BNL-63292

CONF-9606252--3

An Investigation Of Positrons Interacting With Solid Argon, Krypton And Xenon

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Abstract

With this article we intend to shed some light on all the important characteristics of the up-to-date most efficient positron moderators, the rare gas solids. We stress on the importance of the impurities in the performance of the solid rare gas moderators. The impurity factor is linked with the crystalline changes to explain the effect of annealing, and demonstrate the role of impurities in the endurance. Significant increase in the low energy positron yield is observed after repeated anneals. The positron energy distributions from Ar, Kr, and Xe moderators are measured to be about 2 eV (FWHM).

Key words: positron beam, moderators, solid rare gases, impurity

PACS: 78.70.Bj, 61.72.Ss, 78.50.-w

DISCLAIMER

1. Introduction

A decade ago the rare gas solids were used to produce a low energy positron beam [1,2] and the "hot positron model" was introduced to explain the experimental results. The efficiency of the solid rare gas moderators (SRGM) was found to be close to 1% (for Ne), an order of magnitude higher than that of a tungsten single crystal moderator, and the geometry configuration was found to be of importance also. However, the slow positrons, leaving the moderator, had a broad energy spectrum.

The processes of rare gas crystal formation are well studied in the past [3]. Growing a crystal of good quality, from the liquid, is not feasible in ultra-high vacuum applications. To improve on the crystalline structure of grown from the vapor solid, Grund *et al.* [4] introduced the idea of annealing. In the most recent work by Mills *et al.* [5] it was concluded that Ne, Kr and Xe moderators have comparative efficiencies after anneal. In this work we study different aspects of the performance of Ar, Kr and Xe moderators only, since Ne requires complicated apparatus (three-stage refrigerator).

2. Experimental Apparatus

A 19 mCi ^{22}Na source, deposited in a 6 mm-in-diameter spot on the back side of a 6 μm Ti foil, and sealed in a stainless steel capsule, was mounted on the cold finger of a two-stage refrigerator (Fig. 1). The resulting yield was 25% of all available β^+ from the source. Research grade gases with stated purity of Ar (99.9999%), Kr (99.997%), and Xe (99.995%) were used as moderator material. The moderators were crystallized from the vapor onto the pre-cooled (13 K) source assembly. The low energy positrons were transported in magnetic field to a target, biased at -200 V. The annihilation γ-rays were detected in coincidence between two ϕ3"x3" NaI(Tl) detectors. A base pressure of less than $1 \cdot 10^{-10}$ Torr after a thorough bake-out was provided by an open end pumping system, consisting of two Balzers turbo-molecular pumps (500 l/s and 50 l/s), operating in series [6]. The gas analysis was done on a VG SX-200 Quadrupole Mass Analyzer.

3. Results and Discussions

The SRGM efficiency is defined as the ratio of the low energy positron intensity and the β^+ activity of the source. One must note that the β^+ efficiency of the source assembly depends very strongly on the specifics of its preparation and the final efficiency may vary significantly in different laboratories. Of great importance is also the shape in which the moderator is formed, the so called geometry factor. Many applications take advantage of the large increase in the moderation efficiency of conical moderators, relative to "flat face" one (factors of 2-5 [7]), but it would only be fair to compare moderators of the same geometry. The results presented below are for transmission moderator (For metals, see Ref. [8]).

We will employ the role of the impurities to explain the effect of annealing on the moderators efficiencies (Sec. 3.1), and their endurance (Sec. 3.2) and we believe that their presence effects the energy spectra of the slow positrons (sec. 3.3).

3.1. Annealing

The solid rare gas moderators (SRGM) are typically grown from the vapor, sublimated onto a cold surface below the triple point, with no liquid present. It is well known [3] that these prepared crystals are more defective and finer grained than those grown from the liquid. It was experimentally established [4] that the annealing of the moderator crystals after deposition increases their efficiency, but no discussion is given on the driving mechanism of the effect. Because of the positive affinity of the SRGM, the improving of the crystalline structure could not explain the phenomenon by itself. Below, we suggest an explanation that incorporates the link between the crystalline structure and the impurities in the solid.

Grown at 13 K and $2\cdot10^{-5}$ Torr, the rare gas crystals consist of 0.05-0.1 μm grains [3], significantly smaller then the positron diffusion length, Λ (~0.5 μm [1]). It is also known [3], that, in general, the impurities tend to precipitate at the grain boundaries. Under these conditions, the positrons may encounter impurities several times during their random walk in the bulk within their lifetime. Thus, they are provided with an additional energy loss mechanism, not taken into account in the hot positron model. As the loss per collision could scale up to 500 meV, it is reasonable to expect that it may dominate over the phonon scattering.

Ar, Kr, and Xe, annealed to 36 K, 49 K, and 68 K, respectively ($1\cdot10^{-4}$ Torr [9]), have larger grains 0.3-1 μm [3], comparable with Λ. This effectively reduces the local concentration of the impurities in the region, probed by a single positron and the phonon scattering becomes the dominant energy loss process. Therefore, more positrons would reach the surface of the solid, and an increase in the moderator efficiency would be observed. It is logical to expect, that the annealing would have no longer effect, once the grains become much larger than Λ. For example, a Xe crystal with grains measuring between 1-5 μm is produced by annealing to 100 K (0.6 Torr) [3], which gives an idea about the required pressure range.

To avoid the problem with the high pressure, a Kr moderator, formed at 4 mTorr and 13 K, was subjected to anneals up to a fixed temperature, T_a (T_a=42; 45; 48; 51 K), for several consecutive times. Special care was taken to minimize the gas losses due to its pumping out. The maximum reached efficiencies for each of these temperatures had the same value, ε_m, within the statistical limits, and the presented results (Fig. 2, for clarity, T_a=42; 51 K) are normalized to it. The value at zero is related to the efficiency after fabrication. It is clear, that the first anneal is the one that plays the major role, but even higher positron yield could be achieved by repeated anneals. The ε_m of the moderator, annealed equivalently longer at T_a, corresponds to efficiency after one anneal at some higher temperature, for which the vapor pressure could not be handled by the system. The loss of moderator material in further anneals reflected in a lower positron yield.

3.2. Endurance

SRGM were prepared from Ar, Kr, and Xe at 13 K and $2\cdot10^{-5}$ Torr injection pressure. They were annealed as described in Sec 3.1. In agreement with Ref. [5], we find that the deposition temperature does not affect significantly their final efficiency, providing that anneal is done afterwards. The endurance of the moderators was then monitored for over 5 days (Fig. 3a). A change in the moderator efficiency was observed during the first day, but it becomes less significant afterwards. After 5-6 days, the moderators were reannealed and they regained their initial efficiency. With proper normalization, it could be demonstrated (Fig. 3b) that the deterioration in the moderation efficiency is not a result of any internal structural changes, but rather a surface contamination. The efficiency for each gas (Ar, Kr, Xe) is normalized to its value at t=0, the moment when the annealing process is terminated. Thus defined relative efficiencies are monitored for one day. The clearly shown similar behavior does not depend on the type rare gas, therefore, it could be concluded that it is due to surface contamination.

An impurity analysis of the environment shows the dominant presence of the hydrogen and CO, second in quantity. At the operating conditions, the hydrogen does not condense, therefore we attribute the decrease in the moderator efficiency to the CO, physi-sorbed onto the surface. Knowing the partial pressure of CO, $\sim 2 \cdot 10^{-11}$ Torr, and assuming unity sticking coefficient, the rate for its pile-up is close to 2 L per day, which is in a good agreement with the endurance results (Fig.3a,b). The process of "poisoning" the SRGM with the deposition of N_2, O_2 and CO was also confirmed in other tests.

In some situations the pile-up of impurity on the SRGM surface could be used to improve their qualities. Such is the case of sub-monolayer amount of water, which enhances the positron yield of Ar and Kr moderators (Fig. 4) [10]. The effect is attributed to the change in the positron affinity.

3.3. Slow Positron Energy Distribution

The low energy positron spectra of SRGM had been discussed in other work [1]. It must be noted, however, that those moderators were not annealed at high temperature, as the procedure was introduced later. To investigate this effect, our system was modified as follows. Two 92% transmission tungsten grids, spaced at 2 mm, were biased at the retarding voltage, V_R, while the third (92%) one was kept always at ground potential. The grid assembly was mounted on the thermal radiation shield of the refrigerator (~60 K) in order to avoid condensation of the rare gases on it. The moderator was fabricated on 40 μm Al foil, glued onto the Cu source cap. The constructed retarder was expected to have less than 1% energy resolution. A Channeltron Electron Multiplier (CEM) was used in these experiments.

The moderators were grown at high pressure (~10 mTorr) to ensure that no significant amount of impurities from the vacuum environment is added in the crystals during their growth. They were later annealed, until their vapor pressure reached $1\cdot10^{-4}$ Torr. Energy distribution spectra were measured for each type of moderator before and after annealing. The results were normalized to maximum height in the spectra of the annealed crystals (Fig. 5). It is clear that their shape differs significantly from a gaussian. For that purpose, in addition to the full-width-at-half-maximum (FWHM), it is useful to introduce another parameter, f, the fraction of positrons with energy within FWHM. The experimental values for FWHM and f are: 2.0 eV and 50% for Ar, 1.7 eV and 60% for Kr, and 2.2 eV and 70% for Xe.

No major differences were observed in the shape of the positron spectra of annealed and not annealed moderators. In the former case, however, the spectra appeared shifted toward the lower energies, relative to those before annealing. The magnitude of the shift was small for Ar (-0.2 eV) and Xe (-0.4 eV), but significant for Kr (-2.4 eV). The negative charges (secondary electrons) causing the shift are probably trapped in the vicinity of impurities on the surface, which migrated there during the annealing. The migration in Ar is small, because of the low annealing temperatures. Thus the change in the impurity concentration on the surface is small, therefore the small shift. Xe, on the other hand, sublimates at high temperatures, for which most of the common impurities have very high vapor pressure [9] and they may be desorbed. Hence, the surface could be considered to be relatively clean after the annealing, except for water. Anneals at temperatures, suitable for Kr, could lead to an increase of the surface impurities by an order of magnitude, which provides for the large energy shift.

By expectation, according to the hot positron model, the energy distribution of the moderated positrons would be exponential, ignoring final details. In our measurements, two peaks could be distinguished in case of Ar (1.4 eV and 6.0 eV) and Kr (0.8 eV and 3.2 eV). Similar results were shown in Ref. [1], although not discussed, and were also supported in other measurements [11]. At the present, we could not give a conclusive explanation, but we believe that the phenomenon is related to an undescribed energy state in the SRGM.

4. Conclusion Remarks

In this article we presented variety of aspects in the interaction of the positrons with the rare gas solids, used to convert high energy β^+ particles to low energy positron beam. We stressed on the importance of the role of the impurities in the performance of the SRGM. The given explanation of the effect of the anneals takes into consideration the additional energy loss process, provided by the impurities, and incorporates it in respect to the changes in the crystalline structure. The impurity pile-up on the SRGM surface are responsible for the deterioration in the slow positron yield, as it was demonstrated, although in some cases (water) the impurities may enhance the efficiency. To make the research more complete, we measured the energy distribution of the low energy positrons, produced with Ar, Kr and Xe moderators, and found them to be about 2 eV (FWHM). Evidence for surface charge trapping, most likely in impurities, was also observed. We comment on the rather complicated structure in the energy spectra.

Acknowledgments

The authors would like to thank Dr. F. M. Jacobsen for his interest in this research, as well as for his many valuable discussions, suggestions and comments. This work was supported by US DOE under contract #DE-AC02-76CH00016.

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FIGURE CAPTIONS

Fig. 1 Positron beam for solid rare gas studies, experimental set-up.

Fig. 2 Relative efficiency of SRGM versus the number of consecutive anneals.

Fig. 3 Endurance of Ar, Kr and Xe moderators for a week (a), the efficiency is given on the left axis. Normalized to the value at the end of the annealing, the positron yield, monitored for a day (b) indicates that the deterioration of the moderator efficiency is caused by surface contamination.

Fig. 4 The normalized slow positron yield versus the water coverage on the SRGM surface shows the actual increase in the moderation efficiency of Ar and Kr at ~0.1 L water.

Fig. 5 Energy distribution of the slow positrons produced by solid Ar, Kr and Xe, before and after annealing. The E_{th} indicates the positronium formation threshold.

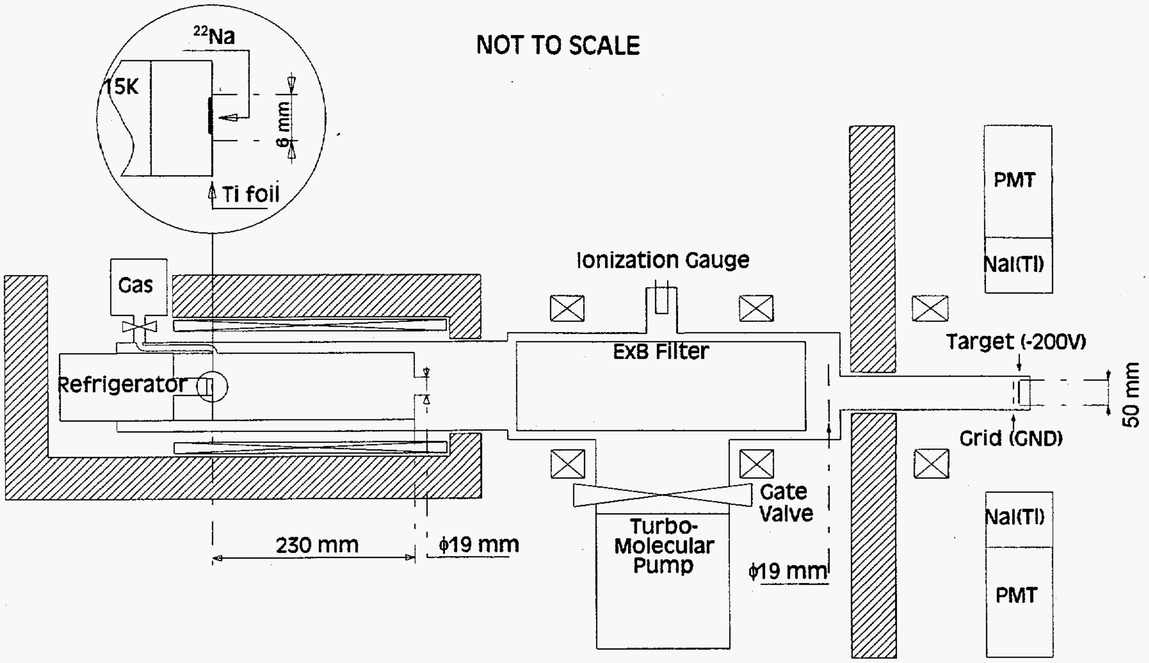
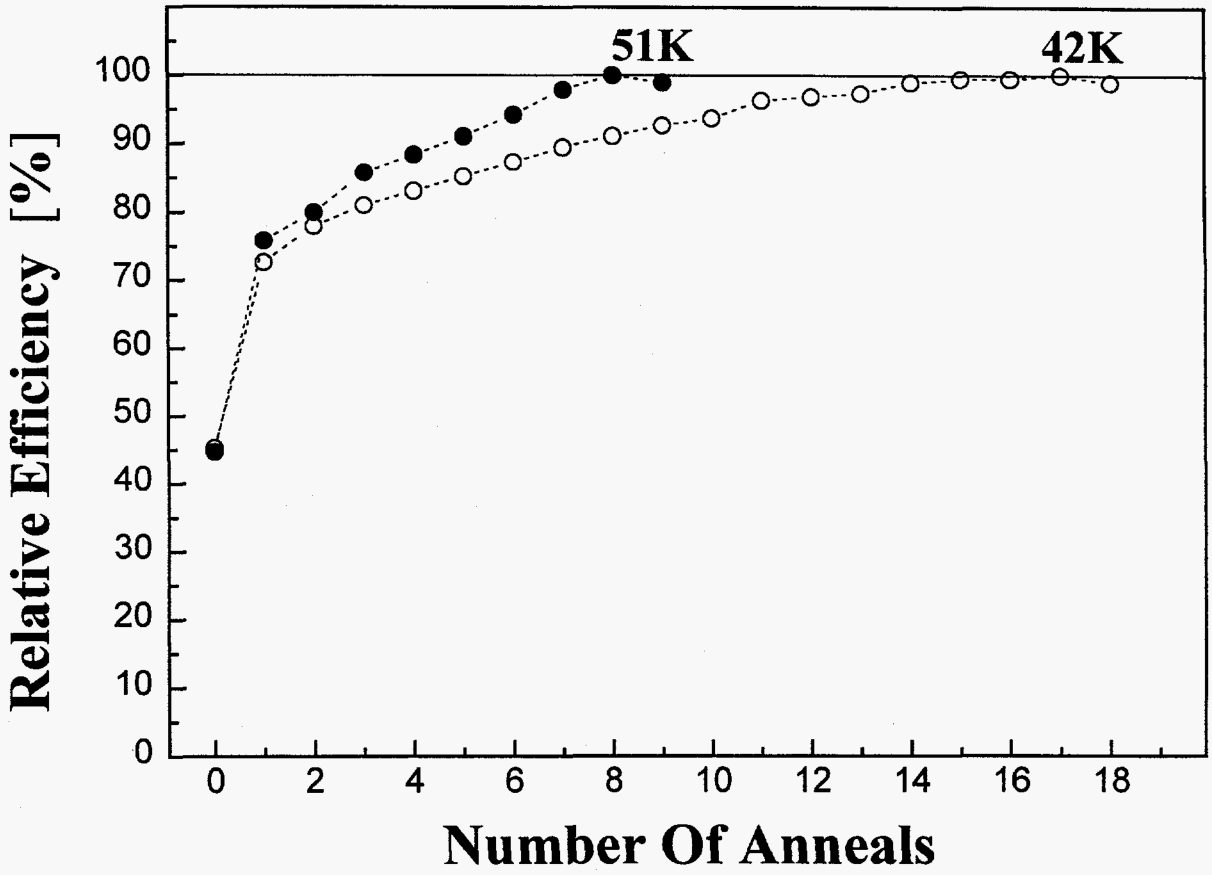
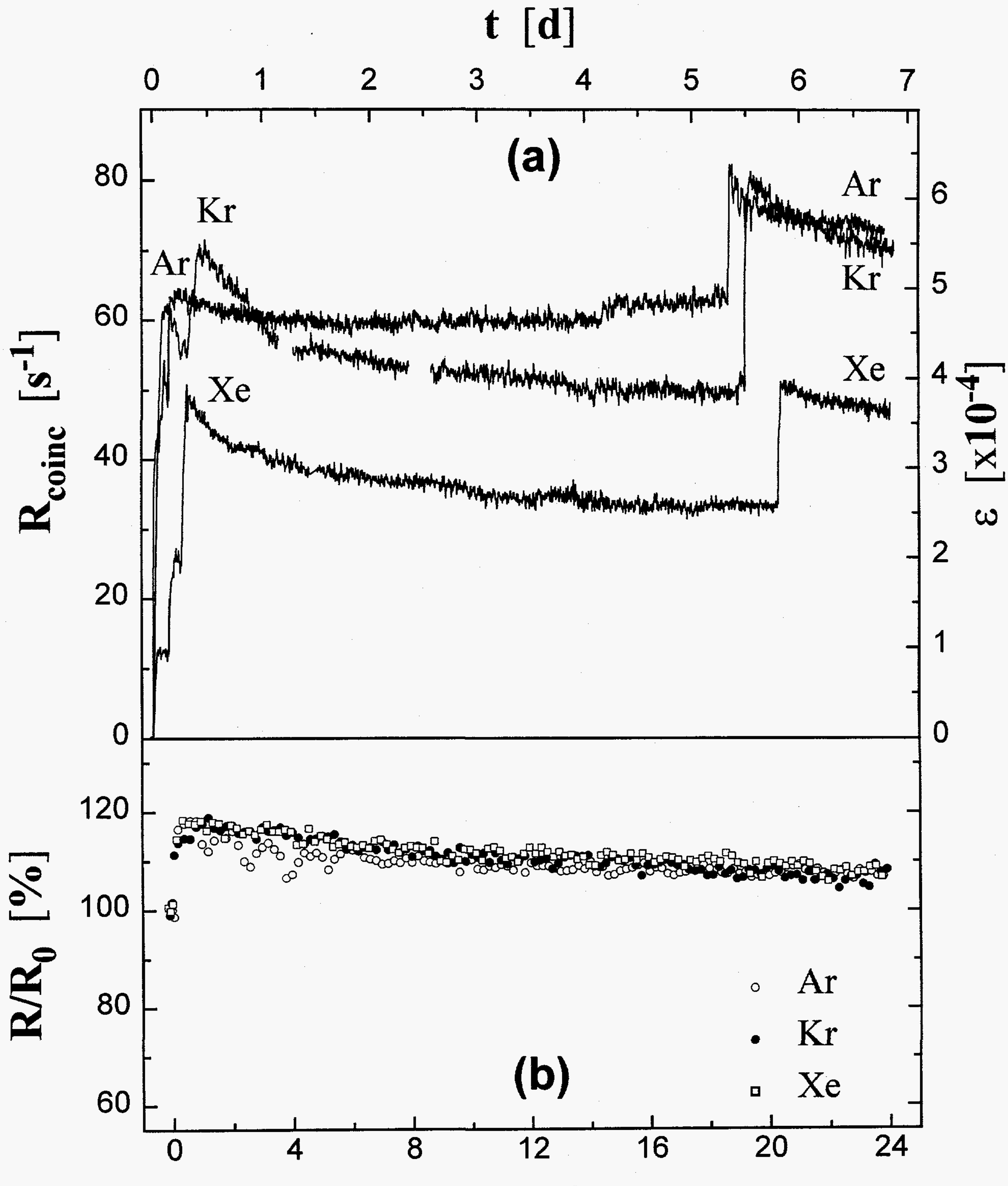

NOT TO SCALE
^{22}Na
15K
6 mm
Ti foil
Gas
Refrigerator
Ionization Gauge
ExB Filter
230 mm
ϕ19 mm
Turbo-Molecular Pump
Gate Valve
ϕ19 mm
PMT
NaI(Tl)
Target (-200V)
50 mm
Grid (GND)
NaI(Tl)
PMT

Fig. 1



51K
42K
Relative Efficiency [%]
0
10
20
30
40
50
60
70
80
90
100
0
2
4
6
8
10
12
14
16
18
Number Of Anneals

Fig. 2



t [d]
0
1
2
3
4
5
6
7
(a)
Ar
Kr
Ar
Kr
Xe
Xe
R_{coinc} [s^{-1}]
80
60
40
20
0
ε [x10^{-4}]
6
5
4
3
2
1
0
R/R_0 [%]
120
100
80
60
○ Ar
● Kr
□ Xe
(b)
0
4
8
12
16
20
24
t [h]

Fig. 3

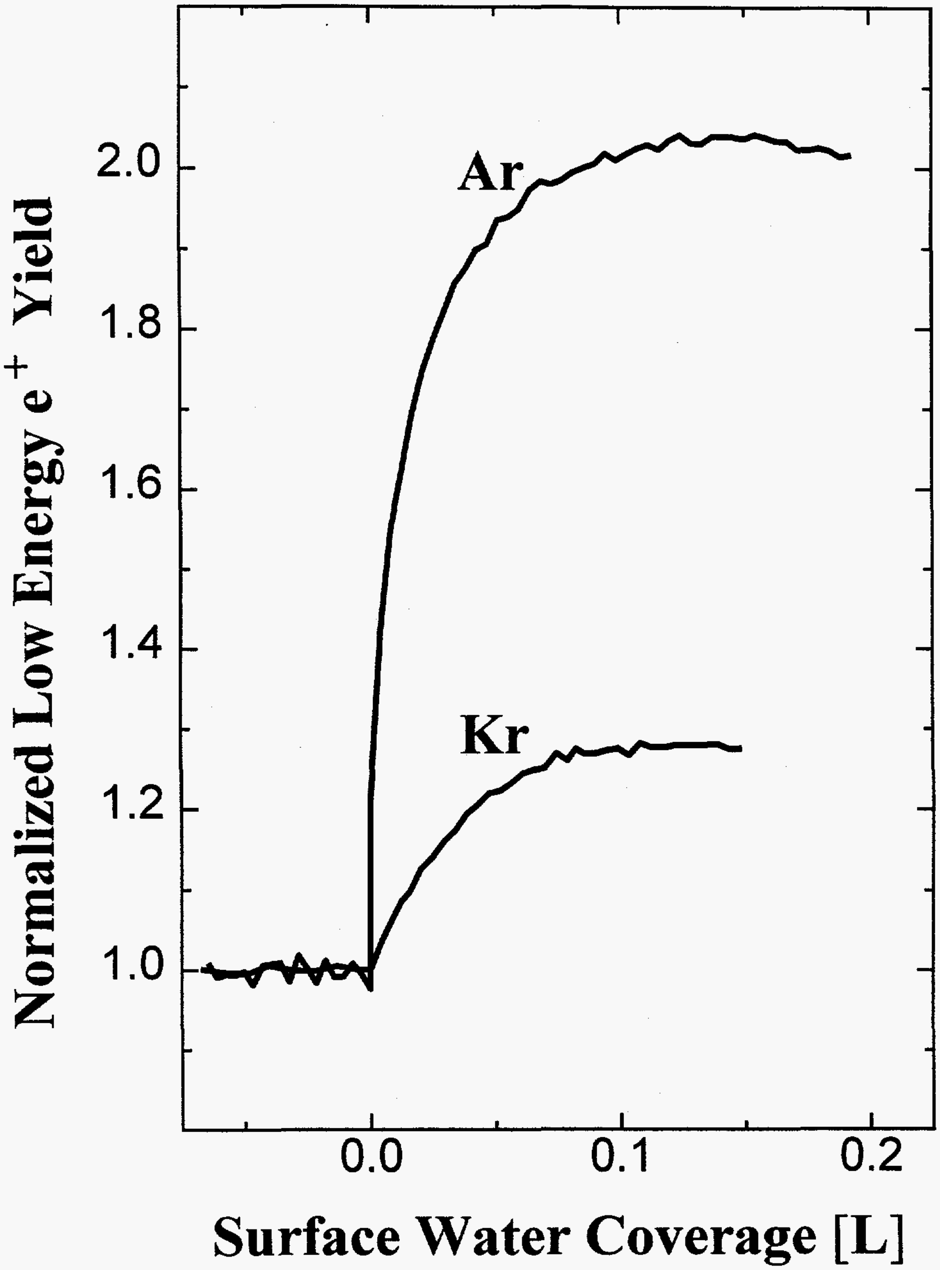
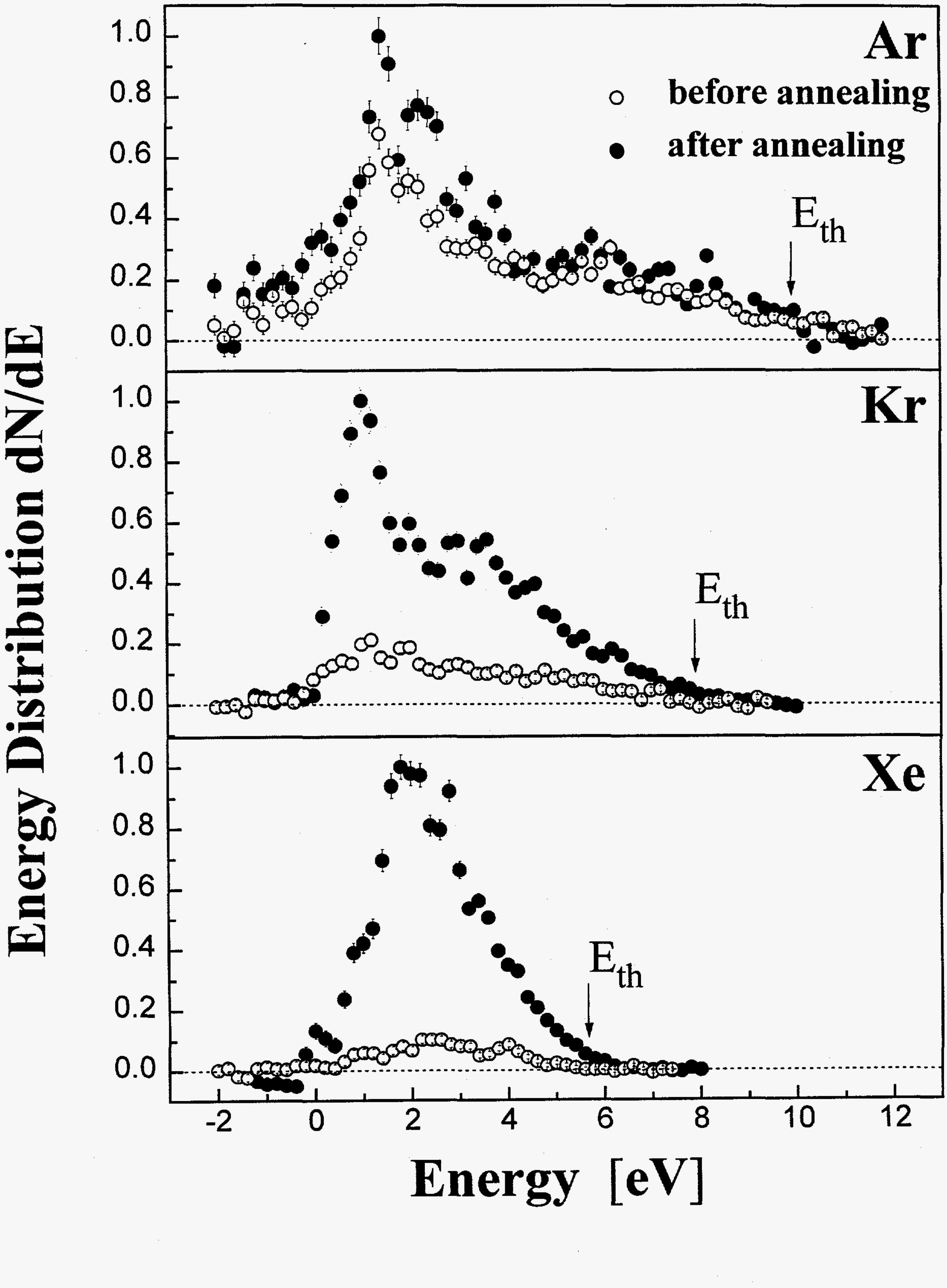

Normalized Low Energy e$^+$ Yield
Surface Water Coverage [L]
Ar
Kr
1.0
1.2
1.4
1.6
1.8
2.0
0.0
0.1
0.2

Fig. 4



Ar
○ before annealing
● after annealing
E_th
Kr
E_th
Xe
E_th
Energy Distribution dN/dE
1.0
0.8
0.6
0.4
0.2
0.0
-2
0
2
4
6
8
10
12
Energy [eV]

Fig. 5