

ICANS XIX,  
19th meeting on Collaboration of Advanced Neutron Sources  
March 8 – 12, 2010  
Grindelwald, Switzerland

## GAS DESORPTION EXAMINATION OF B<sub>4</sub>C RESIN FOR NEUTRON VACUUM CHAMBER

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### ABSTRACT

Gas desorption rate of several types of B<sub>4</sub>C resin was measured by means of throughput method. In particular, the test was focused on investigating gas type, effects of dry-air, grain size (density) effects, and so forth. It is found that water is main contribution to out gas, also dry-air can effectively reduce desorption.

### 1. Introduction

Some neutron instruments, particularly pulse neutron instruments, require a large volume of vacuum chamber (VC) in its neutron flight path. The role of the chamber is not only to avoid the air scattering, also to utilize the chamber as OVC for refrigerators of sample environment (SE) equipments. The dimension of the VC quite depends on required instrument performance. In the case of direct geometry chopper spectrometer, size of the VC trends toward larger, L2 (sample-detector distance) is almost 3-5m. Thus, the chamber volume is estimated to be 30-50m<sup>3</sup>. In this chamber, the inner surface covered with neutron absorber as a liner is usually going to be 100-200m<sup>2</sup>, which yields the enormously large amount of out gas. Some instruments may, moreover, need other components such as vanes, SE equipments that also give rise to out gas. In particular, using the chamber as OVC for refrigerator, its vacuum is required relatively high the order of 10<sup>-4</sup> to 10<sup>-5</sup> Pa. Reasonably large vacuum pumps and controlling system are necessary to operate the smooth experiments. We have performed the gas desorption rate measurements for several types of B<sub>4</sub>C and resin to figure out the reasonable conditions that *effectively shield neutrons with minimum out gas*. This could also give a guiding as select the vacuum system. In this report, the following items are aimed principally in the tests.

- (1) Following the realistic conditions of neutron experiments
- (2) The effects of the dry air
- (3) The grain size (density) difference of the B<sub>4</sub>C mixture

## 2. Experimental Conditions

### 2.1. Throughput method

We define “volume gas”  $pV$  [Pa m<sup>3</sup>] with gas pressure  $p$  and volume  $V$ . A time difference of the volume gas corresponds to gas flow  $Q$ . The  $Q$  is, therefore, defined as follows,

$$Q \equiv V \frac{dp}{dt} + p \frac{dV}{dt} \quad (1)$$

Suppose that a vacuum pump with pumping speed  $S$  is used for evacuating a chamber,

$$\frac{dV}{dt} = S \quad (2)$$

The Eq. 1 is rewritten as,

$$V \frac{dp}{dt} = Q - pS \quad (3)$$

This is called equation of evacuation. We now consider the situation “the evacuating speed is not zero, and the time difference of the pressure in the chamber can be negligible.” as realized in the quasi-stable state,

$$V \frac{dp}{dt} \approx 0 \quad (4)$$

then, the Eq. 3 is

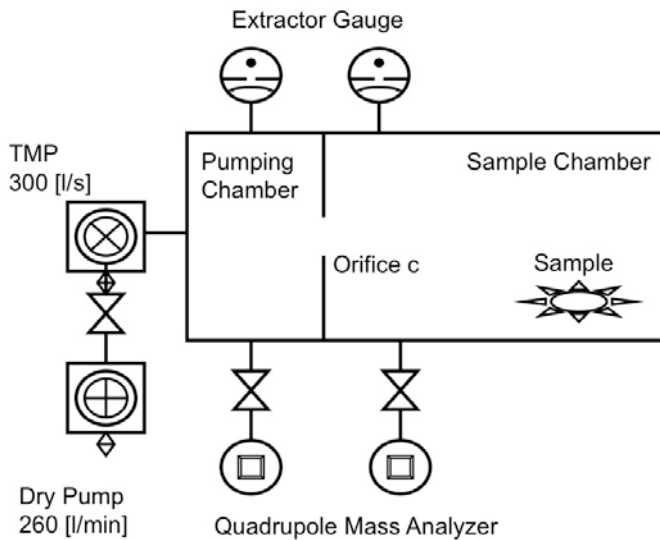
$$q = \frac{P}{A} S \quad (5)$$

with normalized gas flow  $q$  ( $Q = Aq$ ). This is known to be gas flow of throughput.

### 2.2. Measurement devices

The measurement system used in this report is shown in Fig. 1. Two chambers (pump chamber and sample chamber) are connected by an orifice its conductance is determined. The vacuum is measured for both chambers in cases with/without sample. Time dependence of the vacuum without sample is

$$V \frac{dp_{wo}}{dt} = Q_c - c(P_2 - P_1) \quad (6)$$



Here,  $c$  is the conductance of orifice and  $P_1, P_2$  correspond to the vacuum of the pump and sample chamber, respectively. In the case with sample, it is similarly described with gas out of sample  $Q$ ,

$$V \frac{dp_w}{dt} = Q + Q_c - c(P'_2 - P'_1) \quad (7)$$

Considering the limit of high vacuum,

$$\frac{dp}{dt} = \frac{dp_{wo}}{dt} = \frac{dp_w}{dt} = 0 \quad (8)$$

Therefore, the out gas  $Q$  is measured as the following relationship,

$$Q = c(P'_2 - P'_1) - c(P_2 - P_1) \quad (9)$$

Fig. 1 Vacuum measurement system by throughput method

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**2.3. Samples**

The tests were carried out by two series. In the first experiment I, the sample used is listed in Table I. And Table II is the samples for the experiment II.

Table I : Samples measured in experiment I

	Dimension [mm <sup>3</sup> ]	Grain mixing ratio	Binding
Sintered B <sub>4</sub> C	25×5×100	N/A	N/A
Flexible B <sub>4</sub> C	20×33×6	F20(35%)+F40(52%)+F60(3%)	10% (SB)
ISIS B <sub>4</sub> C	29×24×18	unknown	5%
Inorganic B <sub>4</sub> C resin	58f×13	F20(36%)+F40(54%)+F60(3%)	7% (Inorganic)
Crispy B <sub>4</sub> C	75×10×6	F20(38%)+F40(56%)+F60(3%)	3% (SB)
Epoxy (SB)	40f×4	N/A	N/A
Epoxy (SB) (sintered)	40f×4	N/A	N/A

The experiment I is focused on the out-gas measurement test for several types of the sample containing various ratio of bindings. Sintered B<sub>4</sub>C shows great advantage for out gas, though it is not reasonable price for wide coverage. Flexible B<sub>4</sub>C is the mixing ratio, which normally used for the neutron experiments. ISIS B<sub>4</sub>C is the specimen used in ISIS, delivered from Brookvine. For inorganic B<sub>4</sub>C resin, the inorganic resin is used for the binding instead of usual SB. Only 3wt% binding is used for Crispy B<sub>4</sub>C. Epoxy is simply the piece of SB and one of them is sintered by 120°C in an oven. Typical samples are pictured in Fig. 2(a) and 2(b), for Flexible B<sub>4</sub>C and Sample 5, respectively. The “F” number in the grain mixing ratio represents the diameter of B<sub>4</sub>C grains. For instance, F20 corresponds to the diameter range within 850µm to 1180µm, and the case of F40 corresponding grain range is 355-500µm.

For the experiment II, the sample volume was unified, but with different types of mixing ratio, densities. In this second test, we focused on the difference of desorption rate by the variety of sample density.

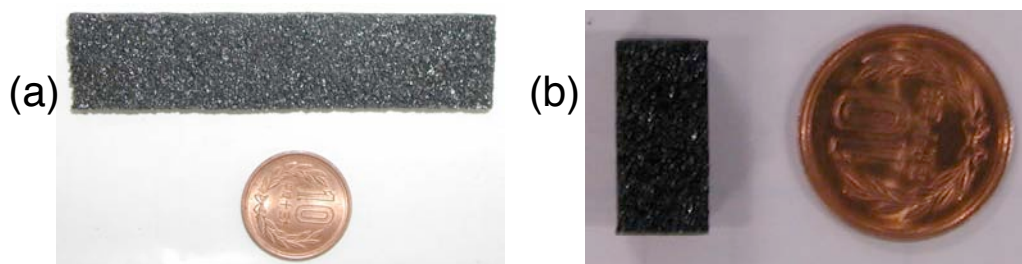


Fig. 2 The samples used in measurements: (a) Flexible B<sub>4</sub>C resin with 10yen coin (10yen coin diameter is 23.5mm), (b) Sample 5 used in experiment II

Table II : Samples measured in experiment II

	Sample volume [cm <sup>3</sup> ]		Density [g/cm <sup>3</sup> ]
Sample 1	92.3	F20(39%)+F40(58%)+F60(3%)	1.37
Sample 2	92.3	F20(29%)+F40(47%)+F60(24%)	1.40
Sample 5	92.3	F20(0%)+F40(14%)+F60(86%)	1.32
Sample Ex2	92.3	F20(0%)+F40(100%)+F60(0%)	1.29

#### 2.4. Experimental procedure

In order to obtain the gas desorption rate under the real neutron experimental condition, we do not necessarily follow a general way. In particular, the following items are originally modified from the way.

- (1) Baking: Baking process of vacuum chambers is essentially indispensable for reduce the out gas from the sample. It is, however, impossible to do so in the real huge chamber, we omit this process.
- (2) Measurement time: It is mechanically hard to measure the short time region (~1 hour) after starting the measurement. In this experiment, measurement time is defined from 1 to around 150 hours.
- (3) Dry-air: It is obvious that using dry-air for opening air the tank has great advantage to suppress adsorption of water in the tank. We tried to clarify the dry-air effects quantitatively.
- (4) Grain size: Also we prepared several types of samples mentioned above, namely, the different  $B_4C$  grain size mixture. This yields difference of density of  $B_4C$  resin. We surveyed the density dependence.
- (5) Base line: Base line is defined as desorption rate without any sample such like background. The gas desorption rate from the sample is obtained by subtracting the base line data from the measured sample data. For the series of measurements (whether using dry-air or not), no big difference has been observed.

Considering above things the first and the second experiment were carried out with the following procedure.

Experiment I:

- After open air the chamber (about 10min.), place the sample.
- Start pumping following leak check, and then start measurement (150 hours)
- Dry-air flow (about 10min.). Start re-measurement.

Experiment II (Dry-air effects):

- Open air for several days, pumping for each pumping time
- Purge with dry-air, start measurement

### 3. Results and Discussion

#### 3.1. Base line measurement

In Fig. 3(a) and 3(b), time evolution of vacuum in the two chambers divided by orifice and

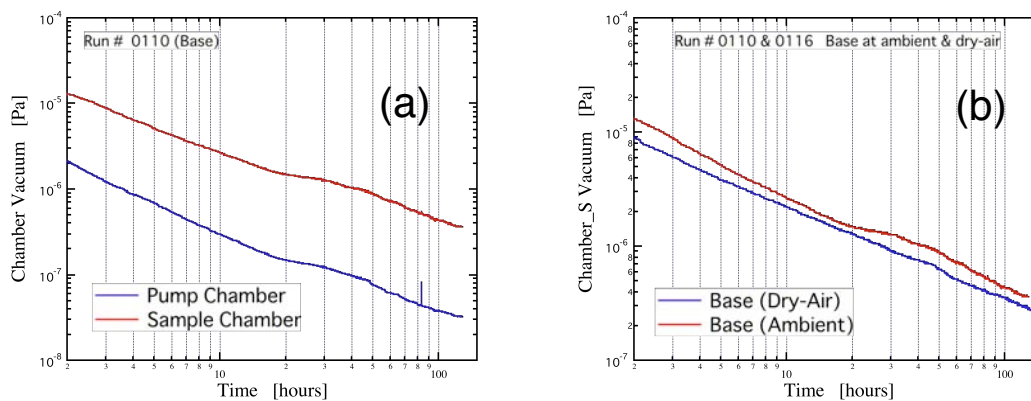


Fig.3: (a) vacuum for both sample and pump chamber, (b) comparison between ambient and dry-air pumping

base line vacuum with/without dry-air are shown. Time dependence of each chamber (chamber-Sample and –Pump) seems almost identical except for the difference of one digit, and one can see the vacuuming process is properly executed. No large effects on the vacuum with/without dry-air have been observed.

### 3.2. Out gas from Bindings

Figure 4 shows gas desorption rate of epoxy resins. A resin without sintering treatment shows enormous out gas whether using dry-air or not as shown in Fig. 4(a). Even sintered treatment of resin shows better out gas (Fig. 4(b)). Quadrupole mass spectrometer measurement reveals a main component in out gas of water as shown in Fig. 4(c). This is quite consistent with that after the sintered resin shows much less gas desorption (sintering has been done with 120°C heating for 2 hours as mentioned above).

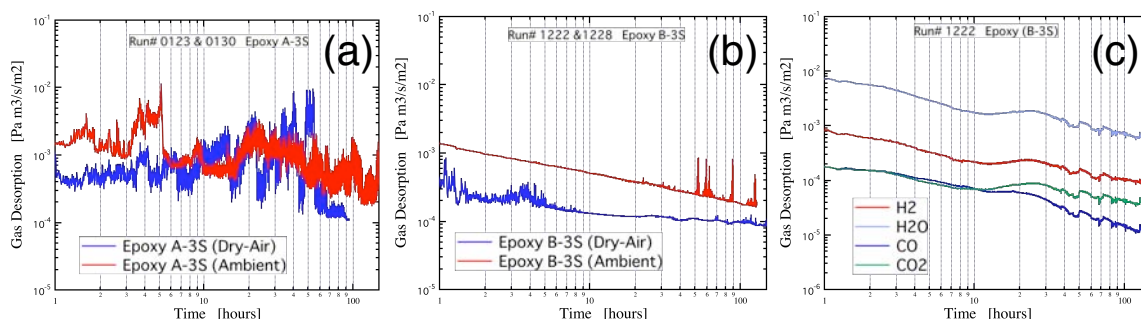


Fig. 4: (a) gas desorption from epoxy resin, (b) sintered epoxy resin, (c) partial pressures of out gas detected quadrupole mass spectrum.

### 3.3. Results of experiment I

On various types of mixture of B<sub>4</sub>C and resin, gas desorption rate (GDR) have measured. In Fig. 5(a) and 5(b) the results are summarized with the case of ambient air and dry-air, respectively. Crispy type B<sub>4</sub>C and ISIS B<sub>4</sub>C shows rather good GDR. Inorganic B<sub>4</sub>C shows relatively high GDR because of not only the large size of the sample but also the large amount of resin. Therefore, the only minimum resin can be used in order to reduce the gas desorption. Dry-air also much affects reduction of GDR for any types of B<sub>4</sub>C except for sintered. Since porous B<sub>4</sub>C resin contains much water in it, dry-air can prevent the re-adsorption of water molecules.

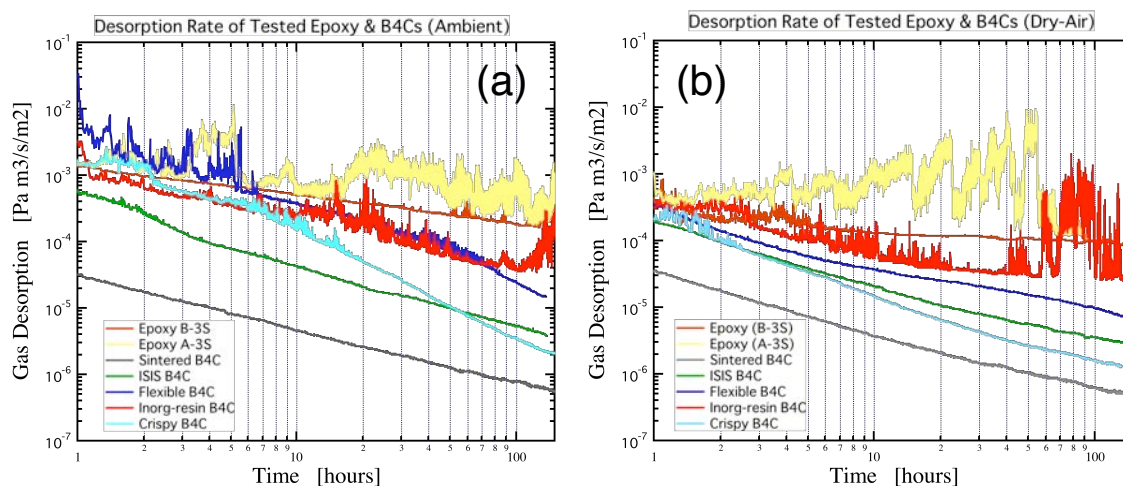


Fig. 5: summary of GDR with (a) ambient and (b) dry-air results



### 3.4. Results of experiment II

The results of experiment II is shown in Fig. 6(a) and 6(b) in the same way as experiment I. All the samples show similar properties, but with a little difference. Initial value as pointed by ① in the figure should in principle identical. The distribution is thought to be a sort of an error. In the section ③ (asymptotic region), we do not find any remarkable differences between ambient and dry-air. However, comparing Sample 2 and SappmleEx 2 at one hour, 27.8% difference in GDR is observed, which is dominant to the 8.5% density difference.

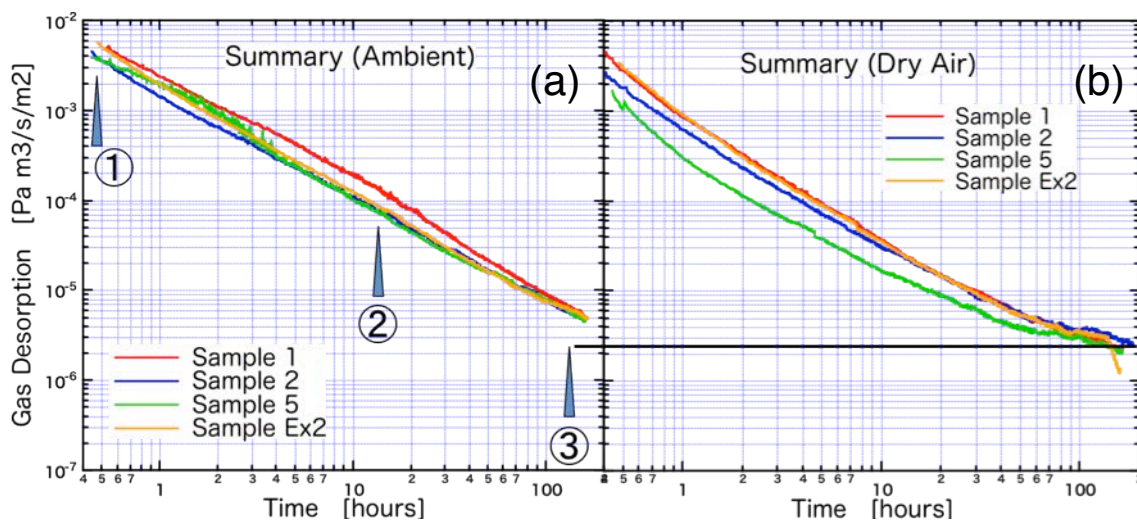


Fig. 6: GDR of the second tests (a) with ambient and (b) with dry-air

In the following, we demonstrated tests for dry-air effects with the following way.

- (1) Open air: for several days to be the same starting condition
- (2) Pre-pumping: for each pumping time
- (3) Purge: purge by dry-air up to 1 atm.
- (4) Pumping (measurement): for around 120 hours

Obtained GDR for each pumping time is summarized in Fig. 7. Difference of the pre-pumping time corresponds to the effect of dry-air. Plateau (or dip) regions appear on pre-pumping data, whereas no such behaviour has been observe on without pre-pumping. Also, plateaus seem to be proportional to pre-pumping time. After plateau region, all the curves recover to the original (no pre-pumping) GDR curvature. Again, this is a quite visible dry-air effect result suggesting introducing a dry-air system is indispensable for saving time.

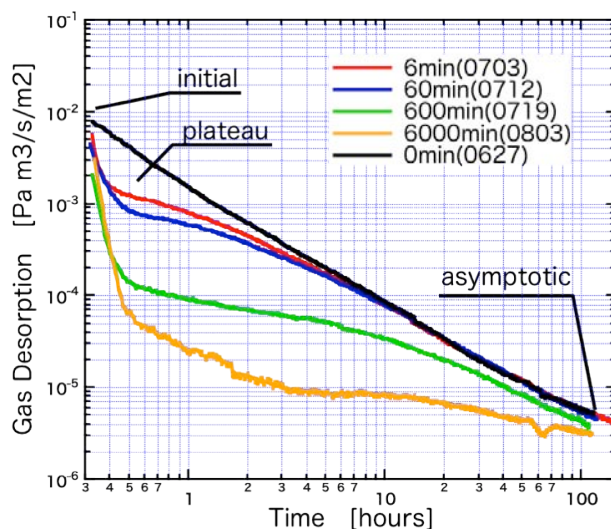


Fig. 7: Dry-air effects.

### 3.5. Discussion

In Fig. 7, we can see a systematic trend. We define some features as indicated in the inset of Fig. 8. The value (a) is defined as a GDR and (b) is the pumping time at plateau region. (c) is the time that gas desorption of 0min. (no pre-pumping and purging by dry-air) data becomes the same order at plateau. Figure 8 is the plot of these values as a function of pre-pumping time. (a) decreases with increasing pre-pumping time, directly reflecting the reduction of water molecules in the chamber by pre-pumping. (b) defined as roughly the center of plateau seems to be almost constant. (c) rapidly increases. This means time gain to achieve the pumping time. If it is retained the “pre-pumped vacuum” by dry-air, you can save this time gain for your experiments.

Now, we discuss about the water out gas in the following. If you start the purge of VC, humid ambient air gives rise to adsorption of water molecules on the VC surface. According to Ref. 1, using the dimensionless equation of evacuation,

$$V \frac{dn}{dt} \equiv Aq^* - \frac{av}{4}n \quad (10)$$

covering ratio of water molecules are estimated. The left side of Eq. 10 is the time shift of the number of water molecules. The second term in the right side is the number of water going through the orifice with  $a$ .  $q^*$  stands for GDR from the wall surface and expressed as follows,

$$q^* = \sigma_m \left( -\frac{d\theta}{dt} \right) \quad (11)$$

with  $\sigma_m = 3 \times 10^{15}$  [mol/cm<sup>2</sup>] and water covering ratio  $\theta$ . Considering remaining meantime of molecules, Temkin type heat of adsorption, then time evolution of the water molecule coverage can be calculated as shown in Fig. 9. The coverage  $\theta$  is plotted as a function of pumping time in second. To reduce 80% of the water coverage, it is estimated to take 10<sup>7</sup>s, which is about 3.9 month!

Obtained time dependence of (a) in Fig. 8(a) shows quite similar properties to the calculation (Fig. 9). From this view point, the parameter (a) can be the value corresponding to the gas desorption at pumped for corresponding time.

### 4. Conclusion

Gas desorption rate of several neutron shielding B<sub>4</sub>C resin has been investigated. Main contribution of out gas turned out water from B<sub>4</sub>C and resin. The resin is particularly undesirable form this point. Also, the difference of B<sub>4</sub>C density and effects of dry-air have been

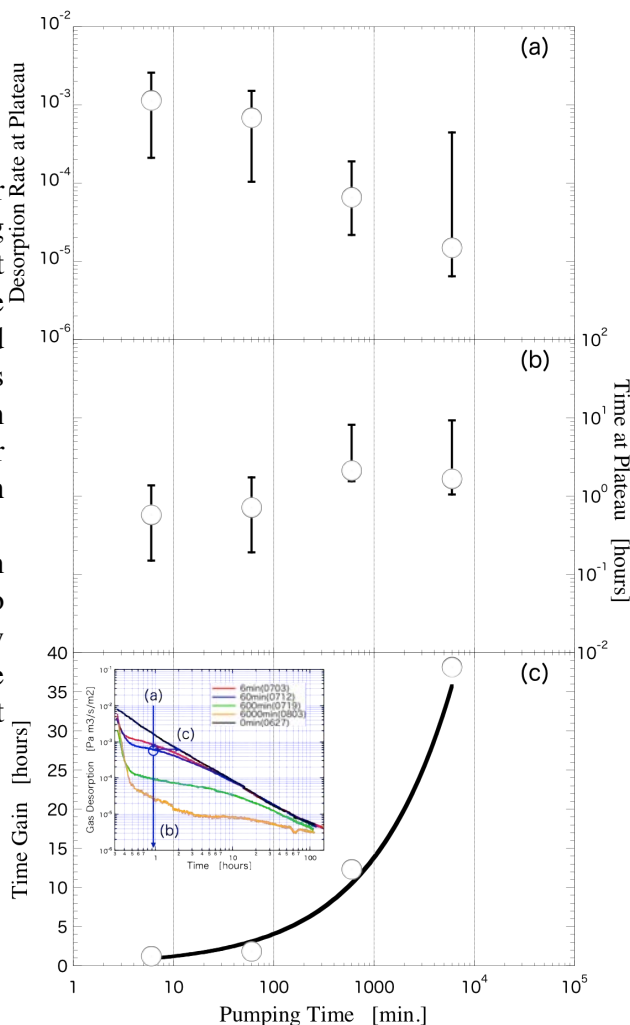


Fig. 8: Pumping time dependence of each reduced parameters. Each parameter is defined in the inset.

tested. We found smaller out gas is released from a larger density sample. Its gain seems to be not so much. However, a large density with small out gas is obviously better for neutron shielding. Purge with dry-air much affects on the gas desorption, and it is worth considering to be installed.

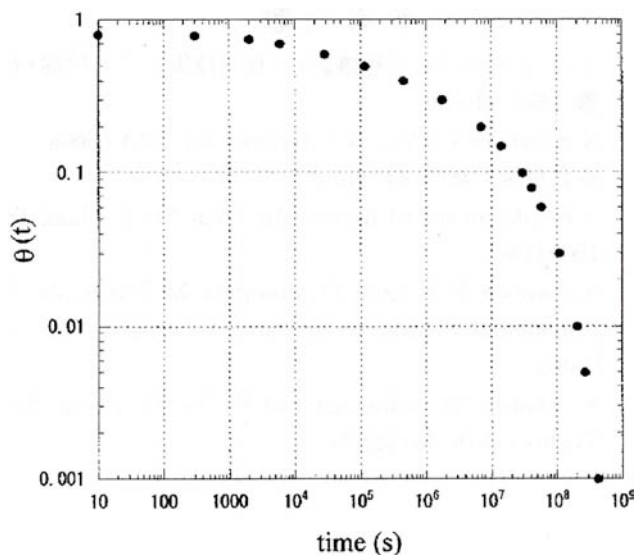


Fig. 9: Calculated water coverage as a function of pumping time. This is from Ref. 1

## Acknowledgements

One of authors TY acknowledges a partial support by the Ministry of Education, Science, Sports and Culture, Grant-in-Aid for Scientific Research (C).

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