POSITRON ANNIHILATION IN TEKTITE AND HYALITE

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Positron lifetime spectra as well as one-dimensional angular distributions of photons coming from the two-photon annihilation of an electron–positron pair were measured at room temperature for samples of natural tektite and hyalite. The data were used for estimation of positronium characteristics in the specimens. The characteristics were analysed in terms of parameters of pores trapping the positronium atoms before their decay. In the analysis two models of the traps were taken into account, i.e. the square potential wells of spherical symmetry with barriers of finite and infinite heights. It was found that the linear dimensions of the pores resulting from these two-trap models are almost the same and they are of the order of 1 nm.

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1. Introduction

For the last several years there has been a growing interest in studying the microstructural properties of naturally occurring materials for which different degree of crystallinity is observed including natural glasses (see [1] for example). It seems that this interest is mainly due to technological applications of amorphous materials.

Experimental investigations of the microstructure of minerals are performed by various techniques making use of phenomena sensitive to the structure of matter. Among the techniques a family of positron annihilation methods is known as a useful tool although it is not very popular among mineralogists. The nondestructive methods use positron and positronium atoms (Ps — the positron–electron bound state) as microscopic probes for the research of matter in different states. Both positrons and positronium atoms have a strong tendency to become localized in low-electron-density sites of condensed matter. Existing of the trapped positrons and positronium atoms in the studied material reflects on the positron

lifetime spectrum and the characteristics of positron annihilation radiation. It makes positron annihilation techniques suitable tools for studying the positron and positronium traps. They can be as vacancies and voids in crystals or atomic-scale holes in amorphous materials for example. Recently, the positronium atoms have widely and successfully been used as probes for a study of the properties of pores in some polymers [2]. This encourages us to perform such studies for natural glasses. In this paper we present the results obtained for two naturally occurring silica materials — tektite and hyalite (perfectly glass-clear opal). As far as we know the materials have never been investigated with positrons and positronium atoms.

2. Experimental and results

The room temperature measurements of lifetime spectra (LS) and one-dimensional angular distributions (AD) of photons coming from the two-photon annihilation of an electron–positron pair were carried out for samples of natural tektite from Czecho-Slovakia called moldavite [3] as well as hyalite from Jordanów in Poland [4]. The specimens in sizes of about 10 mm×10 mm×2 mm had natural or fracture surfaces.

LS were collected by using a lifetime spectrometer with the full width at half maximum (FWHM) of the time resolution function of about 350 ps. Its details are given elsewhere [5]. As the source of positrons the ^{22}Na isotope of the activity of about 4×10^4 Bq was used. It was sandwiched between two identically prepared pieces of the sample. AD were recorded by means of an automated angular correlation spectrometer with the long-slit geometry and FWHM of the angular resolution function of about 1 mrad. The description of such a spectrometer can be found in [6]. During the AD measurements specimens were one-sidedly exposed to positrons provided by a 10^8 Bq source of ^{22}Na.

LS were measured in the time range of 0–14 ns making use of 256 channels of the multichannel analyzer. The total number of counts for one LS was $(7-10)\times10^5$. The spectra were decomposed into three exponentials with different mean lifetimes (τ_1, τ_2 and τ_3) and intensities (I_1', I_2' and I_3') by using the mathematical model of a spectrum proposed by Dauwe et al. [7]. The determined values of τ_3 and I_3' characterizing the long-lived component of LS (the mean lifetime greater than 0.6 ns) are given in Table I. The AD measurements were carried out for the angles from −34 to +34 mrad with the step of 0.5 mrad. The total number of counts for one AD was 1.5×10^6. The obtained distributions were analysed in terms of a sum of three gaussians with different FWHM (Γ_1, Γ_2 and Γ_3) and intensities (I_1, I_2 and I_3). The fitted parameters of the narrowest gaussian, Γ_1 and I_1, are presented in Table I.

A further elaboration of the obtained results was performed under assumption that the contribution to AD described by the narrowest gaussian is associated with the *para*-positronium (the total spin $S = 0$) intrinsic annihilation [8] whereas the long-lived component of LS is conditioned by the pick-off annihilation of positrons forming the *ortho*-positronium atoms (the total spin $S = 1$). This enabled us to use the parameters Γ_1 and τ_3 for determining characteristics of pores trapping the positronium atoms before their decay. It was done accepting the com-

Characteristics of positronium atoms in the samples of tektite and hyalite.

Sample	τ_3(ns)	I'_3 (%)	Γ_1 (mrad)	I_1 (%)
Tektite	1.28 ± 0.04	5.4 ± 0.2	3.5 ± 0.1	4.3 ± 0.1
Hyalite	1.61 ± 0.01	26.3 ± 0.4	3.3 ± 0.1	7.1 ± 0.1

monly used simple models of a pore according to which the pore constitutes for positronium a square potential well of spherical symmetry and assuming that the positronium trapped in the well occupies the ground state before its decay. The proper computations were performed for the well without electrons inside, having the potential barrier of a finite height [9] (the first case) and for the well with a layer of electron cloud inside, located at the surface of the well barrier of an infinite height [2] (the second case). As a result the depth U (the first case) and the radius R of the wells were determined in the studied samples. In the calculations we used the relationships given in the literature. In the first case they were taken from the publication of Bartenev et al. [9]

$$R_U = x\,\frac{2\hbar}{m_0 c\Gamma_1},\qquad\qquad (1)$$

$$U = \frac{m_0 c^2 \Gamma_1^2}{16\sin^2(x)},\qquad\qquad (2)$$

where x is a solution of the following equation:

$$\frac{\tau_0}{\tau_3} = \frac{\sin^3(x)}{\sin(x) + x[1 - \sin^2(x)]^{1/2}}\qquad\qquad (3)$$

under the condition $\mathrm{ctg}\,x < 0$, $\hbar$ is the Planck's constant divided by 2π, m_0 denotes the electron mass, c stands for the velocity of light and $\tau_0 = 350$ ps. In the second case the used formulas were taken from the paper of Jean [2]

$$\tau_3 = \frac{1}{2}\left[1 - \frac{R_\infty^\tau - \Delta R}{R_\infty^\tau} + \frac{1}{2\pi}\sin(2\pi\frac{R_\infty^\tau - \Delta R}{R_\infty^\tau})\right]^{-1},\qquad\qquad (4)$$

$$R_\infty^\Gamma = 2.148\frac{2\hbar}{m_0 c\Gamma_1},\qquad\qquad (5)$$

where $\Delta R = 165.6$ pm is an empirical parameter describing the thickness of a layer of electron cloud located inside the well at the surface of its barrier and τ_3 is expressed in the units of ns. As it is seen from the above equations, in the second case the radius R of the positronium traps can be determined independently from AD (R_∞^Γ) and LS (R_∞^τ) data whereas in the first case determining of $R(R_U)$ requires using both AD and LS parameters simultaneously.

The obtained results are collected in Table II.

Parameters of positronium traps in the samples of tektite and hyalite.

Sample	R_U (pm)	U (eV)	R_∞^τ (pm)	R_∞^Γ (pm)
Tektite	486 ± 14	0.56 ± 0.03	373 ± 5	481 ± 14
Hyalite	524 ± 11	0.57 ± 0.02	411 ± 1	502 ± 11

3. Conclusions

From the obtained data it follows that there are empty pores inside the studied materials which trap positronium atoms before their decay. The mean size of the pores as well as the total free space in tektite are smaller than in hyalite. The values of the mean radius of the pores "seen" by the positronium atoms amount to 0.4–0.5 nm and depend on the method of their estimation. The values of R_∞^Γ and R_U are closed to 0.5 nm whereas the values of R_∞^τ are closed to 0.4 nm. On the other hand the difference in R for the pores of hyalite and tektite is about 40 pm when estimated on the basis of R_∞^τ and R_U and it is about 20 pm when estimated from R_∞^Γ. A question arises why the values of R_∞^τ are essentially smaller than the ones of R_∞^Γ. The difference $R_\infty^\Gamma - R_\infty^\tau$ for tektite and hyalite is about 100 pm and exceeds 20% of R_∞^τ. This is strange in view of the results of Jean [10] who obtained an excellent agreement between values of R_∞^τ and R_∞^Γ for an epoxy polymer studied in various temperatures. It is worth noticing that the unexpected discrepancy between the values of R_∞^τ and R_∞^Γ for studied materials can be decreased by using in Eq. (4) the empirical constant ΔR with greater value. In our computation we have accepted, following [2], that ΔR is 165.6 pm. This value of ΔR was determined on the basis of Eq. (4) and the observed values of τ_3 for molecular solids with the known pore sizes. If it is assumed that $R_\infty^\tau = R_\infty^\Gamma$ one can obtain that ΔR is equal to 214 pm and 202 pm for tektite and hyalite, respectively. It may suggest that the thickness ΔR of the electron layer inside the model pore is different for dissimilar materials (pores). It seems however that drawing the final conclusions on the question requires further experimental data of the kind presented in this paper. As a final point we would like to return to the mentioned paper of Jean [10] on the correlation between R_∞^τ and R_∞^Γ values. From the paper it follows that the AD data for an epoxy polymer were elaborated with the program ACARFIT (from the PATFIT 88 package [11]) which does not take the instrumental resolution into account. It means that the Γ values published by Jean are overestimated and the true values of R_∞^Γ are greater than the R_∞^τ ones as in our studies.

Acknowledgments

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