

## Influence of Atmospheric Gases Present in the Pores of MCM-41 on Lifetime of ortho-Positronium

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**Abstract.** The modification of the extended Tao-Eldrup model accounting ortho-positronium quenching in air is presented. Taking into account quenching by oxygen molecules adsorbed on the surface of porous material gives reasonable agreement between results of the model and the experimental positron annihilation lifetime spectroscopy data. Pore size distributions calculated using this model from the spectra for MCM-41 mesoporous sieve obtained in air, oxygen or vacuum are compared and discussed taking into account effect of ortho-positronium migration from small open pores to the larger ones. The rates of ortho-positronium quenching by air ( $47.2 \mu\text{s}^{-1} \text{MPa}^{-1}$ ), oxygen ( $220 \mu\text{s}^{-1} \text{MPa}^{-1}$ ) or nitrogen ( $1.7 \mu\text{s}^{-1} \text{MPa}^{-1}$ ) obtained from pressure dependences of the lifetimes observed in MCM-41 agree reasonably with the experimental results of other authors, if the correction for oxygen adsorbed on the surface is applied.

### Introduction

Since silica gels are available for study, interaction of ortho-positronium (o-Ps) trapped in their pores with gases influencing o-Ps lifetime is object of interest [1-2]. Explanation of quenching of Ps by interaction with paramagnetic molecules (like  $\text{O}_2$ ) involving their excited complex with Ps was proposed as early as in 1970's ties. However, these studies do not provide practical information about correction which has to be made for results obtained in air. There are experiments that need to be carried in air, e.g. [3], therefore the relation between pore size and o-Ps lifetime in air, analogous to the one given by the Extended Tao-Eldrup (ETE) model [4] for vacuum, seems to be useful. Such relation was calculated from classical model (where Ps is treated as a bouncing ball) taking into account presence of air on the surface of the material [5]. Predictions of the classical model (Fig.1) agree with the experimental data for pore diameters larger than 2 nm. Nevertheless, some assumptions used in that model seem to be incorrect. The relation of o-Ps lifetime in vacuum versus pore size obtained from classical bouncing ball approach was shown to be inaccurate for pores with diameters smaller than 20 nm [6] and there is no reason to assume that classical approach would be more accurate in the presence of air. Another doubtful assumption is the existence of completely filled monolayer of air molecules on the surface of pore at normal temperature and pressure. According to the recent gravimetric studies of gas adsorption on MCM-41 porous silica [7], the layer of air adsorbed on the surface should be filled in less than 0.5%. In order to obtain correct relation between o-Ps lifetime in air and pore diameter a modification of quantum model is needed.

### Materials and Experimental

Mesoporous molecular sieves MCM-41 were synthesized [8] using hexadecyltrimethylammonium bromide (99%, Fluka) cationic surfactant as a template and tetraethoxysilane (98%, Fluka) as a silica source. After synthesis the template was removed by heating up (rate 1 K/min) to 823 K in air for 8 h. Then the sample was stored at 823 K for 5 h in an oxygen flow in order to eliminate the carbon remnants left. Specific surface  $1209 \text{ m}^2/\text{g}$  (BET method), pore volume  $1.17 \text{ cm}^3/\text{g}$  and mean diameter of primary pores 2.8 nm (BJH method) of the obtained material were estimated from nitrogen adsorption isotherm at 77K.

Fast-slow delayed coincidence spectrometer with the detectors equipped with BaF<sub>2</sub> scintillators was used for positron annihilation lifetime spectroscopy (PALS). ‘Stop’ energy window was widely open in order to maximize the efficiency of collecting 3- $\gamma$  annihilation events, what makes a relatively poor time resolution (FWHM  $\sim$  310 ps). The sample in form of two 2 mm thick layers of powder surrounded <sup>22</sup>Na positron source enclosed in a Kapton envelope. During the measurements the sample was kept inside the sealed chamber, where the vacuum about 10<sup>-5</sup> Pa could be maintained or it could be filled by gas under selected pressure up to ambient pressure. Ambient air, high purity nitrogen (99.999%, BOC Gazy) or oxygen (99.5%, Air products) were used to fill the chamber. The coincidence counting rate was about 2.4  $\times$  10<sup>6</sup> per hour. About 3.1  $\times$  10<sup>7</sup> counts per spectrum were collected during 13 hours for MELT analysis [9] and 1.2  $\times$  10<sup>7</sup> counts during 5 hours for LT analysis [10]. All measurements were done at 300 K.

### Quantum model for air

Shapes of the dependence of o-Ps lifetime in pores of silica materials in air versus pore diameter (Fig.1) obtained from experiment [1, 3, 11] and the analogous dependence in vacuum [6] seem to be similar, except for the fact that maximum value of the lifetime observed in air reaches only 85-90 ns. This value is observed for large pores, where pick-off annihilation is negligible and the

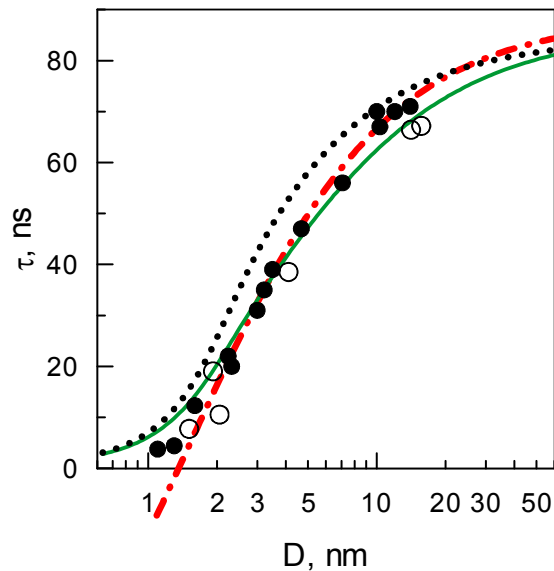


Fig.1 Relation between o-Ps lifetime in air and diameter of free volume: experimental data by Goldanskii's group [1, 11] (solid circles) and Zaleski [3] (open circles), the ETE model for air with adsorption (solid line), the ETE model for air without adsorption (dotted line), classical approximation [5] (dash-dot line).

lifetime is determined mainly by oxygen quenching and intrinsic annihilation. These results led Dull et al. [12] to the conclusion that constant value of air quenching rate at standard pressure  $\lambda_{\text{air}(1\text{atm})} = 4 \mu\text{s}^{-1}$  can be applied to the quantum ETE model, regardless of the pore size, according to equation

$$\lambda = \lambda_{\text{ETE}} + \lambda_{\text{air}(1\text{atm})}, \quad (1)$$

where  $\lambda_{\text{ETE}}$  is o-Ps annihilation rate calculated according to the ETE model [4]. The value of  $\lambda_{\text{air}(1\text{atm})}$  given above differs significantly from 4.8-5.7  $\mu\text{s}^{-1}$  calculated from results of oxygen quenching study [1, 13-15] taking into account 21% content of oxygen in air. The reason of the deviation is adjusting air quenching rate to overestimated value of  $\lambda_{\text{ETE}}$ . Inaccurate results of the ETE model are caused by assumption of cubic pore shape made by Dull et al. [12], what is unlikely for interconnected pore system in silica gels. However, even using higher value of  $\lambda_{\text{air}(1\text{atm})} = 4.8 \mu\text{s}^{-1}$  in Eq.1 still gives poor agreement of model curve and experimental results (Fig.1).

The shortcoming of the approach presented above is absence of taking into consideration adsorption of air molecules on the surface of pores. Even 0.5% coverage of the surface by adsorbed molecules at standard pressure results in appearance of additional quenching centers. If the pores are small enough, like in the case of MCM-41, number of the molecules adsorbed on the surface exceeds number of free air molecules in the volume of the pore. Results of gravimetric studies of adsorption of main components of air, i.e. nitrogen and oxygen, on MCM-41 [7] allow to estimate the ratio between number of adsorbed and free molecules. Assuming cylindrical shape of the pore this ratio depends on pore diameter according to the relation 4.5/D nm. As a first approximation one can assume that quenching rate of air both adsorbed on the surface and free inside the pore is the same. Extending Eq. 1 by taking adsorbed gas into account results in equation

$$\lambda = \lambda_{\text{ETE}} + \lambda_{\text{gas}(1\text{atm})} + 4.5/D \lambda_{\text{gas}(1\text{atm})}, \quad (2)$$

where  $D$  is pore diameter in nm. Fitting Eq.2 to the experimental data obtained in air (Fig.1) gives  $\lambda_{\text{gas}(1\text{atm})} = 4.6 \mu\text{s}^{-1}$ . Adsorption of nitrogen and oxygen is very similar (difference in the amount of adsorbed molecules at standard pressure is about 0.7%) thus the model can be applied to both pure gases as well as their mixture in air. Following this idea an attempt to estimate pure oxygen quenching rate basing on four o-Ps lifetimes observed in MCM-41 in vacuum and in oxygen was undertaken. Obtained  $\lambda_{\text{gas}(1\text{atm})}$  value is  $17 \mu\text{s}^{-1}$ .

## Results and Discussion

**Distribution of pore sizes.** In order to compare the results of both standard ETE model and its version modified for pores filled by gas, measurements of positron lifetimes in MCM-41 were performed in vacuum, air and oxygen. The PALS spectra were analysed by MELT. From histograms of o-Ps lifetimes obtained this way the distribution of pore sizes was calculated. A part of the grid of lifetimes used in MELT (850 lifetimes from 1 ns to 140 ns in exponentially increasing steps) was transformed to the grid of pore diameters [16] according to the appropriate model

version (for vacuum, air or oxygen). The intensities ascribed by MELT to the lifetimes in the grid were transformed into volume per unit of pore size [17] according to the equation

$$\frac{dV}{dD} \propto \frac{dI(\tau)}{d\tau} \frac{d\tau}{dD}, \quad (3)$$

where  $dI(\tau)/d\tau$  is intensity from MELT divided by width of lifetime interval in the grid and  $d\tau/dD$  is the derivative of relation  $\tau(D)$  given by the model.

The results of the calculations for the range of mesopores ( $2 \text{ nm} < D < 50 \text{ nm}$ ) are shown in Fig.2. There are two groups of mesopores of different sizes, indicated by two peaks centred at about 2 nm and 16 nm, observed independent on whether sample is in vacuum or gas environment. In each spectrum there are also two peaks in the range of micropores ( $D < 2 \text{ nm}$ ) not shown in Fig.2. However, it is not clear if these micropores are open for gas penetration and therefore they were not taken into account in the discussion.

The size of smaller pores corresponds to primary cylindrical mesopores of MCM-41 (ordered in hexagonal array), while larger ones are irregular spaces between openwork particles where primary pores are located. This identification is confirmed by difference in width of the distribution of each peak. Well defined shape and size of primary pores results in small and almost symmetrical distribution of diameters ( $FWHM = 0.14 \text{ nm}$  in vacuum), while sizes of irregular interparticle spaces have wide distribution ( $FWHM = 11 \text{ nm}$  in vacuum). Usually there is a tail in the peak from the side of large pore sizes; however this effect may be result of lognormal shape of a peak preferred by MELT.

Even though general similarity between the pore distributions presented in Fig.2, differences between them are also visible. The most disturbing among the differences is discrepancy in position of the peaks. There are several possible reasons for this discrepancy:

1. Incorrect calibration (i.e. value of  $\lambda_{\text{gas}(1\text{atm})}$ ) of the model, which is highly probable because majority of available experimental data was measured 35-38 years ago.

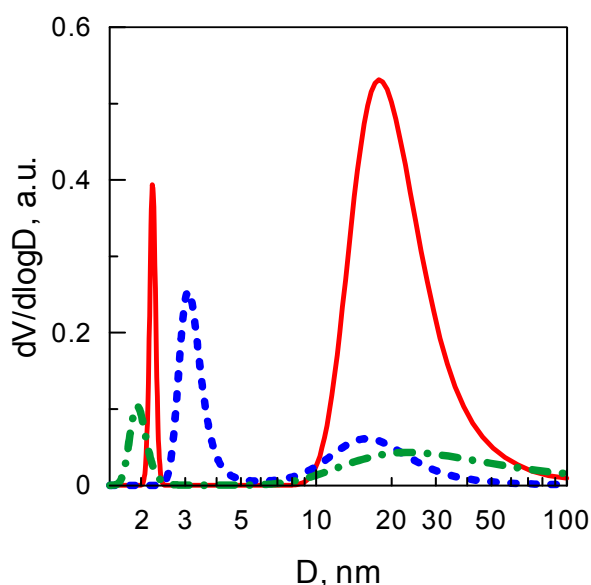


Fig.2 Pore size distribution in MCM-41 obtained from PALS spectra measured in vacuum (solid line), in air (dashed line) and in oxygen (dot-dash line).

2. Various accuracy of results, which is difficult to estimate for each distribution. Air or oxygen quenching causes shortening o-Ps lifetimes of both considered components, whilst decrease of intensity is observed only in the case of the long-lived component. The first factor makes distinguishing between both components harder, but the second one enhances the short-lived component making its lifetime distribution easier to determine.

3. Presence of processes influencing o-Ps lifetimes and dependent on gas pressure, but not taken into account in the model. Such a process is escape of o-Ps from the primary pores to the interparticle spaces [18]. Its presence in the MCM-41 sample produces an inconsistency in the ratios between total volume of the primary pores and the interparticle spaces  $R_v = V_p/V_s$  calculated from the PALS data measured for the sample in vacuum ( $R_v = 0.063 \pm 0.001$ ), air ( $R_v = 0.285 \pm 0.004$ ) and oxygen ( $R_v = 1.14 \pm 0.01$ ). Comparison of these values to the ratio estimated from nitrogen adsorption/desorption isotherms ( $R_v \approx 1.7$ ) shows that  $R_v$  given by PALS is underestimated. Moreover, discrepancy between the values from PALS and nitrogen adsorption/desorption isotherms rises if o-Ps annihilation rate in the primary pores decreases, e.g. from  $57 \mu\text{s}^{-1}$  for MCM-41 in oxygen to  $28 \mu\text{s}^{-1}$  for vacuum. This result confirms migration of o-Ps from smaller free volumes to the larger ones, which is suppressed if o-Ps annihilation rate in the primary pores becomes comparable to or greater than the escape rate. The consequence of the migration is shortening of the lifetime in the primary pores, which leads to underestimated value of pore diameter. This effect explains difference between the diameter for vacuum and oxygen, but not in the case of air, where opposite tendency is observed.

**Pressure dependence of lifetimes.** Assumption that the relation between number of gas molecules in the pores and pressure is linear is a good approximation for both free and adsorbed molecules in the range 0-100 kPa for each oxygen, nitrogen and air [7]. It allows to write Eq.2 in the form dependent on the pressure

$$\lambda = \lambda_{\text{ETE}} + (1 + 4.5/D) \lambda_{\text{gas}} p, \quad (4)$$

where  $p$  is gas pressure and  $\lambda_{\text{gas}}$  is gas quenching rate per unit of gas pressure.

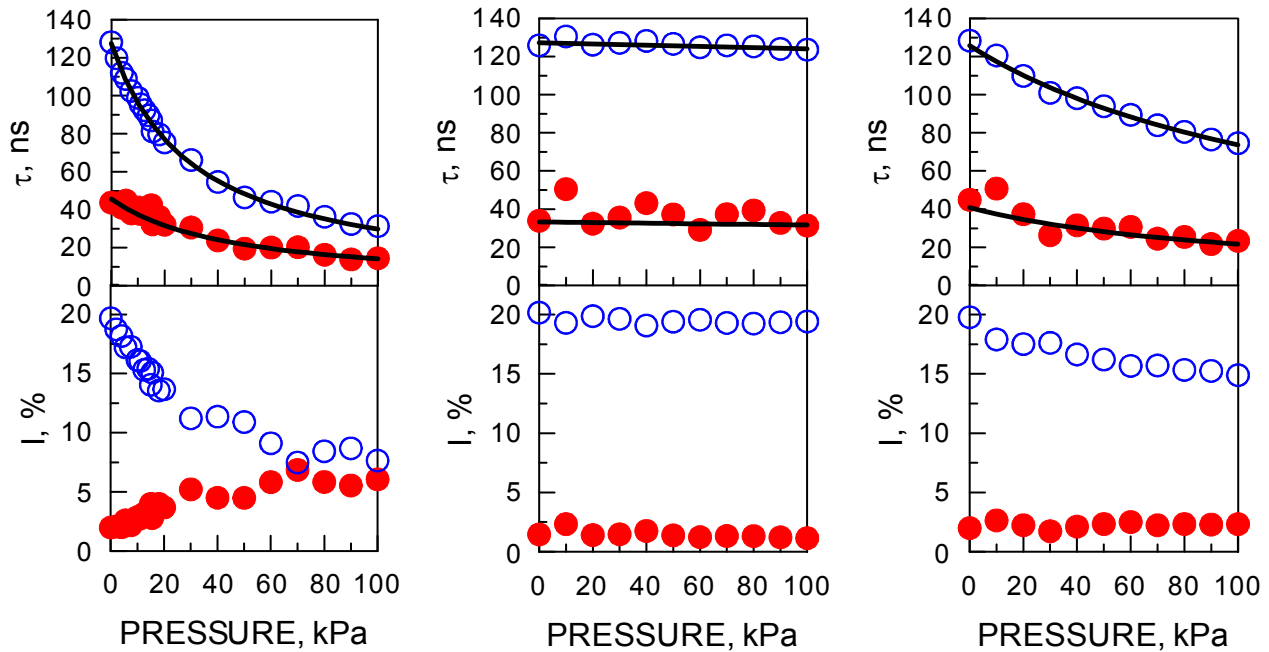


Fig.3 Dependence of o-Ps lifetime and intensity on pressure of oxygen (left), nitrogen (middle) and air (right) for primary pores component (full circles) and interparticle spaces component (open circles) in MCM-41. The lines are the result of fitting Eq.4 to the experimental data.

In order to verify the dependence of o-Ps lifetimes on pressure of gas predicted by the model, PALS spectra for pores filled by oxygen, nitrogen and air under various pressures were measured.

The simplified method of analysis of the spectra by LT was chosen in order to minimize uncertainty of the results. The number of spectrum components was reduced to five by approximating two o-Ps components attributed to micropores by single lifetime. Finite distribution of lifetimes was neglected, what may result in systematic shift of the values, but it is necessary to distinguish both components attributed to mesopores when their lifetimes become close to each other at higher oxygen pressure. The results of the analysis are shown in Fig.3. Expected from Eq.4 hyperbolic-like relation between the lifetimes and gas pressure is visible, as well as decrease of the intensity of the long-lived component. The intensity of the short-lived component seems to be constant or even increases in the case of oxygen, which is the result of suppression of o-Ps migration from the primary pores to the interparticle spaces.

Dependence of o-Ps lifetime and intensity on pressure of oxygen shown in Fig.3 is quite strong, in contrary to weak dependence of these parameters on pressure of nitrogen. This dissimilarity is caused by different o-Ps quenching mechanism in each gas. The quenching in nitrogen is caused mainly by pick-off process occurring during collisions, while quenching by chemical reaction or spin conversion dominates in oxygen. Whether chemical reaction resulting in increase of pick-off rate or spin conversion of o-Ps to p-Ps followed by fast intrinsic annihilation is responsible for oxygen quenching in pores of MCM-41 is hardly to be determined from the positron lifetime spectra. Ratio between probability of quenching by chemical reaction to probability of spin conversion estimated for pores with  $D = 2.4$  nm is about 0.25 [1].

Fitting Eq.4 to the experimental relation presented in Fig.3 allowed to determine values of oxygen, nitrogen and air quenching rates ( $\lambda_{O_2}$ ,  $\lambda_{N_2}$  and  $\lambda_{air}$ ). The results, together with air quenching rate calculated as weighted average of partial oxygen and nitrogen quenching rates ( $\lambda_{(21\% O_2 + 78\% N_2)}$ ), are presented in Table 1. Both  $\lambda_{O_2}$  and  $\lambda_{N_2}$  from the long-lived lifetime are consistent with results of other authors for oxygen [1, 13-15] and nitrogen [19-20]. It is difficult to find reliable data for air quenching rate, but the  $\lambda_{(21\% O_2 + 78\% N_2)}$  is in very good agreement with  $\lambda_{air}$  obtained directly.

Table 1. Oxygen, nitrogen and air quenching rates obtained from the dependence of o-Ps lifetime versus pressure of gas filling primary pores and interparticle spaces of MCM-41. Pore diameters were calculated from the lifetimes extrapolated to 0 Pa using the ETE model.

$D_{ETE}$ , nm	$\lambda_{O_2}$ , $\mu s^{-1} MPa^{-1}$	$\lambda_{N_2}$ , $\mu s^{-1} MPa^{-1}$	$\lambda_{air}$ , $\mu s^{-1} MPa^{-1}$	$\lambda_{(21\% O_2 + 78\% N_2)}$ , $\mu s^{-1} MPa^{-1}$
2.4	$180 \pm 10$	$4.7 \pm 3.5$	$76.5 \pm 10.1$	$39.4 \pm 3.4$
24	$220 \pm 12$	$1.7 \pm 0.2$	$47.2 \pm 5.9$	$47.6 \pm 1.9$

Much worse is agreement of the quenching rates calculated from the pressure dependence of lifetime of the short-lived component. It is possible that due to low intensity of the component the reliability of determination of its lifetime suffers from the approximations assumed during data analysis. It would explain opposite than  $\lambda_{N_2}$  and  $\lambda_{air}$  deviation of  $\lambda_{O_2}(D_{ETE} = 2.4$  nm) for primary pores from  $\lambda_{O_2}(D_{ETE} = 24$  nm) for interparticle spaces, because determination of the lifetime is more accurate in this case due to increased intensity of the short-lived component. There is another explanation of the disagreement if  $\lambda_{O_2}$  is in fact sum of o-Ps quenching rate by oxygen and o-Ps escape rate. Interaction with the gas should slow down the migration of o-Ps along the pore (i.e. decrease of o-Ps escape rate competing with increase of oxygen quenching rate), what would be observed as too slow lifetime shortening with increasing pressure in the primary pores.

## Summary

The relation between o-Ps lifetime and diameter of pore filled by air given by the simplified model presented in this paper reconstructs the tendency observed in experimental data. Nevertheless, the model is not sufficient yet to give precise quantitative results. New experimental data covering the size range of mesopores (well defined pore size and geometry is essential) are needed to develop and verify the model.

Reconstruction of pore size distribution from PALS data can be distorted if the process of o-Ps migration between pores of different sizes is present.

Oxygen, nitrogen and air quenching rates obtained from the dependence of o-Ps lifetime against pressure of gas are in agreement with the data available in the literature only for free volumes having diameter about 24 nm. Results obtained by the same method for smaller pores are not consistent, probably because of some systematic errors or effect of o-Ps escape. However, influence of unknown factors not taken into account in the model and manifesting itself in nanoscale is also possible. In order to elucidate this problem, the measurements analogous to these presented above but for the material where annihilation in small (2-3 nm) and closed (or long enough to neglect o-Ps escape) pores dominates should be performed.

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