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LETTER TO THE EDITOR

Water-induced effects on the positron moderation efficiency of rare gas solids

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Abstract. The first systematic study of the effect of impurities on the positron moderation efficiency of rare gas solids (RGS) is presented. Water contamination in the RGS decreased the moderation efficiency, already observed at the ppb level. A sub-monolayer (~0.1 L) of water on the surface, however, enhances the positron yield by (up to) a factor of 2.1 for Ar and 1.3 for Kr. We explain these observations with changes in the positron affinity, induced by the water dipoles, and suggest further experiments to improve the performance of the RGS.

Low-energy (~eV) positrons are usually produced by using moderators in conjunction with natural radioactive sources or accelerators. In some of them, e.g. W, Ni, Cu and diamond, the material has negative work functions [1, 2], which create the possibility for a fraction of the thermalized positrons, that diffused to the surface before annihilation, to be expelled into the vacuum. In others, such as rare gas solids (RGS), only some epithermal ('hot') positrons are able to escape the moderator, as the work function at the surface is positive [3, 4]. In the latter case higher positron yields are achieved, which are associated with the longer mean free path of the positrons in RGS [3] than in metals or diamond. In general, the contaminants are expected to play a major role in the quality of the RGS by altering the hot positron dynamics. Some evidence for this exists in the literature (for example, compare the results from [5, 6]), but no systematic study has been performed. Our interest in the water, representing an impurity, was motivated by its presence in vacuum systems. This letter will report that the impurities in RGS affect the final efficiency even at ppb level, and are a dominant factor in the energy loss processes above 1 ppm. A totally unexpected result will also be demonstrated in that a small amount of water actually enhances the moderation efficiency. Other possibilities of achieving much larger positron yields of RGS will be given.

A schematic diagram of the experimental apparatus is shown in figure 1. The positrons were obtained using a 19 mCi ²²Na β^+ source, sealed in a capsule by a 6 μ m Ti window. Low-energy positrons were produced by crystallization from the vapour of research grade Ar (99.9999%) or Kr (99.998%) directly onto the source assembly, pre-cooled to 13 K. Accelerated to 100 eV in a 120 G axial magnetic field, they were selected by a $E \times B$

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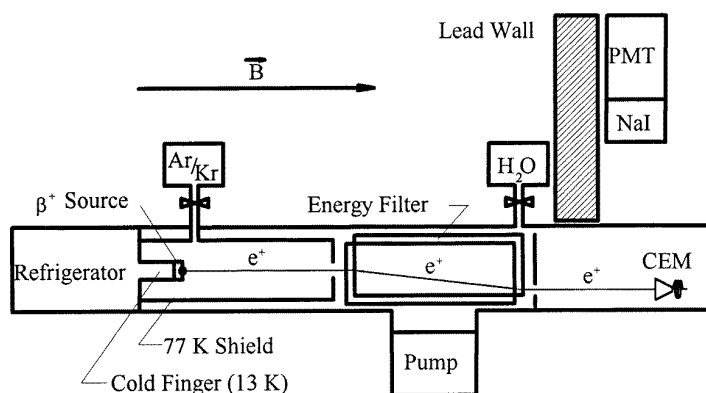


Figure 1. A schematic diagram of the experimental apparatus.

velocity filter and transported to a Channeltron Electron Multiplier, whose efficiency was derived from a coincidence with a $\varnothing 3'' \times 3''$ NaI(Tl) detector. A base pressure in the unbaked system of 1×10^{-9} Torr, dominated by water, was provided by two Balzers turbo-molecular pumps (TMP) (500 l s^{-1} and 50 l s^{-1}) operating in series. The partial pressures of the other impurities were $< 2.5 \times 10^{-10}$ Torr for H_2 and $< 1 \times 10^{-10}$ Torr for the others (CO and CO_2).

The deionized water used in these experiments was further purified by multiple outgassing procedures described below. Ice was produced by cooling to 77 K (LN_2), after which the gases in the reservoir were pumped through the main vacuum system. After it was re-sealed, the reservoir was warmed up to room temperature again. The process of cooling and warming was repeated five or six times until the pressure in the vacuum system did not rise above 10^{-8} Torr while evacuating the gases from the reservoir. A constant flow of the purified water vapour was introduced (at room temperature) into the vacuum system, thus setting the desired pressure in the main chamber in the range of 10^{-6} – 10^{-9} Torr. As the water condenses onto the 77 K shield of the refrigerator, only those molecules passing through the aperture and directed toward the pre-cooled source condensed on it. The attenuation in the water partial pressure at the moderator, as compared with the pressure in the main chamber, was found to be 1.5×10^{-3} . This allowed us precise control over introducing impurities at sub-ppm level. In these experiments the water pressure was first established at the desired level and kept constant throughout the whole experiment. Even at the maximum H_2O partial pressure at the source (10^{-9} Torr), the water would form a sub-monolayer before the rare gas injection, and thus β^+ attenuation would be insignificant. The moderator was produced afterwards.

The rare gas injection pressures were optimized for maximum efficiency of clean moderator (no water injected) and estimated at 2.4 mTorr for Ar and 0.6 mTorr for Kr. The calculations are based on (i) the pressure, as read in the main system and corrected for the ionization cross-section ratios of Ar/N_2 (1.19) and Kr/N_2 (1.81) [8], (ii) the pumping speed of the TMP, 400 l s^{-1} for Ar and 500 l s^{-1} for Kr, as stated by the manufacturer, and (iii) the conductance of the aperture. The estimated values agreed with the reading of the pressure when the system was closed (no pumping) for a short period of time. The water concentration in RGS given in this letter is the ratio of the partial pressures of water to those of the rare gases (Ar/Kr), assuming sticking coefficient unity at 13 K for all of their vapours.

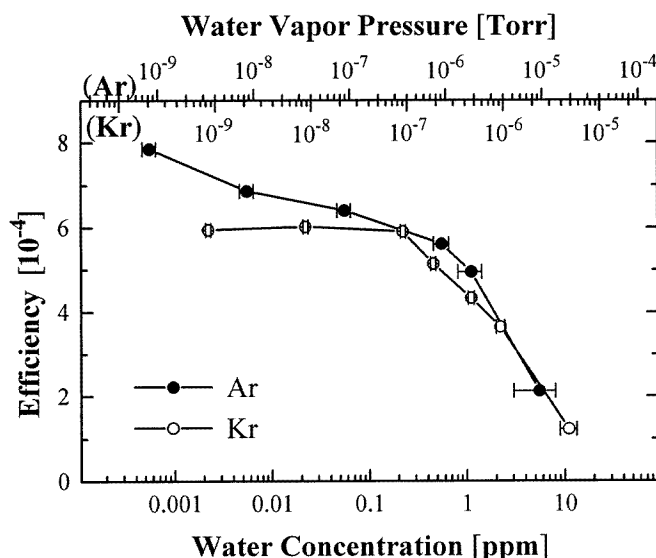


Figure 2. Moderation efficiency of Ar (solid points) and Kr (open points) as a function of the water concentration in the bulk crystal. The top scale represents the water pressure as measured in the main chamber.

The results we obtained for the moderation efficiency as a function of the water concentration in the RGS are shown in figure 2. The measured water pressures in the main system are given on the top scale for both cases, Ar and Kr. As expected, a large amount of water in the rare gas crystal (>1 ppm) results in a significant decrease in the moderation quality of the crystal. Either the water dominates the energy loss process of the hot (\sim eV) positrons by reducing their energy below the affinity level, or positrons are lost to positronium formation at energies lower than the threshold for the RGS. Of greater importance, however, is the fact that even a small amount of water (\sim ppb) still plays a role in the positron dynamics, which could strongly reflect as underestimation in the evaluation of the positron diffusion length in RGS [3]. This conclusion is still valid even if the water concentration is wrong by a full order of magnitude if there are smaller sticking coefficients (0.1) for Ar and Kr. The difference in both curves simply accounts for the intrinsic purity of the gases, stated to be <1 ppm H_2O in Kr and <0.1 ppm H_2O in Ar. The experiments were repeated at 0.21 and 25 mTorr Ar injection pressures and revealed the same dependence. There is no reason why other impurities (CO , O_2 , N_2 , etc) present in the moderator crystal would play a different role.

In order to separate the influence of the water in the system on the RGS positron yield, the effect of its deposition onto clean moderators was investigated. Ar and Kr moderators were prepared at 25 and 6 mTorr, respectively. They were annealed until their vapour pressure reached 1 mTorr, after which their low-energy positron yields were normalized to unity. The water coverage on the surface was achieved by injecting it at 1.5×10^{-10} Torr (1×10^{-7} Torr in the main chamber). These experiments yielded a totally unexpected result (figure 3). A small amount of water, ~ 0.1 L ($1 \text{ L} = 10^{-6}$ Torr s), actually enhances the moderation efficiency by a factor ranging between 1.80 and 2.05 for Ar and 1.22 and 1.27 for Kr in different experiments. More detailed study revealed slightly different values of the water coverage in Ar and Kr for which the maximum efficiency is reached, 0.13 ± 0.01 L

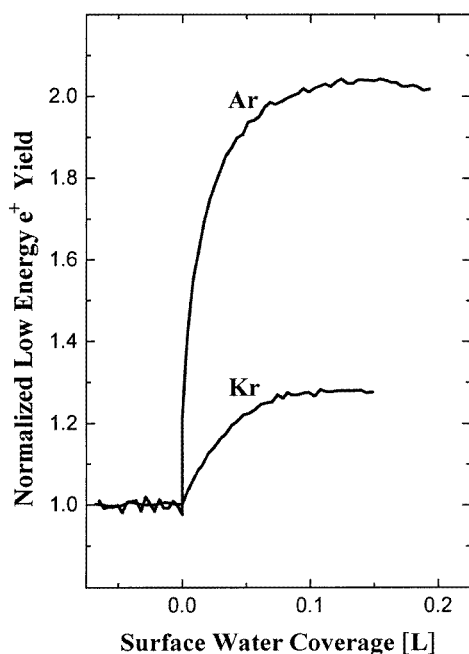


Figure 3. The best achieved enhancement in the moderation efficiency for Ar and Kr. The low-energy positron yield, normalized to its value before the water injection, presented as a function of the estimated surface water coverage.

(Ar) and 0.11 ± 0.01 L (Kr). Most recently, we have observed the same effect ($\times 1.15$ increase) in the moderation efficiency of Kr in conjunction with stronger source, 1.5 Ci ^{64}Cu . If the water deposition continues further (>0.2 L), the positron yield fits to an exponential decrease with 30% per Langmuir rate for the following 5–6 L due to energy losses in the water molecules on the RGS surface.

The present explanation of the observed phenomenon at 0.1 L water coverage is related to either water-induced changes of the surface dipole, D , which affects the positron work function ϕ_+ , or field-assisted moderation caused by trapped electrons in the vicinity of the water molecules. The latter, however, has been found to be accompanied by a negative shift of the slow positron energy spectrum of the order of several volts (-15 to -20 V) [9]. By scanning the $E \times B$ velocity filter, we conclude that no energy shift (>0.5 V) takes place, thus we rule out the field-assisted moderation. We suggest that the effect is caused by an affinity change, $\Delta\phi_+$, toward less positive values of ϕ_+ , by changing the surface dipole barrier. Thus, positrons with energy within ϕ_+ and $\phi_+ - \Delta\phi_+$ are able to escape in vacuum. Indeed, if the water molecules are arranged with the hydrogen atoms toward the RGS surface, which is an energetically favourable orientation, the water dipole would contribute to such an affinity change. Simple calculations, based on the number of water molecules (1 L = 0.27 monolayer), all oriented in the same direction, and assuming unity sticking coefficient, yield $\Delta\phi_+ = -0.2$ eV. However, the long-range Coulomb forces also need to be taken into account in the energetic approach to the problem. When in close proximity, the water molecules would sense the repulsive electrostatic forces, which will eventually counterbalance the dipole orientation described above. In such cases, larger water coverage would lead to either orientation of the dipoles and the positrons will undergo

energy loss processes, a fact which is in agreement with the experiment.

The above explanation could motivate further extended studies. Instead of water, we suggest the use of materials with large dipole moment and a simple structure to avoid the energy loss processes. Good candidates would be alkali halides, the best of which would be CsI. As some of them have dipole moments five times greater than that of water, they would lead to an affinity change of the order of 1 eV. We expect that their optimum coverage would be the same, 0.1 L, because the effect of the Coulomb forces would not change. Thus, one may expect a factor of 5–10 increase in the positron yield of RGS.

In conclusion, the present study of the impurities in and on RGS reveals their importance for achieving good solid rare gas moderation efficiencies for low-energy positrons. Even at the ppb concentration level, the impurities have an impact on the diffusion length of the epithermal positrons. The surprising effect of 0.1 L water coverage creates new possibilities for reaching higher positron yields of RGS. A further detailed investigation, involving alkali halides, is needed to determine the changes in the surface dipole barrier.

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