Vol. **107** (2005) *ACTA PHYSICA POLONICA A* No. 4

Proceedings of the 35th Polish Seminar on Positron Annihilation, Turawa, Poland 2004

Role of Trapped and Solvated Electrons in Ps Formation

S.V. Stepanov[a,*], V.M. Byakov[a], C. He[b], T. Hirade[c]
and K.V. Mikhin[a]

[a]Institute for Theoretical and Experimental Physics, 117218 Moscow, Russia
[b]Photonics Research Institute, AIST
Umezono 1-1-1, Tsukuba, Ibaraki 305-8568, Japan
[c]Japan Atomic Energy Research Institute, 319-1195 Tokai, Japan

Role of trapped and solvated electrons in Ps formation is discussed. Combination of thermalized positron with such electrons is possible from the viewpoint of the energy balance and may results in Ps formation. This process proceeds during all e^+ lifetime in matter. Fitting of raw experimental e^+–e^- annihilation spectra has to be based on an adequate physical input, which often leads to necessity of nonexponential deconvolution of the spectra. We have interpreted the Ps formation data in polyethylene, ethylene-methylmethacrylate and polymethylmethacrylate in dark and in light vs. time of the measurement and temperature. Parameters characterized accumulation of trapped electrons and their recombination with counter ions and positrons are obtained.

PACS numbers: 61.80.Fe, 36.10.Dr, 78.70.Bj, 82.35.Lr

1. Introduction

According to modern views [1, 2] formation of positronium (Ps) atom in dielectric media proceeds as follows. Fast positrons, born, for example, as a result of β^+-decay of ^{22}Na, during a fraction of a picosecond lose their kinetic energy on ionization of a medium. Some of the thermalized positrons may combine with track electrons, created in e^+ ionization slowing down and form Ps atoms. Ps formation reaction, $e^+ + e^- \to$ Ps, takes place at the terminal part of the positron track just after thermalization of e^+ and e^-. In Ref. [3] basing on the temperature dependence of the Ps yield in n-propanol there was concluded that the main Ps

---

[*] corresponding author; e-mail: Sergey.Stepanov@itep.ru

precursor is intratrack quasi-free electron, e^-_{qf}. However, the question about the contribution of the solvated species in the Ps formation remains open.

Recently the possibility of Ps formation in polymers at low temperatures as a result of interaction of e^+ with localized electrons was experimentally proved [4]. Later on more detailed investigations were carried out in different polymer matrices in a wide temperature range [5, 6]. It was shown that an important factor, which determines the Ps yield, is an influence of ionizing irradiation of the samples caused by e^+ source. Electrons formed as a result of ionization of the medium and escaped ion–electron recombination at low temperature (below glass transition point), are stabilized on structural defects and survived for a long time. These electrons are called trapped electrons, e^-_t. Usually their concentration increases with time t_m of the measurement. An increment of the Ps formation is attributed to the reaction $e^+ + e^-_t \rightarrow \mathrm{Ps}$. Direct confirmation of realization of this reaction was obtained in age momentum correlation (AMOC) studies [7].

Illumination of the sample by visible light ("bleaching") delocalizes trapped electrons and stimulates their recombination with counter ions. In this case this additional channel of Ps formation disappears and Ps yield drops down. A similar effect is achieved by means of heating of the sample (above glass transition temperature). As an example, *o*-Ps formation probability measured in low density polyethylene (LDPE) in dark and in light is shown in Fig. 1.

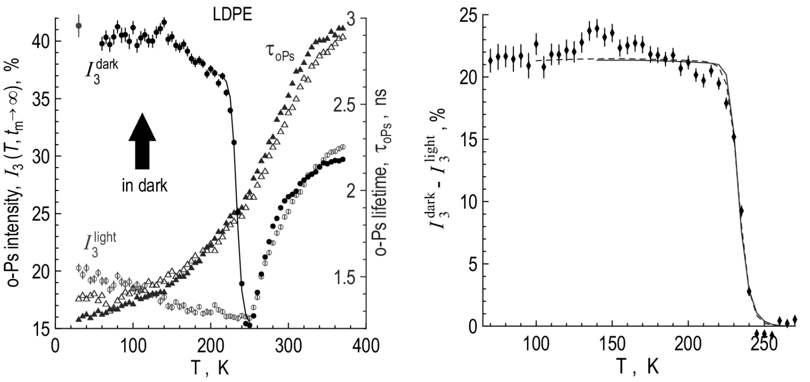

Fig. 1. Left: Temperature dependences of *o*-Ps formation probability (○, ●) and *o*-Ps lifetime (Δ, ▲) in low density polyethylene in dark (filled symbols) and in light (open symbols) [5]. The data are obtained in equilibrium conditions ($t_m \rightarrow \infty$). Each point was measured during 1 h and after that T was increased in a step of 5 K. So e^-_t concentration reaches its maximum equilibrium value at any given T. Right: The difference of I_3 in dark and in light vs. T (◆). It simulates the contribution of the trapped electrons to the Ps formation. Lines are drown basing on Eqs. (10–13). Solid line corresponds to the 2nd order Eq. (1) and dashed line reflects accumulation of the e^-_t according to the 1st order Eq. (2).

2. Accumulation of trapped electrons and radicals

A trapped electron disappears only if it recombines with a positive ion.[†] The question about an order of a chemical reaction, which governs accumulation kinetics of e_t^-, is related to the correlation in positions of e_t^- and positive counter ions. These species are formed (in spurs mainly) because of ionizing radiation from the positron source. Most of them recombine, but some fraction escapes intratrack ion–electron recombination. Afterwards these electrons transform into e_t^-.

Recombination between e_t^- and counter ions takes place in case of occurrence of two uncoupled events: (1) presence of the counter ions close enough to precursors of e_t^- (which is mobile therefore this mobility will affect the maximum concentration of e_t^-) until trapping and (2) activation of e_t^- by molecular motion or appearance of a good chance for e_t^- to tunnel to the ion, which governs by temperature-dependent (in general case) rate constant k_ti. Multiplication of probabilities of these two events gives a standard recombination term, $-k_\mathrm{ti}c_\mathrm{t}c_\mathrm{i}$, which transforms into $-k_\mathrm{ti}c_\mathrm{t}^2$, because the concentration of electrons equals the concentration of the ions ($c_\mathrm{t} = c_\mathrm{i}$) at any time. So the concentration of e_t^- obeys the following equation:

$$\frac{\mathrm{d}c_\mathrm{t}}{\mathrm{d}t_\mathrm{m}} = J - k_\mathrm{ti}c^2{}_\mathrm{t}, \qquad c_\mathrm{t}(0) = 0 \quad \Rightarrow \quad c_\mathrm{t}(t_\mathrm{m}) = \sqrt{\frac{J}{k_\mathrm{ti}}\frac{\mathrm{e}^{2\sqrt{Jk_\mathrm{ti}}t_\mathrm{m}} - 1}{\mathrm{e}^{2\sqrt{Jk_\mathrm{ti}}t_\mathrm{m}} + 1}}. \tag{1}$$

Here t_m is the duration of the measurement, k_ti is the recombination rate constant and J describes e_t^- production. Indeed, the tunneling recombination of ion pairs with short distance will be very fast and they may not be detected by EPR or positrons. Moreover, the de-trapped electrons have a certain possibility of re--trapping. In this case recombination term will look more complicate and may depend on c_t.

The first order equation

$$\frac{\mathrm{d}c_\mathrm{t}}{\mathrm{d}t_\mathrm{m}} = J - \frac{c_\mathrm{t}}{\tau_\mathrm{t}}, \qquad c_\mathrm{t}(0) = 0 \quad \Rightarrow \quad c_\mathrm{t}(t_\mathrm{m}) = J\tau_\mathrm{t}(1 - \mathrm{e}^{-t_\mathrm{m}/\tau_\mathrm{t}}) \tag{2}$$

could be valid in the case when it is sufficient just to activate the electron and recombination definitely takes place. If $J\tau_\mathrm{t}^2 \sim 1/k_\mathrm{ti}$ Eqs. (1) and (2) predict a similar time variation of $c_\mathrm{t}(t_\mathrm{m})$.

Longtime irradiation of the sample may lead to accumulation of radicals in the bulk (similar to that of e_t^-). They may capture intratrack e^- and e^+ and, hence, suppress Ps yield. Accumulation of radicals in LDPE and their ability to capture Ps precursors are clearly seen in Fig. 1 when e_t^- are removed by bleaching [5]. Above glass transition temperature e_t^- concentration is negligible, but radicals still remain, but in a smaller amount. However, investigation of this effect requires a more elaborated model.

[†] For the sake of simplicity we neglect here possible reactions with radicals $\mathrm{e}_\mathrm{t}^- + \mathrm{R} \rightarrow \mathrm{R}^-$, host molecules (or their fragments) $\mathrm{e}_\mathrm{t}^- + \mathrm{M} \rightarrow \mathrm{M}^-$ and so on.

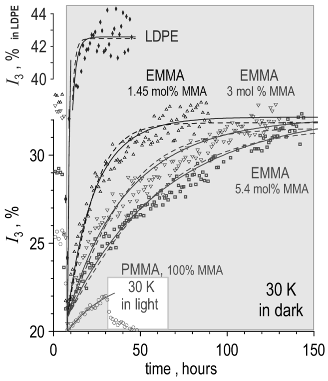

Fig. 2. Time dependences of I_3 in LDPE, EMMA, and PMMA in dark after rapid cooling of the samples from room temperature down to 30 K [10]. It is also shown the result of bleaching of PMMA sample. Solid lines are the result of fitting of the data using Eqs. (10) and (2). Dashed lines are drown basing on Eqs. (10) and (1).

It seems that in polymethylmethacrylate (PMMA) and ethylene-methylmethacrylate (EMMA) accumulation of such radicals does not proceed so efficiently as in LDPE (Fig. 2). When the light is switched on, *o*-Ps intensity in PMMA drops down exactly to the initial value, $I_3(t_\mathrm{m} \to 0)$ (contrary to the case of LDPE).

3. A model of Ps formation in polymers: account for e_t^-

To describe Ps formation process we have to characterize the behavior of e^+ and e^-. Using coincidence Doppler broadening technique it was demonstrated that the carbonyl group (>C=O, entering MMA-group) efficiently traps thermalized positrons [8]:

$$\mathrm{e}^+ + \mathrm{CO} \to \mathrm{COe}^+. \tag{3}$$

So we adopt that if the mole fraction of the MMA-group is larger than 1%, all the positrons are undergone reaction (3). However, e^+ is not strongly bound in COe^+ state and may form Ps, reacting with intrablob or trapped e^-, if it resides close enough. This statement follows from the fact that in all investigated EMMA and PMMA samples initial (at $t_\mathrm{m} \to 0$) *o*-Ps yield is practically the same ($\approx 20\%$) and does not depend on the concentration of the MMA-groups.

We assume that accumulation of e_t^- takes place uniformly in the bulk of the sample and can be described either by Eq. (1) or Eq. (2).

Unfortunately, we have to ignore completely kinetics of intrablob processes (ion–electron recombination, solvation, out-of blob diffusion and so on), because their account leads to necessity of non-exponential deconvolution of raw experimental lifetime spectra, which is not done yet. So we are forced to replace the time-dependent concentration of intrablob electrons around the positron by its averaged over free e$^+$ lifetime. With this rough assumption we come to the following equation for e$^+$ distribution:

$$\frac{\partial c_{\rm p}}{\partial t} = D_{\rm p}\Delta c_{\rm p} - (W_{\rm bl} + W_{\rm t} + \lambda_{\rm p})c_{\rm p}, \qquad c_{\rm p}(r,0) = \frac{\exp(-r^2/a_{\rm p}{}^2)}{\pi^{3/2}a_{\rm p}^3}. \tag{4}$$

Diffusion term, $D_{\rm p}\Delta c_{\rm p}(r,t)$, may be neglected because we assume that the positrons are immediately (at $t \approx 0$) captured by CO-groups, so $D_{\rm p} \to 0$. $\lambda_{\rm p}$ is the e$^+$ annihilation rate of e$^+$ in this state. $a_{\rm p}$ characterizes initial distribution of the thermalized e$^+$. $W_{\rm bl}(t_{\rm m}) \approx k_{\rm ep}c_e + k_{{\rm p}R}c_R(t_{\rm m})$ is the rate of disappearance of e$^+$ owing to reactions with intrablob species (including Ps formation and possible capture of e$^+$ by radicals; c_R is their concentration). $W_{\rm t} = k_{\rm tp}c_{\rm t}(t_{\rm m})$ is the Ps formation rate with a trapped electron. Integrating Eq. (4) over the sample, one obtains the equation for e$^+$ survival probability as a function of t and $t_{\rm m}$:

$$\dot n_{\rm p} = -W_{\rm p}n_{\rm p} \quad\Rightarrow\quad n_{\rm p} = {\rm e}^{-W_{\rm p}(t_{\rm m})t}, \qquad W_{\rm p} = W_{\rm bl}(t_{\rm m}) + W_{\rm t}(t_{\rm m}) + \lambda_{\rm p}. \tag{5}$$

One may write equations for the formation probabilities of p-Ps and o-Ps:

$$\dot n_{p{\rm Ps}} = \frac{W_{\rm bl} + W_{\rm t}}{4}n_{\rm p} - \lambda_{p{\rm Ps}}n_{p{\rm Ps}}, \qquad n_{p{\rm Ps}}(0) = 0, \tag{6}$$

$$\dot n_{o{\rm Ps}} = \frac{3(W_{\rm bl} + W_{\rm t})}{4}n_{\rm p} - \lambda_{o{\rm Ps}}n_{o{\rm Ps}}, \qquad n_{o{\rm Ps}}(0) = 0, \tag{7}$$

which can be easily solved using Eq. (5). Finally we may obtain the number $\Delta C(t)$ of e$^+$–e$^-$ annihilations in the channel of the time analyzer (corresponding to the time moment t):

$$\begin{aligned}\frac{\Delta C(t)}{\Delta t} &= C_{\rm tot}(\lambda_{p{\rm Ps}}n_{p{\rm Ps}} + \lambda_{\rm p}n_{\rm p} + \lambda_{o{\rm Ps}}n_{o{\rm Ps}})\\ &= C_{\rm tot}\Big[\frac{\lambda_{p{\rm Ps}}(W_{\rm bl}+W_{\rm t})}{4(\lambda_{p{\rm Ps}} - W_{\rm p})}\left({\rm e}^{-W_{\rm p}t} - {\rm e}^{-\lambda_{p{\rm Ps}}t}\right)\\ &\qquad + \lambda_{\rm p}{\rm e}^{-W_{\rm p}t} + \frac{3\lambda_{o{\rm Ps}}(W_{\rm bl}+W_{\rm t})}{4(W_{\rm p} - \lambda_{o{\rm Ps}})}\left({\rm e}^{-\lambda_{o{\rm Ps}}t} - {\rm e}^{-W_{\rm p}t}\right)\Big].\end{aligned} \tag{8}$$

This equation does not contain background contribution from random coincidences and it is not convoluted with the resolution function. Equation (8) is normalized on the total number $C_{\rm tot}$ of counts: $\int_0^\infty \frac{\Delta C(t)}{\Delta t}{\rm d}t = C_{\rm tot}$. Equation (8) is similar to the conventional 3-exponential representation of the lifetime spectrum

$$\frac{\Delta C(t)}{\Delta t} = C_{\rm tot}\left(\frac{I_1}{\tau_1}{\rm e}^{-t/\tau_1} + \frac{I_2}{\tau_2}{\rm e}^{-t/\tau_2} + \frac{I_3}{\tau_3}{\rm e}^{-t/\tau_3}\right), \tag{9}$$

where intensities $I_{\rm i}$ and lifetimes $\tau_{\rm i}$ are considered as free adjustable parameters with the only constrain: $I_1 + I_2 + I_3 = 1$. So the total number of the fitting parameters in Eq. (9) is five. Equation (8) also contains five parameters: $W_{\rm bl}$, $W_{\rm t}$, $\lambda_{p{\rm Ps}} \equiv 1/\tau_1$, $\lambda_{\rm p}$, and $\lambda_{o{\rm Ps}} \equiv 1/\tau_3$. As for the rest, Eq. (9) has no physical motivation and it completely ignores relationships between preexponential factors and lifetimes predicted even by naive Eq. (8). So it is difficult to attribute any physical meaning to the parameters involved in Eq. (9).

For example, from Eq. (8) it follows that $I_1 = -(W_{\rm bl} + W_{\rm t})/4(\lambda_{p\rm Ps} - W_{\rm bl} - W_{\rm t} - \lambda_{\rm p})$. It could be even negative! The meaning of $1/\tau_2$ is not the annihilation rate of free e^+ on molecular electrons as it is usually believed (in our notation e^+ annihilation rate is $\lambda_{\rm p}$). Actually $1/\tau_2$ should be associated with $W_{\rm p}$, the total decay rate of e^+, which depends on $c_{\rm t}$, $t_{\rm m}$, and T.

According to Eq. (9), a coefficient before $\lambda_{o\rm Ps} e^{-\lambda_{o\rm Ps} t}$ in Eq. (8), is I_3:

$$I_3(t_{\rm m}) = \frac{3}{4} \frac{W_{\rm bl}(t_{\rm m}) + W_{\rm t}(t_{\rm m})}{W_{\rm bl}(t_{\rm m}) + W_{\rm t}(t_{\rm m}) + \lambda_{\rm p} - \lambda_{o\rm Ps}}. \qquad (10)$$

It is $I_3^{\rm dark}(t_{\rm m})$, if the measurements are conducted in dark ($W_{\rm t}(t_{\rm m}) \neq 0$), in light when $W_{\rm t} = 0$ (all $e_{\rm t}^-$ are bleached) it gives

$$I_3^{\rm light}(t_{\rm m}) = \frac{3}{4} \frac{W_{\rm bl}(t_{\rm m})}{W_{\rm bl}(t_{\rm m}) + \lambda_{\rm p} - \lambda_{o\rm Ps}}. \qquad (11)$$

Thus, we do not consider $W_{\rm bl}$ as an adjusted parameter, but calculate it from Eq. (11) using experimental data on $I_3^{\rm light}(t_{\rm m})$ in light.

Because the experimental data in Figs. 1, 2 were obtained using Eq. (9) we have no other possibility as to use Eq. (10) for fitting the $I_3^{\rm dark}(t_{\rm m})$ data shown

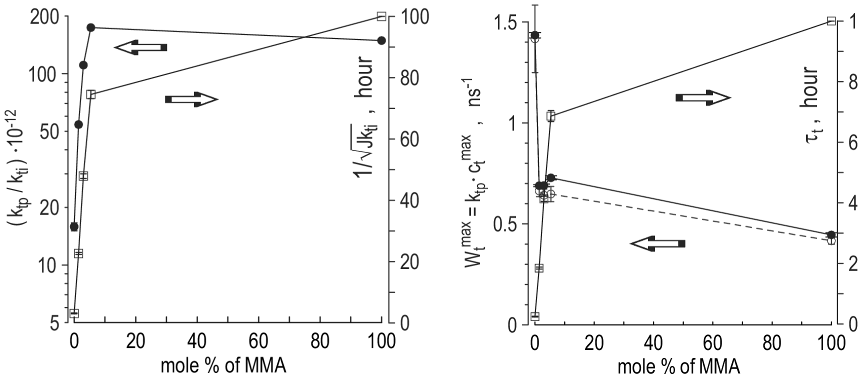

Fig. 3. Concentration dependences of the parameters, which determine Ps formation. Using Eq. (1) we obtain $k_{\rm tp}/k_{\rm ti}$ ratio, ●, and $\sqrt{Jk_{\rm ti}}$, □ (left side). If we use Eq. (2) (right side), we obtain $\tau_{\rm t}$ (□) and the product $W_{\rm t}^{\rm max} = k_{\rm tp} J \tau_{\rm t} = k_{\rm tp} c_{\rm t}^{\rm max}$, ●, which is the maximal recombination rate of e^+ and $e_{\rm t}^-$. Open circles show $W_{\rm t}^{\rm max} = k_{\rm tp}\sqrt{J/k_{\rm ti}} = k_{\rm tp} c_{\rm t}^{\rm max}$ in case of usage of Eq. (1). Both dependences of $W_{\rm t}^{\rm max}$ vs. MMA content (shown by ● and ○) coincide within experimental uncertainty. Because of a lack of data on $I_3^{\rm dark}(t_{\rm m})$ in case of PMMA, the values of $\sqrt{Jk_{\rm ti}}$ (in Eq. (1)) and $\tau_{\rm t}$ (in Eq. (2)) were arbitrary fixed to 100 h.

in Fig. 2. At a given $t_{\rm m}$ and temperature $W_{\rm t}$ depends only on two parameters. If Eq. (1) is used, they are dimensionless ratio of the rate constants $k_{\rm tp}/k_{\rm ti}$ and $(Jk_{\rm ti})^{-1/2}$, which has a meaning of the $\rm e_t^-$ lifetime. In case of Eq. (2), they are $Jk_{\rm tp}$ and $\tau_{\rm t}$. These parameters are obtained from the fitting of the experimental data. Results presented in Fig. 3. We have also used the following numbers for $I_3^{\rm light}(t_{\rm m}=0)$, $\lambda_{\rm p}$, and $\lambda_{o{\rm Ps}} = 1/\tau_3$ at $T = 30$ K gathered in the Table [9].

TABLE

Values of parameters used in calculations.

Sample	$I_3^{\rm light}(t_{\rm m}=0)$, %	$\lambda_{\rm p}^{-1} \approx \tau_2$, ns	$\lambda_{o{\rm Ps}}^{-1}$, ns
LDPE	20	0.44	1.31
all EMMA	20.8	0.37–0.38	1.31–1.35
PMMA	20	0.44	1.68

It is interesting that taking typical numbers for equilibrium concentration of trapped electrons in polyethylene ($c_{\rm t}^{\rm max} \approx (4-5)\times 10^{15}$ $e_{\rm t}^-$/g) and PMMA ($c_{\rm t}^{\rm max} \approx 10^{17}$ $e_{\rm t}^-$/g) known from independent EPR-measurements [4], from the data shown in Fig. 3 we immediately obtain quite reasonable values of $k_{\rm tp}$: 1.5×10^{14} $\rm M^{-1}s^{-1}$ in PE and 6×10^{12} $\rm M^{-1}s^{-1}$ in PMMA.

4. Temperature dependence of I_3 in LDPE

Basing on the results obtained above, we can consider temperature variation of I_3 in polyethylene (Fig. 1). As before, we adopt that the difference of I_3 in dark and in light is due to the contribution of the trapped electrons. When T increases (close to glass-transition point) various molecular motions in the sample come into play. They disentangle trapped electrons and stimulate their recombination with counter ions. So $\rm e_t^-$ concentration decreases rapidly. Experimental data shown in Fig. 1 were obtained in equilibrium conditions (each point in was measured during 1 h and after that T was increased on 5 K). So $\rm e_t^-$ concentration reached its maximum equilibrium value at any given temperature. It implies that

$$W_{\rm t}(t_{\rm m}) = k_{\rm tp}c_{\rm t}(t_{\rm m}\to\infty) = \begin{cases} k_{\rm tp}\sqrt{J/k_{\rm ti}}, & \text{in case of Eq. (1),} \\ k_{\rm tp}J\tau_{\rm t}, & \text{in case of Eq. (2).} \end{cases} \tag{12}$$

We shall take into account an influence of temperature on the equilibrium concentration of $\rm e_t^-$ in Eq. (12) by adding a simple Arrhenius-like term to the recombination constant $k_{\rm ti}^{30\ \rm K}$ (or to recombination rate $1/\tau_{\rm t}^{30\ \rm K}$) of $\rm e_t^-$ with counter-ion at 30 K[‡]:

$$k_{\rm ti}(T) \approx k_{\rm ti}^{30\ \rm K}(1 + {\rm e}^{E/\tilde{T}_{\rm g} - E/T})$$

[‡] For simplicity we relate here all temperature variation of I_3 to $k_{\rm ti}$ variation only. One may expect that temperature dependences of J and $k_{\rm tp}$ are smaller than that of $k_{\rm ti}$.

or

$$\frac{1}{\tau_{\mathrm{t}}(T)} = \frac{1}{\tau_{\mathrm{t}}^{30\ \mathrm{K}}}(1 + \mathrm{e}^{E/\tilde{T}_{\mathrm{g}} - E/T}) \tag{13}$$

(here T and $\tilde{T}_{\mathrm{g}}$ is in energy units). Hence, we obtain a new pair of adjustable parameters: the activation (detrapping) energy, E, and $\tilde{T}_{\mathrm{g}}$. Note, that $\tilde{T}_{\mathrm{g}}$ should not be identified with the glass transition temperature, it is just the temperature when the Arrhenius exponent becomes equal to unity. Curves in Fig. 1 (right side) represent the result of the fitting based on Eqs. (12, 13). Usage of the 2nd order kinetics gives $E = 2.1(1)$ eV and $\tilde{T}_{\mathrm{g}} = 229(1)$ K, but the 1st order one gives $E = 1.32(6)$ eV and $\tilde{T}_{\mathrm{g}} = 232(1)$ K.

5. Results and discussion

A quite often experimental treatment of the lifetime spectra reduces to their exponential decomposition and interpretation of obtained intensities as formation probabilities of respective positron states (p-Ps, free e^+, and o-Ps). It requires crude assumptions about kinetics of intratrack processes including Ps formation, namely we have to suppose that the Ps formation duration is negligibly small. As a result, the physical meaning of the parameters obtained is rather questionable.

An account for the decay kinetics of the intratrack electrons (even in the simplest, for example, exponential form) immediately results in a complicate (non-exponential) shape of the lifetime spectrum, which can be resolved basing on a proper physical model of the processes [10]. Of course, it elongates deconvolution time of each spectrum, but with the help of modern computers it is not a limiting factor, which may terminate the whole investigation.

In this paper we have paid "high price" for obtaining three-exponential lifetime spectrum: all kinetics related to blob electrons was ignored. However preexponential factors turn out to be related to each other. These constrains had to be explicitly taken into account *during* deconvolution of the spectra. Unfortunately, even this simple strategy has not been realized yet. We have restricted ourselves to interpretation of the temporal, temperature and concentration variations of the I_3 parameter using our Eqs. (10, 11). Fitting data for LDPE, EMMA, and PMMA, we have come to the following conclusions:

- at 30 K the reaction rate constant k_{tp} is 13 orders as large as the recombination rate constant k_{ti}. Estimated absolute values of k_{tp} for PE ($\sim 2 \times 10^{14}$ $\mathrm{M}^{-1}\mathrm{s}^{-1}$) and PMMA ($\sim 10^{13}$ $\mathrm{M}^{-1}\mathrm{s}^{-1}$) seem quite reasonable;
- concentration of $\mathrm{e}_{\mathrm{t}}^-$ reaches its equilibrium value within a characteristic time, which varies from 3 h in LDPE to 70 h in EMMA samples (probably even longer in PMMA). It strongly grows up at a low (about 1 mole%) MMA concentration;
- temperature dependence of I_3 is interpreted under the assumption that the recombination rate constant of $\mathrm{e}_{\mathrm{t}}^-$ with a positive ion is the sum of temper-

ature independent and Arrhenius terms. Activation energy of e_t^- lies within 1–2 eV. Its particular number depends on the order of chemical equation applied for the description of accumulation kinetics of the trapped electrons;

- our model is in agreement with recent data [11] indicating that in PE I_3 does not decrease with an increase of external electric field neither at 150 K, nor at 250 K. We attribute this observation to e^+ trapping by radicals produced as a result of irradiation by the positron source. At higher temperatures radicals disappear because of R+R-recombination, e^+ mobility grows up and E-field effect becomes visible.

References

[1] O.E. Mogensen, *Positron Annihilation in Chemistry*, Springer-Verlag, Berlin 1995.

[2] S.V. Stepanov, V.M. Byakov, in: *Principles and Applications of Positron and Positronium Chemistry.* Eds. Y.C. Jean, P.E. Mallone, D.M. Schrader, World Scientific Publ., Singapore 2003, p. 117.

[3] V.M. Byakov, V.I. Grafutin, O.V. Koldaeva, *Radiat. Phys. Chem.* **10**, 239 (1977).

[4] T. Hirade, F.H.J. Maurer, M. Eldrup, *Radiat. Phys. Chem.* **58**, 465 (2000).

[5] C. He, T. Suzuki, V.P. Shantarovich, L. Ma, M. Matsuo, K. Kondo, Y. Ito, *Phys. Lett. A* **313**, 223 (2003).

[6] T. Hirade, *Radiat. Phys. Chem.* **68**, 375 (2003).

[7] N. Suzuki, T. Hirade, F. Saito, T. Hyodo, *Radiat. Phys. Chem.* **68**, 647 (2003).

[8] T. Suzuki, C. He, V. Shantarovich, K. Kondo, E. Hamada, M. Matsuo, L. Ma, Y. Ito, *Radiat. Phys. Chem.* **66**, 161 (2003).

[9] C. He, T. Suzuki, L. Ma, M. Matsuo, V.P. Shantarovich, K. Kondo, Y. Ito, *Phys. Lett. A* **304**, 49 (2002).

[10] C. Dauwe, B. Van Waeyenberge, N. Balcaen, *Phys. Rev. B* **68**, 132202 (2003).

[11] Y. Ito, T. Suzuki, *Radiat. Phys. Chem.* **66**, 343 (2003).