle the sample was degassed by ramping up to constant temperature. The measurements were conducted between 1800 and 2400 K. At these temperatures, trapping and surface effects have been shown to be negligible. Data sets, consisting of seven measurements for protium, and nine for deuterium, respectively, were simultaneously fitted by spectral modelling. Relying on a Bayesian approach, the 4-dimensional probability density of the model parameters (the activation energies for diffusion and solution, E_A and E_S , as well as the corresponding pre-exponential factors D_0 and S_0) was explored using Markov-Chain Monte-Carlo (MCMC) sampling. Post-processing of the coefficients for solubility and diffusivity yields a substantially lower activation energy E_A for protium diffusion than the formerly recommended one from Frauenfelder [1], [2].

For the first time, also a reliable value for the solute diffusion of deuterium was determined. The measured quotient of the pre-exponential factors is in reasonable agreement with the prediction of Vineyards transition state theory ($\frac{D_0^H}{D_0^D} \propto \frac{\sqrt{m_D}}{\sqrt{m_H}} = \sqrt{2} = 1.41$ [3]) if one assumes the same activation energy for both isotopes. Based on this outcome, the extrapolation to the diffusivity of tritium shall be realised by the usage of the revised activation energy of deuterium and the classical mass scaling [3].

The solubility for protium and deuterium is found to be equal, with noticeably higher solute energy E_S compared with [1].

[1] R. Frauenfelder, „Solution and Diffusion of Hydrogen in Tungsten“, J. Vac. Sci. Technol. 6, 388, 1969

[2] R. Causey and T. Venhaus, “The use of tungsten in fusion reactors: A review of the hydrogen retention and migration properties”, Physica Scripta T, vol. 94, pp. 9-15, 2001

[3] G. H. Vineyard, “Frequency factors and isotope effects in solid state rate processes“, *Journal of Physics and Chemistry of Solids*, vol. 3(1), 121-127, 1957