Development of New Detector System for Charged Particle Emission

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Abstract

A charged particle detector system for high temperature gas permeation experiments was developed. We employed a phoswich scintillation counter which consists of a thin YAP(Ce) scintillator and a thicker plastic scintillator, BC-444. Scintillation pulse signals were recorded by a digital storage oscilloscope to realize careful off-line analyses by using the pulse shape discrimination (PSD) technique. Moreover, the system has large plastic scintillators surrounding the reaction chamber: they serve as veto counters and reject cosmic-ray events. Consequently, the system can identify energetic charged particles for emission rate as low as 3 counts/day. In a long period measurement of gas permeation through a Pd/CaO/Pd complex foil, a slight difference of counting rate between D_2 and H_2 gas was observed in the high energy region. It is suggested that high energy charged particles, most probably α particles, are emitted during D_2 gas permeation.

1. Introduction

Charged particle detection is the most sensitive method to verify the occurrence of nuclear reactions in condensed matter. A CR-39 track detector has generally been used in electrolysis and desorption experiments.[1-4] However, the detector cannot provide a time profile of the reaction rate, and it is difficult to get a good energy calibration as well as particle identification. Furthermore, background events caused by cosmic rays and natural radioisotopes cannot be easily suppressed. In order to overcome the disadvantages, we have developed a new charged particle detector system which is suitable for the type of gas permeation experiment originally reported by Iwamura et al. [5]

2. Detector system

The kernel of the new detector is Ce doped YAlO3 scintillator (YAP(Ce)), which is chemically and mechanically stable even in high temperature hydrogen atmosphere[6-8] where a Si semiconductor detector does not work normally. A variation of light yield of the YAP(Ce) is within +2 and -8% below 200°C: the temperature dependence is negligible below 100°C (i.e., in the present gas permeation experiment). The decay time of scintillation light of the YAP(Ce) is about 30 ns.

In our detector system, the YAP(Ce) crystal (ϕ20 × 0.5 mm) is coupled to a BC-444 plastic scintillator (□20 × 5 mm) which emits scintillation light with slower decay time (~280 ns) to form a phoswich detector. As is shown in Fig. 1, a photomultiplier tube (PMT), which is placed outside of the chamber and kept at room temperature, is connected with the phoswich by a quartz light guide. An energy calibration curve of the YAP(Ce) was obtained with a 5.5 MeV α particle from ^{241}Am source, together with the reported values.[9]

Electric pulses from the PMT are recorded by digital storage oscilloscope and analyzed to identify energy and particles. Figure 2 shows schematically a function of the phoswich detector to distinguish necessary events from those of background originating mainly from cosmic rays. Charged particles (~50 MeV α and 12 MeV proton) emitted from a sample stop within the YAP(Ce) crystal due to large energy loss of them and make a fast electric pulse (YAP(Ce) component) whose shape is shown at a left part in Fig. 2. On the other hand, cosmic rays or high energy charged particles from another direction can pass through both the YAP(Ce) and the BC-444 scintillators, and therefore, make a pulse with a longer tail (BC-444 component) as shown in the right side pulse shape in Fig. 2, where the time 0 in Fig. 2 corresponds to the trigger time of the digital oscilloscope.

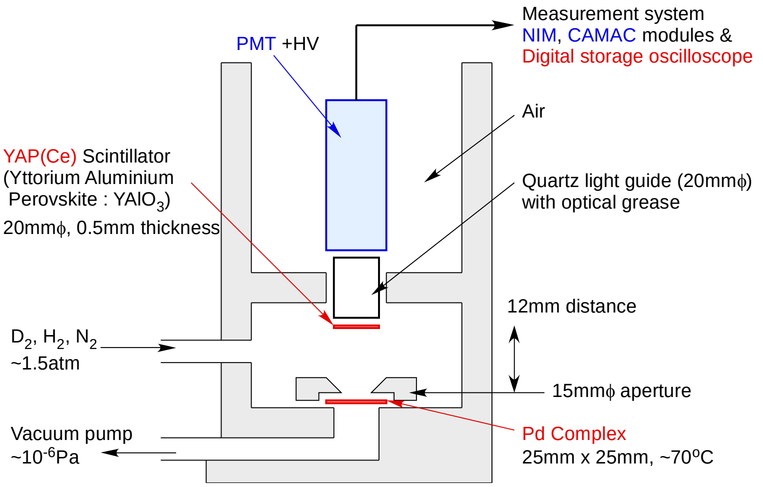

Figure 1. Schematic view of an experimental setup. A phoswich detector is placed 12 mm above a sample and covers a solid angle of 18 %. One side of the sample is pressurized by D_2 or H_2 gas and the opposite side is evacuated by a vacuum pump. The gas is absorbed into the sample and permeates through in it due to density divergence.

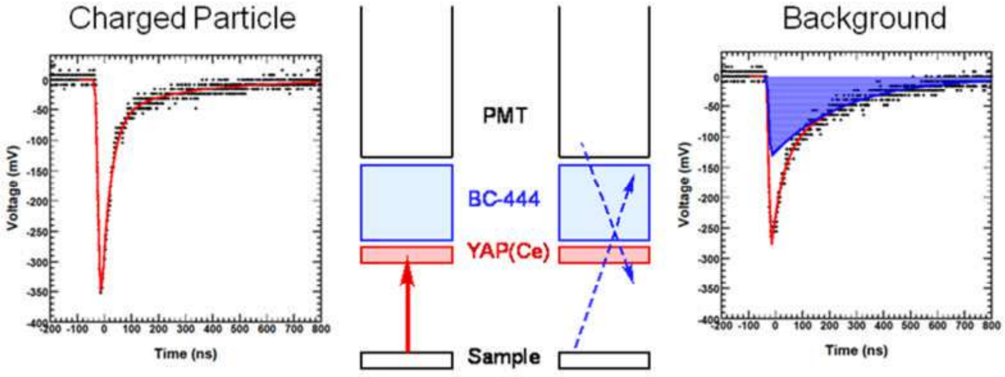

Figure 2. Function of YAP(Ce)/BC-444 phoswich detector and pulse shape discrimination technique. Heavier charged particles such as α particles or protons stop within the YAP(Ce) (left plot). On the other hand, energetic particles can pass through the YAP(Ce) and enter the BC-444, so that a pulse shape has a slower decay component caused by the BC-444 indicated by the blue area, in addition to a fast component indicated by the red curve (right plot).

Discrimination of the events was made by two steps. First, two time windows are set for the pulse shape spectrum, and light yield in each window was deduced: a short window covers between -25 and 75 ns while a long one between -50 and 800 ns. We excluded events with a sufficiently long tail in accordance with the ratio of the two yields. Next, a decay constant of each pulse shape was examined carefully, and events having an α-like pulse shape were finally selected: they are designated pulse shape discriminated (PSD) events. We can assume that the PSD events are heavier charged particles than proton (p, d, α etc.).

In addition to the phoswich detector, the system includes large plastic scintillators which surround the gas permeation chamber and serve as veto counters. These detectors reject 60% of counts as cosmic ray events. Consequently, the new detector system can identify charged particles emitted from the sample during gas permeation for quite low counting rate such as ~ 3 counts/day at several MeV region, because the background level is below 1 count/day.

3. Gas permeation

We have used the detector system to observe charged particles during the gas permeation experiment. Samples of Pd/CaO/Pd complex foil (the gas permeatable area is circular shape with 15 mm diam.) were provided by groups at Iwate University and Mitsubishi Heavy Industries (MHI). Those from Iwate Univ. were prepared fully based on the original recipe of the MHI. In the present work, 4 samples were examined: 2 from Iwate (I-1 and I-2) and 2 from MHI (M-1 and M-2). Foils of I-1, M-1 and M-2 have a Cs deposited layer on the surface as described in Ref. 5.

As shown in Fig. 1, the sample was placed at a position 12 mm below the phoswich detector. Temperature of the sample was controlled by ohmic heaters to be about 70°C. Maximum pressure of D_2 and H_2 gas was 1.5 atm and a gas flow rate was typically about 0.1 ~ 0.5 ccm. It should be noted that the MHI group claims that a much higher flow rate, ~ 3 ccm, is important to achieve nuclear transmutations, but we have not been able to achieve such high rates in this study. After the D_2 gas permeation continues for about 1 week we evacuated the chamber until the pressure falls below 10^{-4} Pa, promoting D_2 gas desorption. Measurement with the H_2 gas permeation, which can be regarded as a background run, was carried out with the same sample after the D_2 gas desorption. We ignored an effect of residual gas in the Pd complex sample, since its quantity was considered very small. Finally, we acquired data during 1 week for each gas and under a vacuum as well.

4. Results

In the offline analyses, energy spectra of the PSD events were made for D2 gas (denoted D2), H_2 gas (H_2) and vacuum (Vac.). Figure 3 shows such spectra for the I-1 foil: counting rate and total counts are plotted. In the lower energy region, the PSD becomes worse because the absolute pulse size is smaller due to lack of light intensity. However in the higher energy region we can clearly reject background events. The counting rate of the background for E > 7 MeV is less than 1 count/day. In order to discuss the observed events statistically, the events are summed up for three bins, 1, 2, and 3, as indicated in Fig. 3. In region 1 of the D_2 spectrum, total counts are clearly larger than the background (H_2 and Vac.).

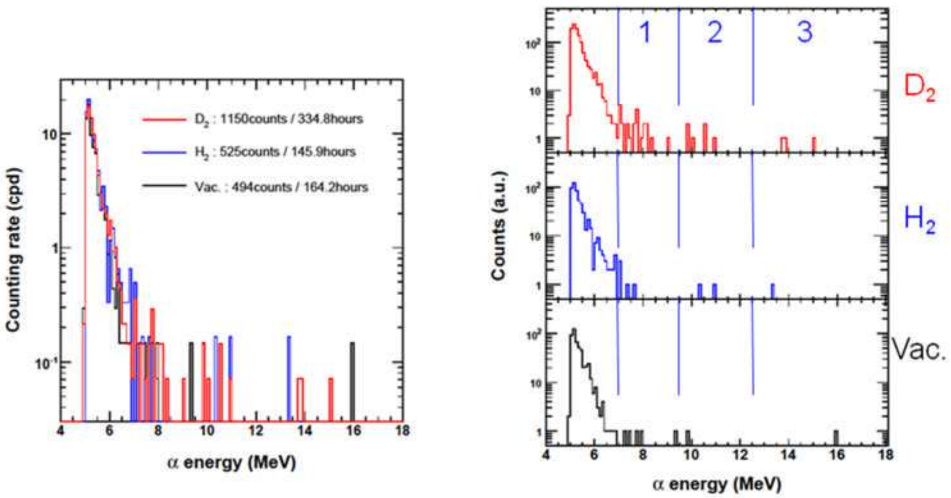

Figure 3. Energy spectra of the PSD events obtained with the I-1 foil. Duration of the D_2 run is 2 times longer than the other cases. Counting rate at lower energy region is not different each other, since background events cannot completely rejected because of smaller pulse shape.

In the present study, we performed the D_2 run first and obtained several counts in region 1. In the H_2 run carried out after the D_2 and Vac. runs, however, no significant count was observed. Then, we retried the D_2 run and observed several additional counts again, in region 1. A total of 22 event counts were observed in region 1, while 9 counts were observed during the background runs (with H_2 and Vac.).

Spectra for the M-1 foil are shown in Fig. 4. Significant events are seen in region 2.

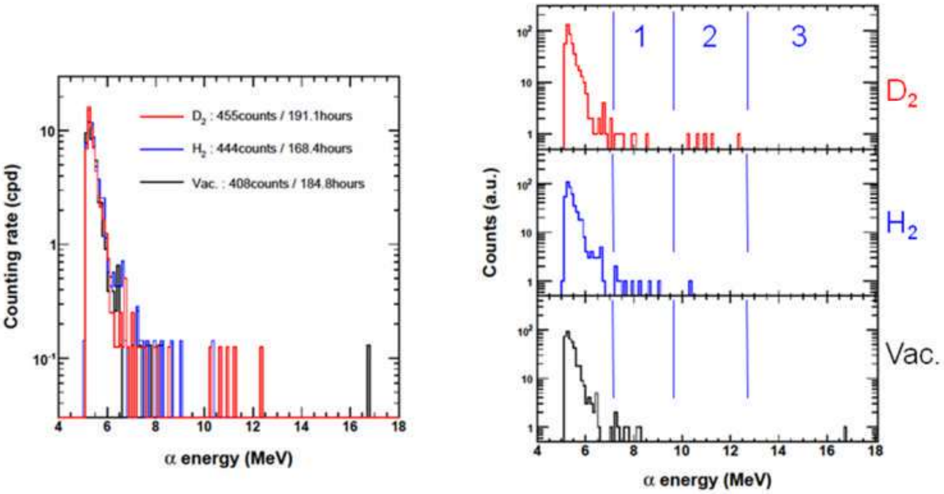

Figure 4. Same as Fig. 3 but for the M-1 foil.

Figure 5 summarizes the deduced counting rate for E > 7 MeV for I-1 and M-1 samples. Red points are data for D_2 runs and black points for background runs (H_2 and Vac.). Vertical error bars with dashed line show 90 % confidence level estimated from the Poisson distribution. As above mentioned, we have significant events in region 1 for the I-1 foil and in region-2 for the M-1 foil. In both cases the error bars of the foreground are not overlapped with those of the background, and the confidence levels are more than 99% in both cases.

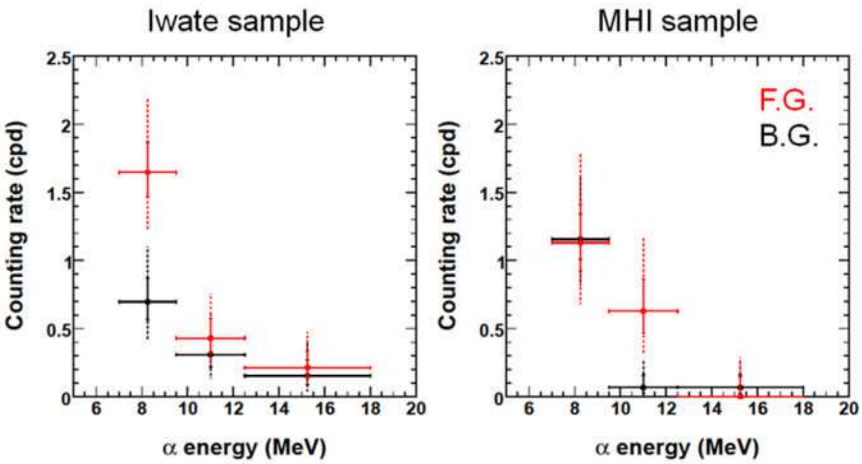

Figure 5. Counting rate for the I-1 (left) and M-1 (right) foils for E > 7 MeV. Data for the D2 run and the background run (H_2 and Vac.) are plotted with red and black points, respectively. Vertical error bars indicate a confidence level of 68.3% (solid line) and 90% (dashed line) derived from the Poisson distribution.

Although we tried 4 samples, significant events of charged particle emission were observed only in the I-1 and M-1 cases. The I-2 foil which has no Cs deposition on the surface showed no emission within the same time period. Although the M-2 foil has a Cs layer, no clear difference was observed between foreground and background. We conjecture that this might be due to a low permeation rate of D_2 gas through the foil: the flow rate was less than 0.1 ccm.

It should be noted that we cannot completely eliminate high energy PSD events in the background runs with this detector system. In other words, there may be several MeV charged particles related to cosmic rays. We have confirmed that such events are not negligible. At present, we consider that these background events are due to elastic recoils of cosmic rays, such as the p(μ, μ)p and d(μ, μ)d reactions, that deposit large energy to the detector. The phoswich detector alone cannot identify these charged particle background events. Although use of the VETO detectors is the only way to eliminate these events, the phoswich detectors cannot be surrounded perfectly in the present work. Therefore an increase of foreground counts is required to improve the reliability with this setup.

5. Conclusion

We have developed the new detector system for charged particle detection in the gas permeation experiment. The YAP(Ce)/BC-444 phoswich detector provides particle identification and VETO detectors, and decrease background events to 1 count/day at several MeV region. This system can identify significant events with a counting rate as low as 3 counts/day for clear evidence of charged particle emission.

Candidate charged particles emitted from the Pd complex foils were observed for the D2 gas permeation with reliability of over 99% in 2 of 4 experiments. These are most likely to be α particles with kinetic energy between 7 and 12 MeV. It should be stressed that these events are caused neither by electronic noises nor by natural radio isotopes. Therefore, we infer that deuteron related nuclear reactions occurring in the sample emit these charged particles. At present, the number of the observed events is so small that the reaction mechanism can hardly be inferred. We conjecture that high permeation rate could induce high reaction rate and, thereby, material improvement are strongly required for the next step.

Acknowledgement

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