Brief Reports

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Comments on positron annihilation and the vacancy properties of Mg

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Recent publications concerning the vacancy properties of Mg, as deduced from positron-annihilation-spectroscopy data, are judged in the light of earlier information based on lattice parameter and bulk thermal expansion data and tracer self-diffusion results. It is apparent that the separate analyses of the data from these various sources in terms of conventional monovacancy properties lead to inconsistent conclusions when they are considered as a whole.

It has been shown that correlations between the threshold temperature (T_v) for positron trapping at vacancies in metals (under conditions of thermal equilibrium), with the melting temperature T_m, or the vacancy formation energy h^f, depend, essentially, on two features.[1] One is that the product of the specific trapping rate of positrons at vacancies (μ) and the free or bulk lifetime of the positron (τ) is similar for all the metals concerned and the other is that h^f has an approximately linear dependence on T_m.

If the measured positron annihilation parameter is F, and the limits F^f and F^v correspond to values of F for annihilation from the free and (vacancy) trapped states, respectively, it can be shown[1] that at T_v, the product $\mu\tau$ bears the following relation to the equilibrium vacancy concentration C_v,

$$C_v(T_v) = \left[\left(\frac{\Delta F}{\delta} - 1\right)\mu\tau\right]^{-1} , \tag{1}$$

where $\Delta F = (F^v - F^f)$ and δ refers to the relative sensitivity of the measurement[1] [all values in (1) refer to $T = T_v$]. In the earlier work[1] the data were analyzed for $\Delta F/\delta = 20$ or, in terms of Eq. (1),

$$C_v(T_v) = (19\mu\tau)^{-1} . \tag{1a}$$

Two very recent investigations of positron annihilation in Mg (Refs. 2 and 3) have shown an apparent value of 720 K for T_v, a value of 5 for $\Delta F/\delta$ and, from one of the reports,[3] an apparent bulk lifetime of 240 ps at 720 K. The data show that the low value of $\Delta F/\delta$ stems from a relatively small value of ΔF, compared to results for other metals where strong vacancy trapping is evident.

For Mg, in terms of Eq. (1), then

$$C_v(720\text{ K}) = (4\mu\tau)^{-1} . \tag{1b}$$

Earlier work[4] on the relative bulk and lattice parameter thermal expansion $\Delta(1,a)$ of Mg, shows that C_v at 720 K is $\approx 9\times10^{-5}$. Insertion of this value for C_v and the above result for τ leads to $\mu = 1.1\times10^{13}\text{ s}^{-1}$. The result for μ and the product $\mu\tau$ are 20 times lower than corresponding values for other metals where strong positron-vacancy interactions are apparent.[1] This seems to be in sympathy both with theoretical predictions of the strength of positron-vacancy interactions in Mg (Ref. 5) and also with the relatively small effect of defect trapping on the positron signal vis-à-vis such effects for other metals[2,3]; it also suggests that it is invalid to apply the usual T_v/h^f correlations to the positron-annihilation-spectroscopy (PAS) data for Mg.

In both of the PAS investigations of Mg it was assumed (1) that the signal at T_v (720 K) was due to positron interactions with equilibrium monovacancies, and (2) that the empirical formula[6]

$$h^f \approx 14k_BT_v \tag{2}$$

could be applied (k_B is Boltzmann's constant). The outcome led to associated vacancy parameters which either directly or indirectly are inconsistent with the results of the $\Delta(1,a)$ work.

For example, a plausible set of vacancy-related parameters, judged to be consistent with the PAS data, was given[3] as $h^f = 0.85$ eV, $S^f = 2k_B$, and $\mu = 1.5\times10^{14}\text{ s}^{-1}$ (S^f is the entropy of vacancy formation). Substitution of the product $\mu\tau (= 1.5\times10^{14}\times220\times10^{-12})$ into (1b) or the above values of

TABLE I. Vacancy concentrations (C_v) and self-diffusion coefficients (D) for close-packed metals at their melting temperatures.

Metal	$C_v \times 10^4$	$D \times 10^9$ cm^{2}s^{-1}	Ref.
Mg	7		4
	1.7		3
	0.4		2
		25	8, 9
Al	9		10
		18	11
Au	7		12
		14	13
Cd	5		14
		14	15
Zn	5		16
		13	17
Ag	2		18
		7	19
Cu	2		20
		6	21
Pb	2		22
		0.7	23

h^f and S^f into the usual equation for C_v,

$$C_v = e^{S/k} e^{-h^f/kT}, \quad (3)$$

leads to $C_v(720\text{ K}) \approx 5 \times 10^{-6}$, which is an order of magnitude lower than the corresponding value determined by the $\Delta(1,a)$ method.

It has been suggested[7] that the results of the $\Delta(1,a)$ study of Mg may be subject to uncertain error, associated with the use of more than one sample for the measurements. It is apparent, however, that at least in one respect, namely, the value of $C_v(T_m)$, the results of the $\Delta(1,a)$ study are more in line with the general systematics of vacancy-dependent properties of close-packed metals than are the Mg vacancy parameters inferred from the positron annihilation data.[2,3] This is illustrated in Table I, where it is shown that higher values of $C_v(T_m)$ are generally associated with higher values of the self-diffusion coefficient at T_m. This trend is completely contradicted by the $C_v(T_m)$ values for Mg deduced from the h^f and S^f parameters assumed in the PAS work.

The information available at present suggests that the vacancy concentrations of Mg may not be abnormally low and that the relatively small effect of positron trapping at vacancies on characteristic positron-annihilation parameters may be indeed symptomatic of a relatively low value for the specific positron trapping rate at vacancies in Mg.

Two steps which might help to resolve the present uncertainty regarding the vacancy defect properties of Mg are (1) to make further direct measurements of C_v via the $\Delta(1,a)$ technique, but avoiding the experimental features which led to criticism[7] of the original study[4] and (2) to use the slow positron beam technique which, apparently, has a relatively high sensitivity to weakly trapping defects,[24] to make a further study of the equilibrium temperature dependence of positron annihilation in Mg.

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