

Positron Life Time Calculations of Defect in α -Iron Containing Hydrogen

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Abstract. Positron lifetime quantum-mechanical calculations were carried out for investigation of point defects in α -Fe containing hydrogen atoms. The electron wave functions have been obtained in the local density approximation [LDA] to the density functional theory [DFT]. On the bases of calculated results, the behaviour of empty nano-voids and nano-voids with hydrogen were discussed. It was found that hydrogen in larger three-dimensional vacancy clusters changes the annihilation characteristics dramatically. The hydrogen atoms are trapped by lattice vacancies. These results provide physical insight for positron interactions with defects in α -Fe and can be used for prediction of hydrogen generation for the design of fusion reactors.

Keywords: Model Calculations, Defects in Solids, Positron Life Time.

INTRODUCTION

The knowledge on the microstructure evolution in pure α -Fe is very important [1]. Positron spectroscopy is capable of monitoring the void formation process over the full range of vacancy-cluster size and it is capable of monitoring of micro-voids even at the earliest stages, which has a strong impact on understanding the micro-void formation phenomena in advanced fusion reactor materials. The method is based on positron trapping at open volume defects and it may be used for the characterization of mono-vacancies and vacancy clusters. This paper treats the positron lifetime calculations with defects in α -Fe lattice containing hydrogen atoms. The electronic structure calculations for super cell size are studied for vacancies, nano-voids and vacancy clusters containing hydrogen. The investigations were motivated by an interest to vacancy cluster behaviour in neutron-irradiated metals of the fusion reactors design. Iron is important material used in the fusion reactors due to its high temperature melting point (1811°K) and other specific characteristics like Young modulus (211GPa), bulk modulus (170GPa), Brinell hardness (490MPa) etc. more than one part and each part should be labels (a), (b), etc.

Calculation Method

The computer simulations were performed using Finnis-Sinclair [FS] N-body potential [2]. The total energy U_{tot} of the model crystal consists of two terms:

$$U_{\text{tot}} = U_{\text{N}} + U_{\text{p}}, \quad (1)$$

where U_{N} is the *many-body* term and U_{p} is the *pair* term. U_{N} is written as:

$$U_{\text{N}} = -A \sum_i \sqrt{\sum_{j(\neq i)} \phi(R_{ij})}, \quad (2)$$

where R_{ij} is the distance between the i -th atom and the j -th atom and

$$\begin{aligned} \Phi(R) &= \{(R-d)^2 + \beta(R-d)^3/d, & R \leq d \\ &= 0, & R > d \end{aligned} \quad (3)$$

for iron. The pair potential term U_{p} is written as

$$U_{\text{p}} = \frac{1}{2} \sum_{ij} V_{\text{p}}(R_{ij}), \quad (4)$$

$$\text{where } \begin{aligned} V_{\text{p}}(R) &= \{(R-c)^2 (c_0 + c_1 R + c_2 R^2), & R \leq c, \\ &= 0, & R > c \end{aligned} \quad (5)$$

The positron lifetimes in the defects are calculated according to the method of Puska and Nieminen [3, 4]. This method gives a reliable positron lifetime value. The electron density distribution $n(\mathbf{r})$ in a crystal is approximated by superimposing the spherical averaged charge densities $n_{\text{at}}(\mathbf{r})$ (r is the distance from the atomic center) of an isolated atom:

$$n(\mathbf{r}) = \sum_i n_{\text{at}}(|\mathbf{r} - \mathbf{R}_i|), \quad (6)$$

where \mathbf{R}_i is the position at which the i -th atom is placed. The Coulomb potential $V_{\text{C}}(\mathbf{r})$ is, therefore, represented by superimposing the function $V_{\text{at}}(r)$ of distance r from an atom:

$$V_{\text{C}}(\mathbf{r}) = \sum_i V_{\text{at}}(|\mathbf{r} - \mathbf{R}_i|). \quad (7)$$

The correlation potential $V_{\text{corr}}(n(\mathbf{r}))$ is due to the interaction between the positron and the host electron. Boronsky and Nieminen [5] have determined the practical form of the function $V_{\text{corr}}(n)$. The total potential felt by a positron is

$$V(\mathbf{r}) = V_{\text{C}}(\mathbf{r}) + V_{\text{corr}}(n(\mathbf{r})). \quad (8)$$

The positron wave function $\psi_+(\mathbf{r})$ as the numerical solution of the Schrödinger equation can be obtained through the iteration process of the Kimball-Shortley method

[6]. The energy eigenvalue of the positron wave function is obtained for every iteration, and this process is continued until the eigenvalue reaches a saturated minimum value.

The positron annihilation rate is expressed by

$$\lambda = \pi r_0^2 c \int dr n_+(r) \times [n_v(r) \Gamma_v(n_v(r)) + n_c(r) \Gamma_c + n_d(r) \Gamma_d], \quad (9)$$

where n_+ , n_v , n_c and n_d represent density of positron, valence, core and d-electrons, respectively. Γ_v , Γ_c and Γ_d are the corresponding enhancement factors due to the correlation between a positron and electrons. According to Brandt and Reinheimer [7];

$$\Gamma_v(n_v) = 1 + (r_v^3 + 10)/6,$$

where $r_v = (3/4\pi n_v)^{1/3}$ is the density parameter of the valence electrons. For core electrons, Γ_c has been estimated as 1.5. We determined the constant value for Γ_d so that the calculated lifetime in the matrix agrees with the experimental result. When we set the $\Gamma_d = 2.20$, the matrix lifetime 108 [ps] is reproduced in iron. From the annihilation rate λ , the positron lifetime is given by $\tau = 1/\lambda$.

Calculating process for a matrix with dimension 42x42x42 (total 18,522 atoms) was used. The defect calculations were done at constant volume and relaxing only the atomic position in a super cell at equilibrium lattice parameter for α -Fe equal to 2.85 Å.

Results and Discussion

The positron lifetime calculations have been performed in a bcc iron lattice containing vacancies, empty nano-voids and nano-voids consisting of 1-28 vacancies and for vacancies containing equal number of hydrogen atoms. The lifetime of a trapped positron is sensitive to the vacancy defects. In FIGURE 1 is shown the isometric plot of the calculated positron density distribution (PDD) in an iron vacancy.

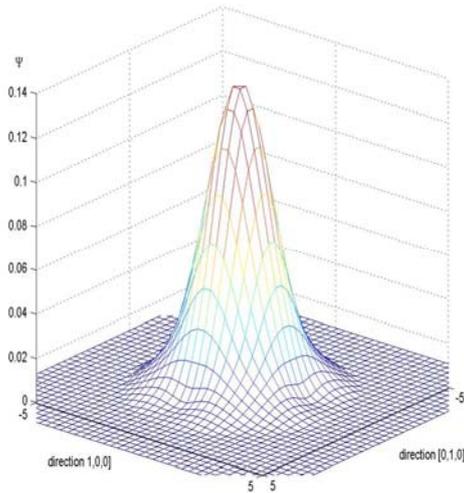


FIGURE 1. Localized positron wave function in bcc α -Fe with one vacancy

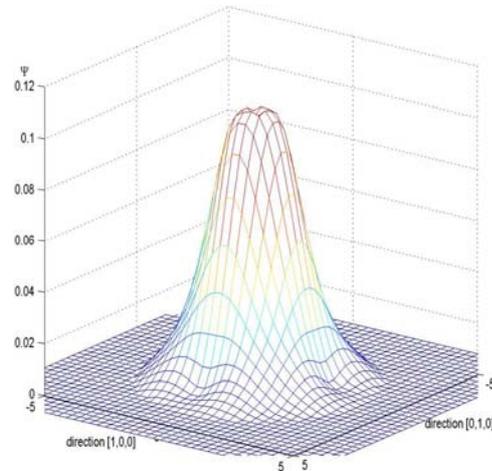


FIGURE 2. Localized positron wave function in bcc α -Fe with one vacancy and one hydrogen atom

The structure of vacancies in iron has been determined experimentally by positron interactions with defects [8] and therefore it is suitable for testing the computational method [9,10]. The lifetime of a trapped positron is larger than that of the positron annihilating in the free state, because the density of electrons inside a vacancy is less than that in the interstitial region. In FIGURE 2 are shown the isometric plot for the computed PDD in an iron mono-vacancy bound to a hydrogen atom (1v-1H). The isometric of the computed PDD show that a central hole appears in the PDD compared to the case of an empty vacancy. This can be explained by the presence of the hydrogen atoms around the center of the void resulting in the exclusion of positrons from the center of the complex. The computed positron lifetime for an iron mono-vacancy is 179 [ps] and for 1v-1H is 146 [ps], respectively. We wish to point out that in the present work, relaxation calculations for lattice atoms have been performed to obtain the energetically most “stable” configuration. At the beginning of the computation, the included atoms are located at the center of the created vacancy. The relaxation of the complex is computed, while coordinates of the lattice “border” atoms are not fixed. In the vacancy-hydrogen case when relaxation is considered, the electron density distribution is spread not in the center of the hole but at the vacancy border, while PDD is predominantly at the center of the vacancy. This explains the computed longer positron lifetime value. The present calculation gives a value of positron lifetime as 108 [ps] for a perfect iron lattice. The calculated value of the positron lifetime in iron is supported by the experimental value of 107 [ps] reported earlier in the literature [8], the values are in good agreement. Our calculations show that a positron in iron is bound to open defects even if they contain hydrogen atoms. The lifetime of positrons in voids is longer than that for a mono-vacancy and gives information on the presence of vacancy-clusters with two or more vacancies. The variation of the calculated positron lifetime τ as a function of the vacancy-clusters size is shown in FIGURE 3, where a correlation between the number of vacancies and

positron lifetime can be seen. The longer lifetime corresponds to three-dimensional vacancy-clusters, namely, nano-voids, which are observed in bcc and fcc metal lattices.

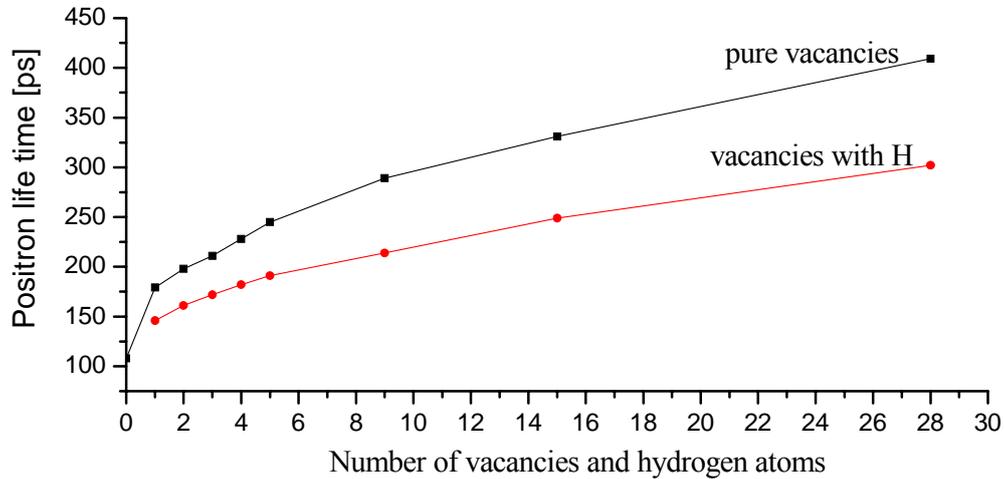


FIGURE 3. Calculated positron lifetime as a function of vacancies number(v) and vacancies bound with hydrogen atoms(H): $nH = nv$

CONCLUSIONS

The calculated results show that the interaction of positrons into one type of defects is different from trapping into other types of defects and with positron interaction in the bulk - 108 [ps]. Typical increasing of positron lifetime depends on the size of defect in the lattice. The calculated positron lifetime for a nano-void with hydrogen atoms is shorter than that for an empty nano-void of the same size, because the electrons from the center of the void attract positrons. The vacancy clusters in α -Fe without hydrogen are active positron traps, if once they are bound with hydrogen, they become less effective in the trapping of positrons. The DFT calculation of positron lifetime for vacancies and nano-voids containing hydrogen leads to interesting practical applications since hydrogen, empty voids and voids containing hydrogen are expected to have different diffusive properties in new materials.

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