Error analysis in a D(H)/Pd gas-loading system when using isothermal calorimetry

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Abstract. In the field of low energy nuclear reaction, isothermal calorimetry is often used to calculate the excess heat power released in the system. Due to the structure of the reaction chamber and the differences of the thermal conductivities of the gases used in the calibration and triggering experiments, the chamber will have different temperature gradient or distribution which leads to errors when calculating the excess heat power using isothermal calorimetry. Different gas pressure also has an influence on the heat transfer.

1. Introduction
The abnormal phenomenon that metal lattice absorbed in a certain amount of deuterium or hydrogen atoms under certain conditions could be released excess heat exceeding chemical energy in the field of condensed matter nuclear science continues to attract the attention and research from scientists since 1989[1]. Scientists and researchers have tried to use different ways to get excess heat energy in many systems[2]. For the existing or newly proposed theories do not match with experimental results well[3], instability and unsatisfactory repetition rate of experimental results have caused many scientists to hold pessimistic and negative attitudes towards the research[4,5]. Calorimetry plays an important role in the thermal measurement of low energy nuclear reaction. Using the wrong calorimetry may lead to wrong results. Even today, there are still some people[6] questioning the calorimetry Fleischmann and Pons used in their experiments. In previous work[7,8], approximately one hundred watts of excess heat was measured in a deuterium/palladium gas-solid system by using an isothermal calorimeter. Take the differences of the thermal conductivities of the gases used in the calibration and triggering experiments and the structure of the reaction chamber into consideration, there would be different temperature distribution or gradient in the reaction chamber. Beside this, the gas pressure also have influence on the heat transfer modes which also leads to a different temperature distribution. Therefore, using temperature to calculate the thermal power released by the system would produce errors. In order to re-examine the previous work, the sample was changed into inert materials and a series of calibration experiments with different types of gases and pressures were performed.
2. Experimental

2.1. Materials and Apparatus
Because the system in previous work is unavilible, a new system which was similar to the old was constructed. Figure 1 is the schematic diagram of the gas-loading system and figure 2 is the schematic diagram of the reaction chamber. In Figure 1: where 1-vacuum valve; 2-Kf25 negative pressure gauge; 3-four-way connection; 4-GL/4 positive pressure gauge; 5-aviation joint; 6-Cf35 apron angle valve; 7-reaction chamber; 8-transition pipe; 9-vacuum gauge; 10-molecular pump; 11-bench; 12-vacuum mechanical pump; 13-hydraulic bellows I; 14-hydraulic bellows II; 15-molecular sieve. In figure 2: where 1-circulating water inlet; 2-ceramic tube; 3-heating wire; 4-ceramic tube; 5-experimental palladium wire; 6-circulating water outlet; 7-stainless steel reaction chamber. The 3 and 5 are wound on ceramic tubes respectively. T1 to T4 are Pt-100 platinum resistance thermometers placed inside the reaction chamber to measure the temperatures.

![Figure 1. Schematic diagram of the system](image1)

![Figure 2. Schematic diagram of the reaction chamber](image2)

2.2. Calibration
In order to verify whether the difference in thermal conductivity of the gas caused the system measuring excess heat power, the experimental palladium wire in the system was removed, hydrogen, argon and nitrogen gas were selected for calibration experiments. The system was calibrated by 1 atm hydrogen, argon and nitrogen respectively. Figure 3 shows the fitting curves and the relationship between applied power and temperature.

![Fitting curves and relationship between applied power and temperature](image3)

(a) T4-sample  (b) T2-heating wire
3. Analysis and discussion

Generally speaking, because the thermal conductivity of hydrogen is much higher than that of nitrogen and argon, when the calibration experiment is carried out, the temperature at the same point in hydrogen calibration will be lower than that of nitrogen and argon. In figure 3, T1, T2 and T3 fit this rule. Figure 3(b) shows that the thermometer is placed between the two ceramic tubes, which is closest to the heating source, so it has the highest temperature. Figure 3(c) shows the temperature above the ceramic tube, which is the same rule because the heat transfer is not blocked. Figure 3(a) shows the temperature of the reaction wire. The ceramic tube is also a material with low thermal conductivity. Most of the heat flow is directly conducted up or down due to the obstruction of the ceramic tube. However, when the input power is over 350 W, the temperature of T4 is higher than that of nitrogen and argon calibration. This is because the convection heat transfer increased gradually. With the increase of temperature (the increase of input power), hydrogen transfers more heat than nitrogen or argon. Because T1 and T3 are blocked by ceramic tubes, even in the case of nitrogen or argon calibration, the heat transferred through the ceramic tube wall will be less than the heat transferred by the gas when the heating source has a higher temperature. This leads to an abnormal temperature in T1. It can be concluded that the structure of the reaction chamber has an important influence on the temperature gradient and distribution. If this effect is not taken into account, these temperature differences will be calculated as excess heat power in the triggering experiment, resulting in large errors in the experimental results.

Figure 3 fitting curves and the relationship between applied power and temperature

Figure 4 The fitting curves and the relationship between pressure and temperature at 380 W applied power in hydrogen calibration
In the calibration experiment of 380 W input power and different hydrogen pressure, we focus on the temperature change of T2. When the pressure is higher than 10 kPa, the temperature of T2 increases with the increase of pressure. In the pressure range of 10 kPa to 100 kPa, the thermal conductivity of hydrogen is almost not affected by pressure, but the high pressure gas will affect the heat radiation transfer. When the pressure is less than 10 kPa, the effect of gas heat conduction gradually decreases, and approaches zero at low pressure. The proportion of heat radiation is increasing, resulting in lower pressure and higher temperature. At the same time, there is another effect that exacerbates this phenomenon. Due to the use of alumina ceramic tube, some micro pores may appear in the ceramic tube during the making process. Due to the Knudsen effect, when the molecular free path of the gas is greater than or close to the porosity linearity, the thermal conductivity of the porous material will decrease significantly. This effect may also be one of the reasons for this phenomenon. Therefore, with the change of air pressure, there is a combined convection and radiation heat transfer process in the reaction chamber, which has a certain influence on the temperature gradient and distribution. Because the results of reaction chamber and thermometer are not the same, there may be more significant temperature difference under some specific pressure and some certain points, resulting in the calculation of greater excess heat power. This result has a great error, and researchers need to pay special attention to this point.

In the research field of low energy nuclear reaction, because the structure of the reaction chamber and the position of the thermometer are also different, researchers should pay special attention to this problem when using the temperature to calculate the excess heat power, otherwise they may get wrong results. Through experiment results, we have learned the cause of this error, and some advises are given to reduce the error:

1. The calorimeter should be calibrated with the same type of gas use in triggering experiment, change sample into inert materials or remove it in the calibration.
2. The pressures should be the same in the calibration and triggering experiments.
3. Use other calorimetry, such as heat-flow (Seebeck) calorimeter.

4. Conclusion
Due to the different thermal conductivity of gases used in calibration and experiment and the structure of reaction chamber, complex heat transfer process occurs in the reaction chamber. Finally, there are different temperature distributions and gradients in the reaction chamber under different types of gases and pressures. The experimental results may have large errors when the excess heat power of the system is measured by isothermal calorimetry. Researchers in the field of low energy nuclear reaction need to pay special attention to this problem and put forward some suggestions to reduce the error.

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