Development of Hydrogen Sensor with Solid Conductor for In-pile Measurements in Neutron Irradiation Tests

N. Otsuka [1], S. Kitagishi [1], T. Saito [1], Y. Endo [1]
C. Yamamura [2], K. miura [2], Y. Matsui [1] and K. Tsuchiya[1]

1: Japan Atomic Energy Agency, 4002 Narita, Oarai, Higashiibaraki, Ibaraki 311-1393, Japan
2: Sukegawa Electric Co., Ltd, 3-19-5 Namekawa-honchoi, Hitachi, Ibaraki, 317-0051, Japan

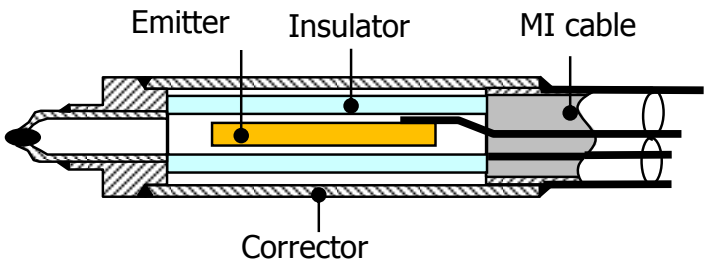
Introduction

The JMTR is one of the foremost testing/research reactors and is used for contribution to many nuclear research and development.

To get data with high accuracy for fuel and material behavior studies in irradiation tests, we have developed many types of measuring instruments such as LVDT, SPND and gas sensors with solid conductors. In particular, the LVDT and SPND can be used to get the data with high accuracy under neutron irradiation environment. On the other hand, the irradiation tests of the hydrogen/oxygen gas sensors with solid conductors were carried out in JMTR, however the data with high accuracy was not obtained, yet.

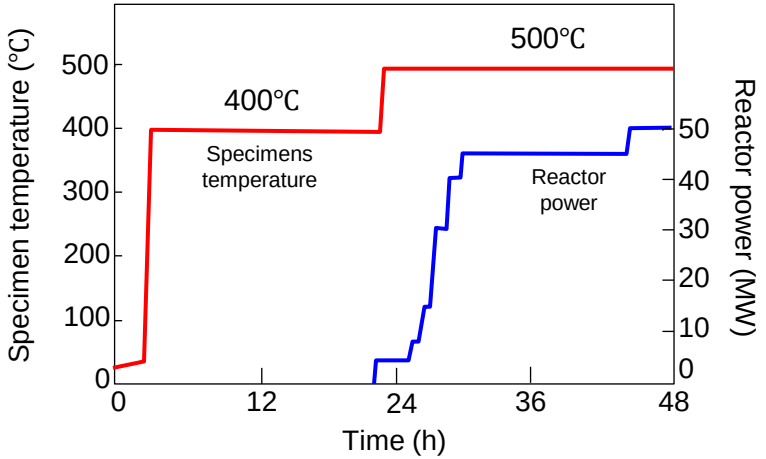
Background

Measurement instruments



- Self-Powered Neutron Detector (SPND)
- Linear Voltage Differential Transformer (LVDT)
- Gas sensor with solid conductor

Irradiation technique



- Temperature control technique
- Environmental control technique
- Neutron control technique

We started to develop the hydrogen sensor for high-temperature and high-radiation conditions with high accuracy.

Development of Hydrogen sensor with solid conductor

The following two works were done
for development of Hydrogen sensor

1. Out-pile test of Hydrogen Sensor with Solid conductor

Electronic motive force with H2 concentration was measured to research the characteristic of hydrogen sensor under un-irradiation conditions.

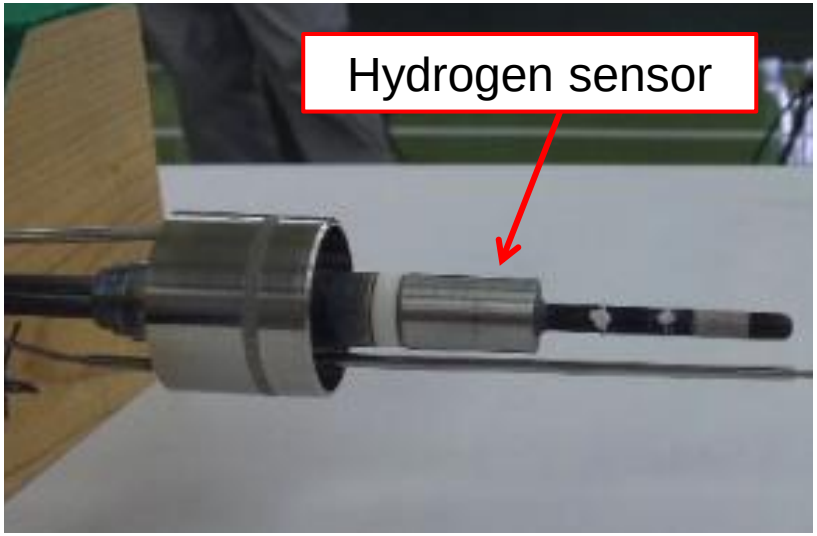
2. Fabrication of irradiation capsule for the hydrogen sensor

irradiation capsule was estimated by GENTC code to confirm the fabricated capsule is able to control temperature of the hydrogen sensor from 400℃ to 600℃.

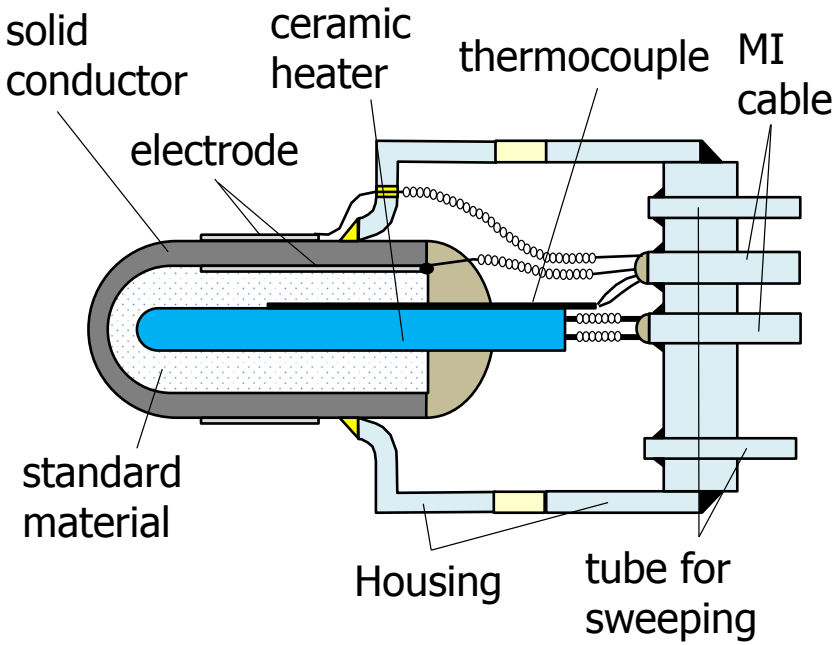
Configuration of Hydrogen sensor with Solid conductor

Specification of hydrogen sensor

Item	specification
Operating Temperature	400～600 (℃)
Measurement range of H_2 concentration	0.01～1 (%)
Environment in irradiation test	He
Fast neutron flux	1.91×10^{16} ($m^{-2} \cdot s^{-1}$)



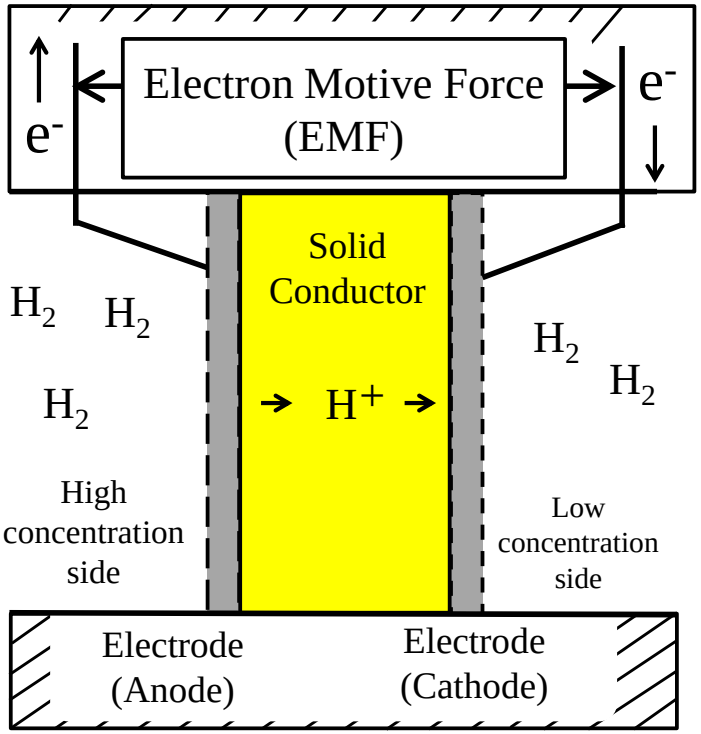
Structure of Hydrogen sensor



No.	Parts	Materials
1	Solid conductor	$CaZr_{0.9}In_{0.1}O_{3-\alpha}$
2	Electrode	Platinum
3	Standard material	$AlPO_4 \cdot xH_2O$ $La_{0.4}Sr_{0.6}CrO_{3-\alpha}$
4	Ceramic heater	Al_2O_3
5	thermocouple	chromel-alumel

Principle of hydrogen measurement

Concept of hydrogen sensor with Solid conductor



Reaction on electrodes

Anode side : $2H^+ + 2e^- \rightarrow H_2$

Cathode side : $H_2 \rightarrow 2H^+ + 2e^-$

Nernst Equation

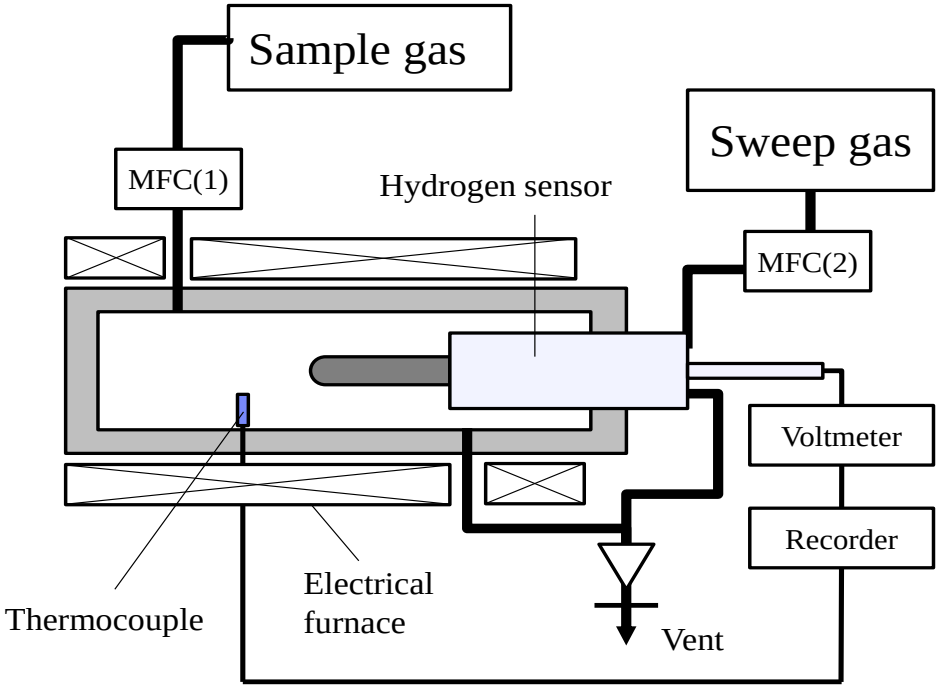
$$E = \frac{RT}{2F} \ln\left(\frac{P_{H2}(1)}{P_{H2}(2)}\right)$$

E : Electron Motive force(EMF)
R : gas constant
T : Temperature of hydrogen sensor
$P_{H2}(1)$: H_2 concentration in environment
$P_{H2}(2)$: Inner H_2 concentration of Hydrogen sensor

Temperature	Inner H_2 concentration
400˚C	3.84×10^{-11} ppm
500˚C	1.11×10^{-8} ppm
600˚C	7.64×10^{-7} ppm

Experiment for Out-Pile test

experiment device

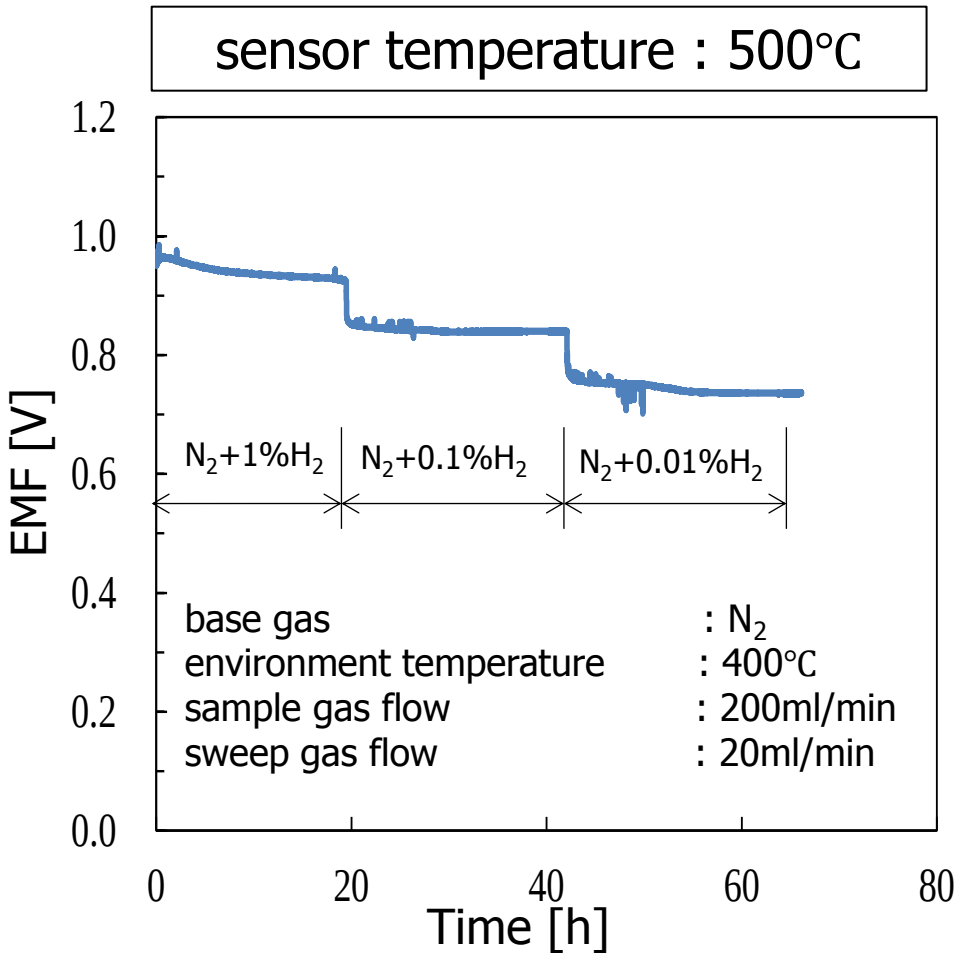


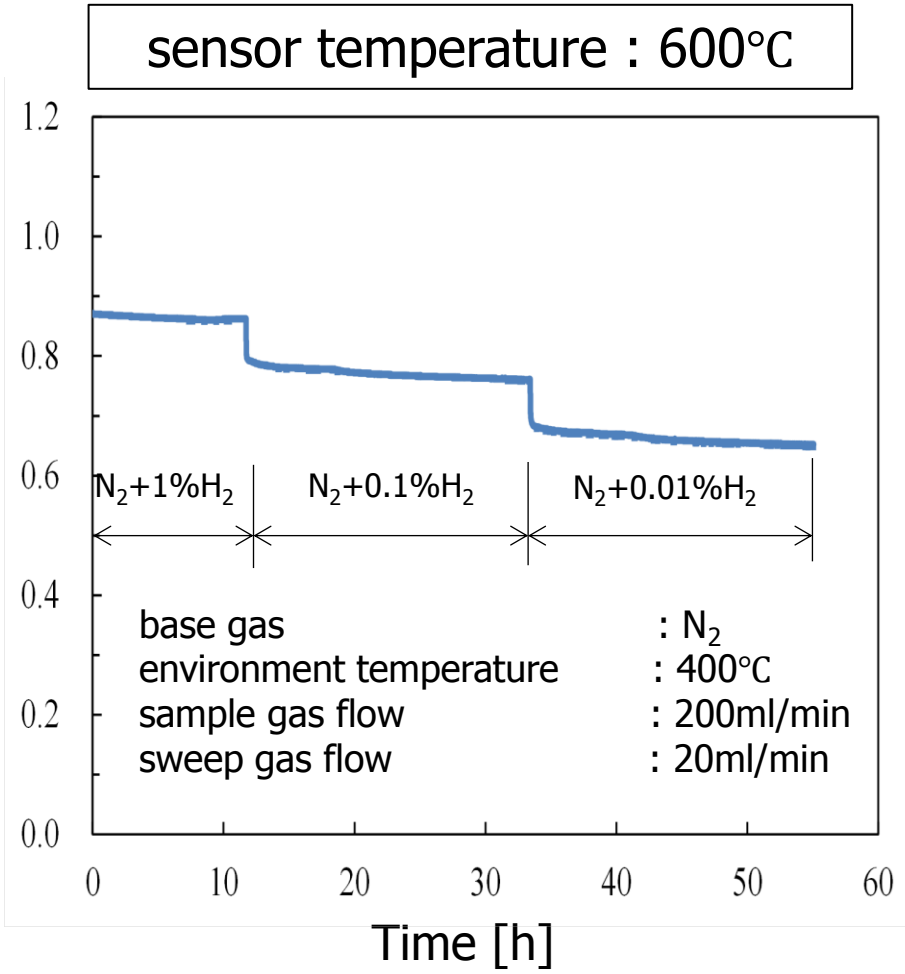
Composition of Sample gas

Base of Sample gas	H_2 concentration in Sample gas
He	0.01, 0.1, 1%
N_2	0.01, 0.1, 1%

The experiment device was prepared to research the characteristic of hydrogen sensor under un-irradiation.

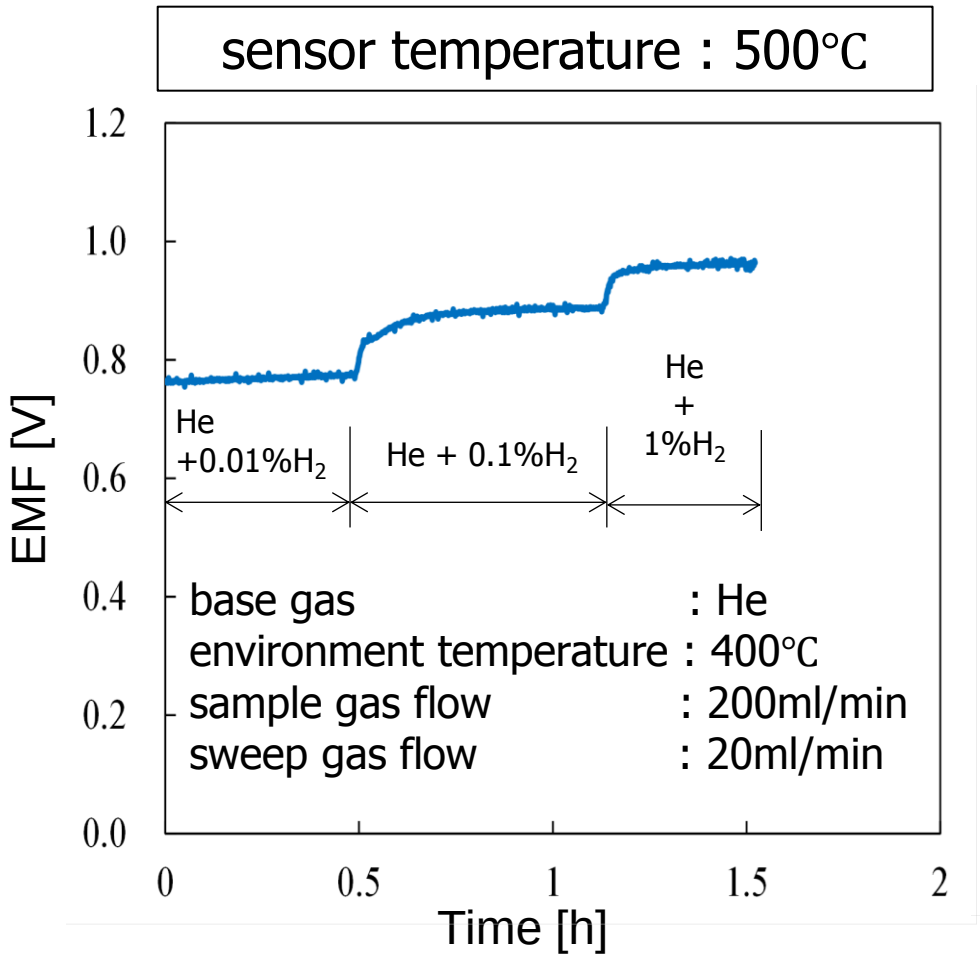
Results of EMF under N_2+H_2 environment

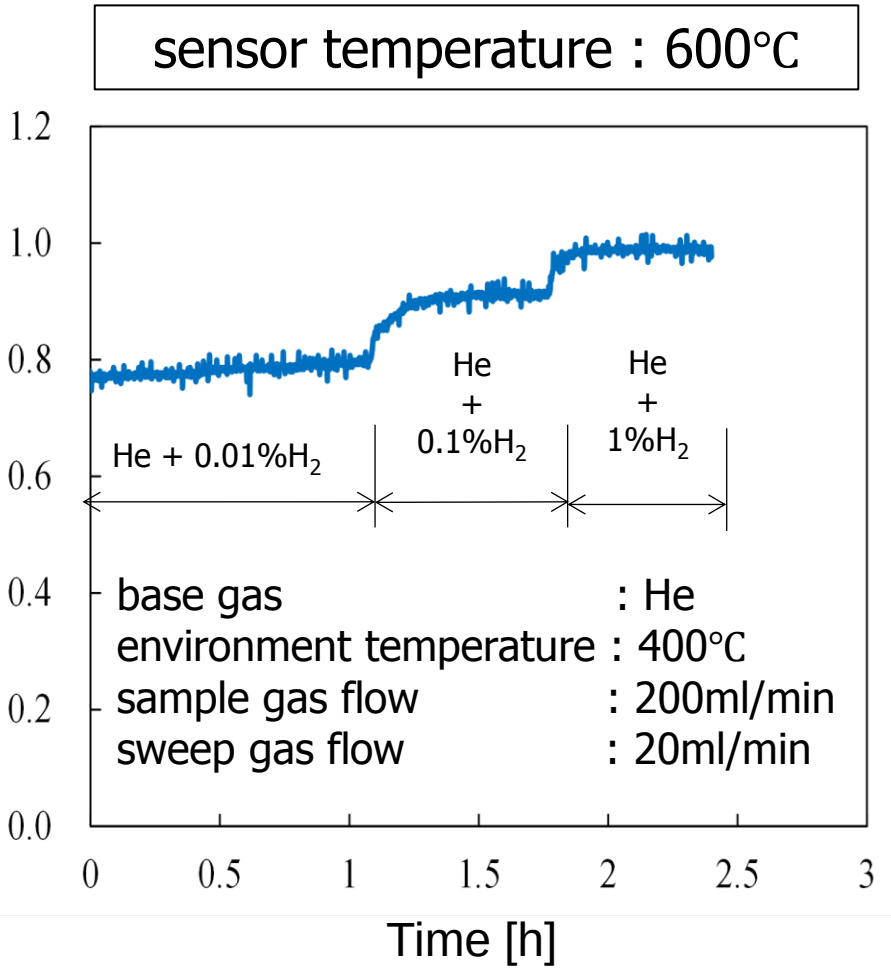




EMF of the hydrogen sensor was decreased with H_2 concentration change at each temperature

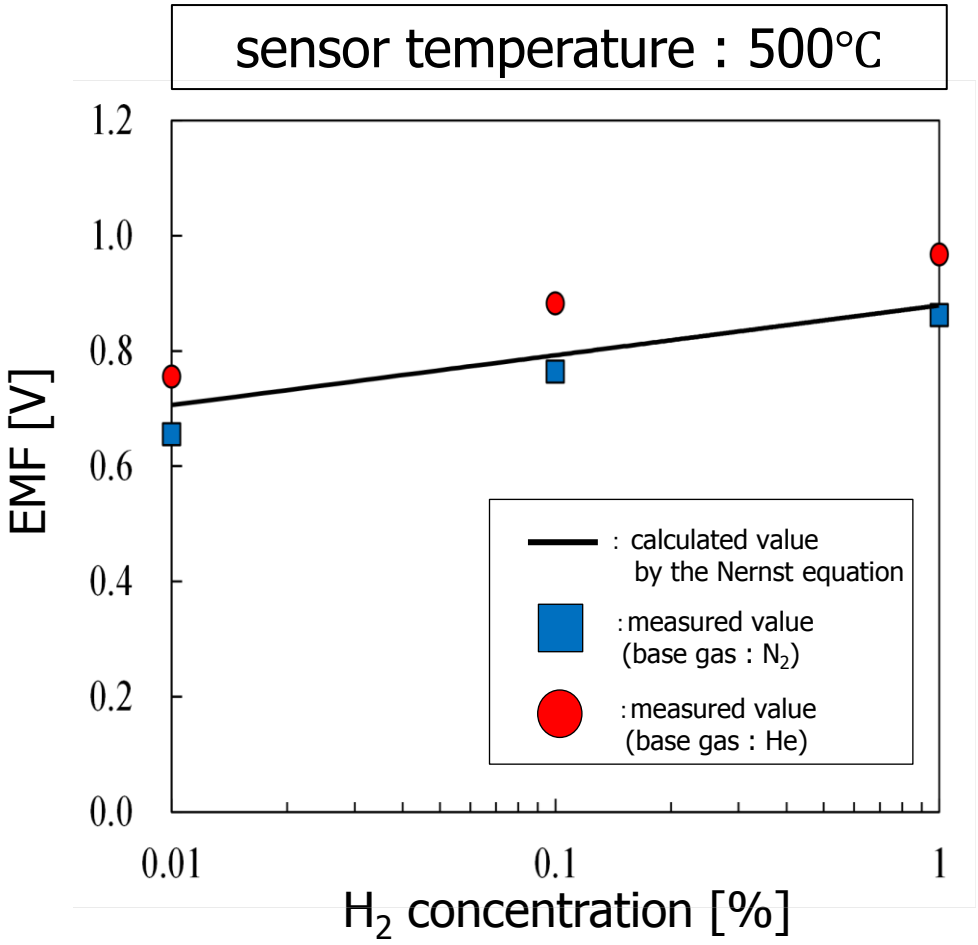
Results of EMF under He+H_2 environment

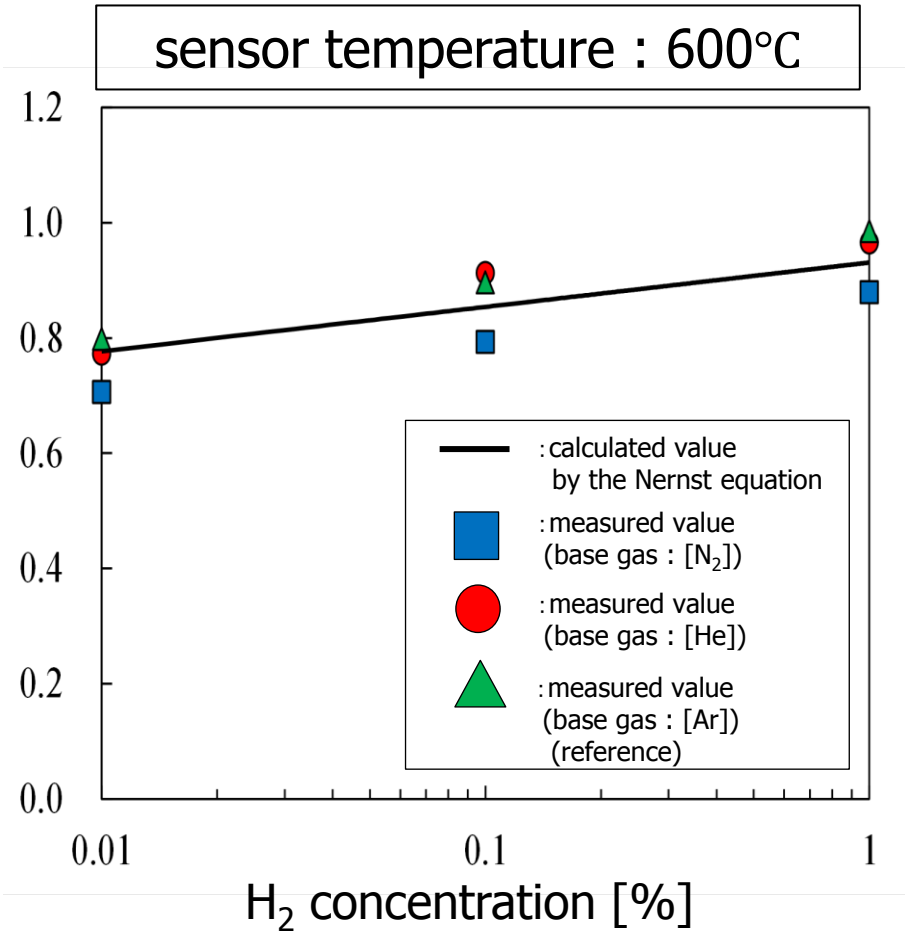




EMF of the hydrogen sensor was increased with H_2 concentration change at each temperature

Relationship between H_2 concentration and EMF





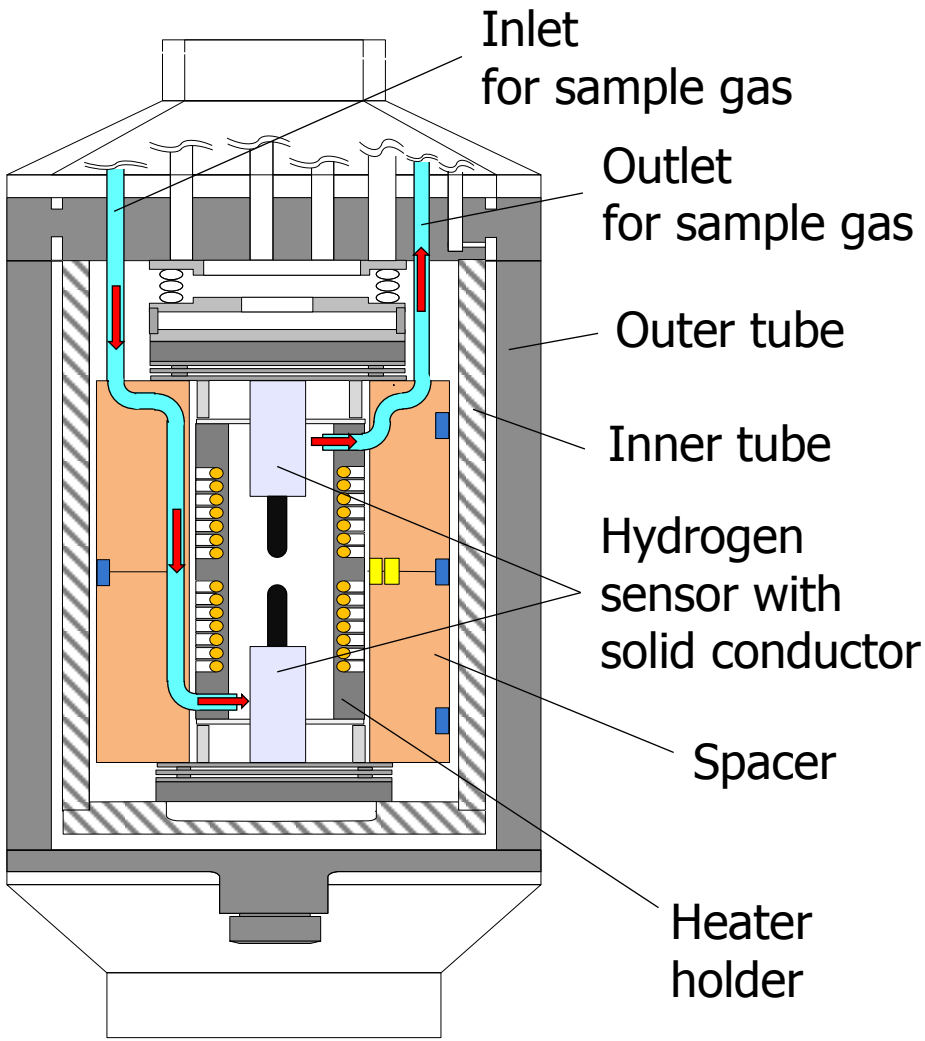
It was found that the hydrogen sensor was able to measure H_2 concentration with high accuracy in each base gas conditions.

Schematic diagram of the irradiation capsule in JMTR

Irradiation condition

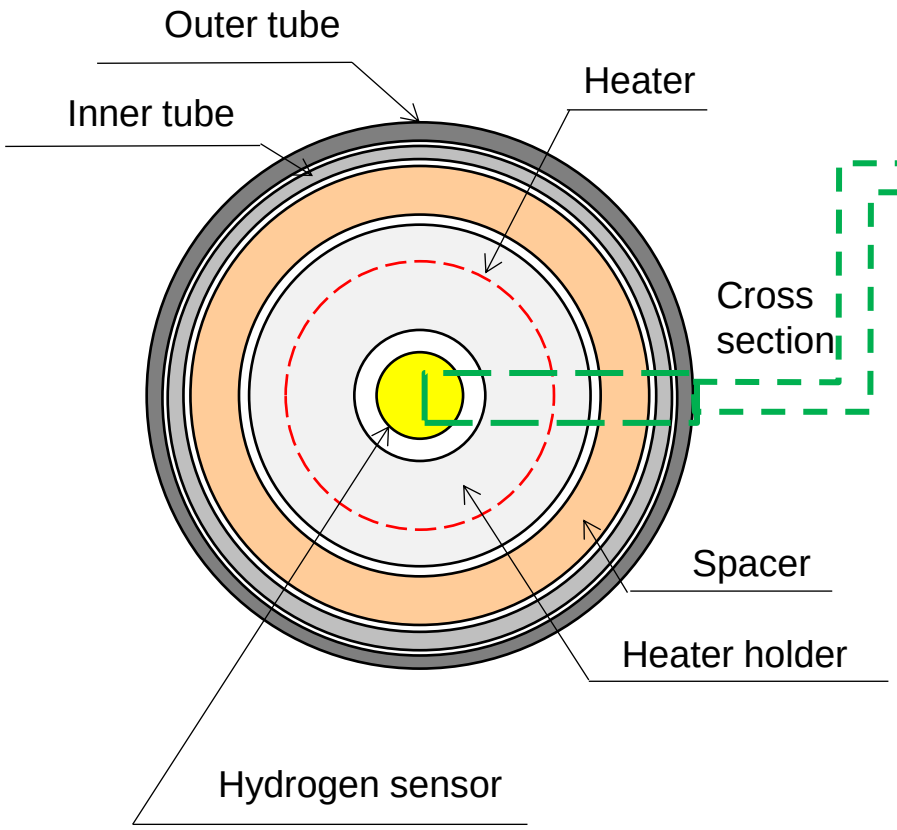
Item	Specification
Temperature in irradiation test	400～600 ℃
Sample gas flow rate	50～200 cc/min
H_2 concentration in sample gas	0.01～1 %
fast neutron flux	1.91×10^{16} $m^{-2} \cdot s^{-1}$
Irradiation cycle	10 cycles

Structure of irradiation capsule

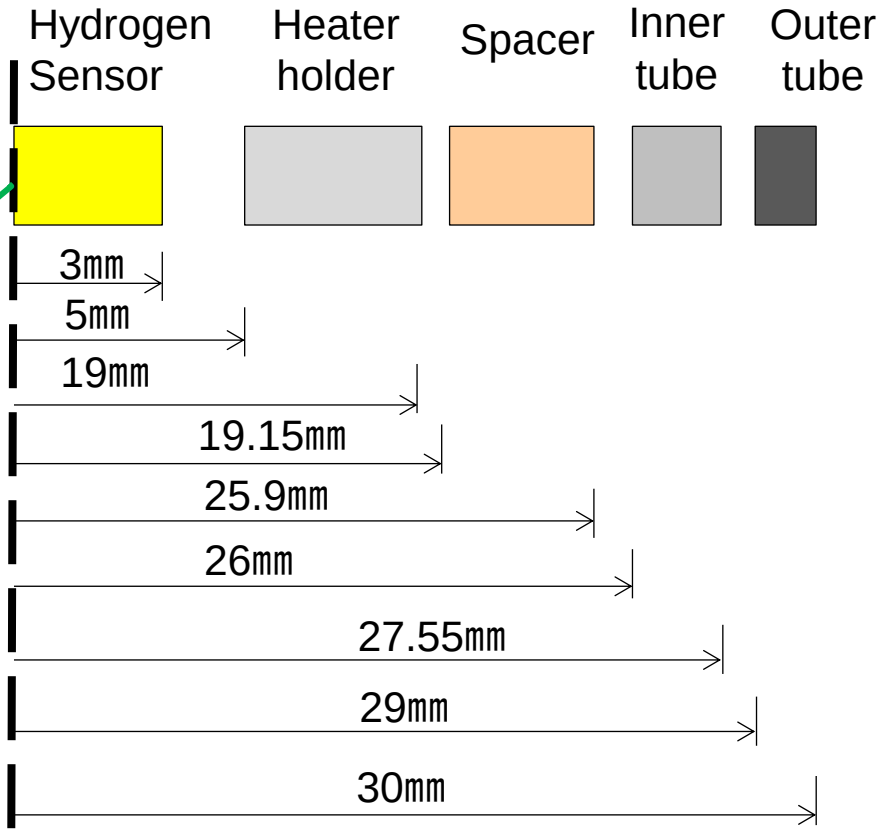


Temperature evaluation of hydrogen sensor in irradiation capsule

Model for temperature calculation



Distance from center of capsule



Inner temperature of the irradiation capsule was estimated by GENTC code to confirm the fabricated capsule is possible to heat the hydrogen sensor up to 400~600℃

Calculation conditions

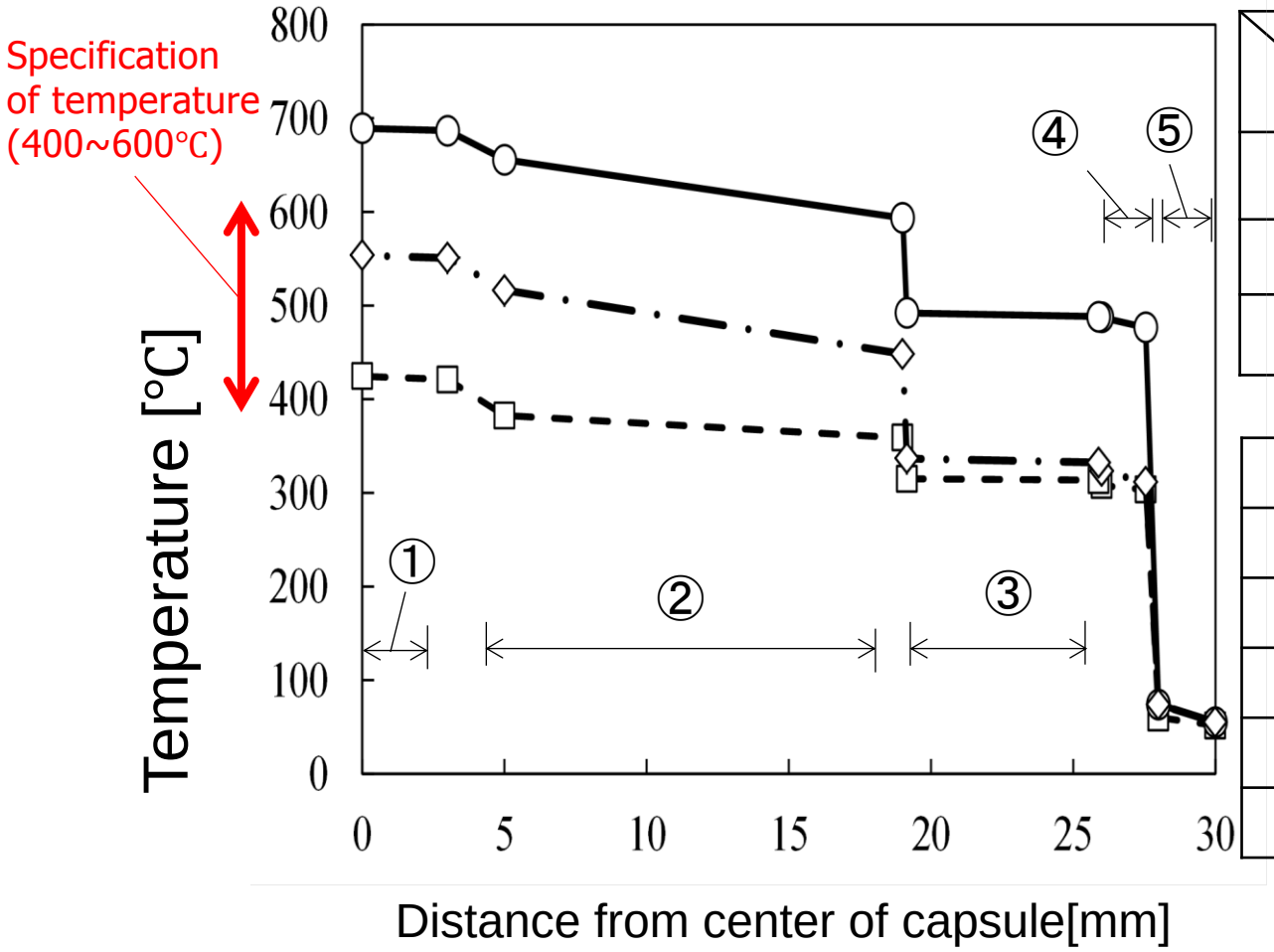
Environments and heater output

Item	value
Gamma heating rate	0.82(W/g)
Water Temperature	49 (°C)
Heat transfer coefficient of outer capsule surface	2.33 (W/cm^2/°C)
Environment inside capsule	He dump, 2/5He
Output of capsule heater	0, 150, 200 (W/cm)

Materials of parts

Part	material
Hydrogen sensor	$CaZr_{0.9}In_{0.1}O_{3-\alpha}$
・Heater holder ・Inner tube ・Outer tube	316SS
Spacer	Al

Temperature distribution in Irradiation capsule



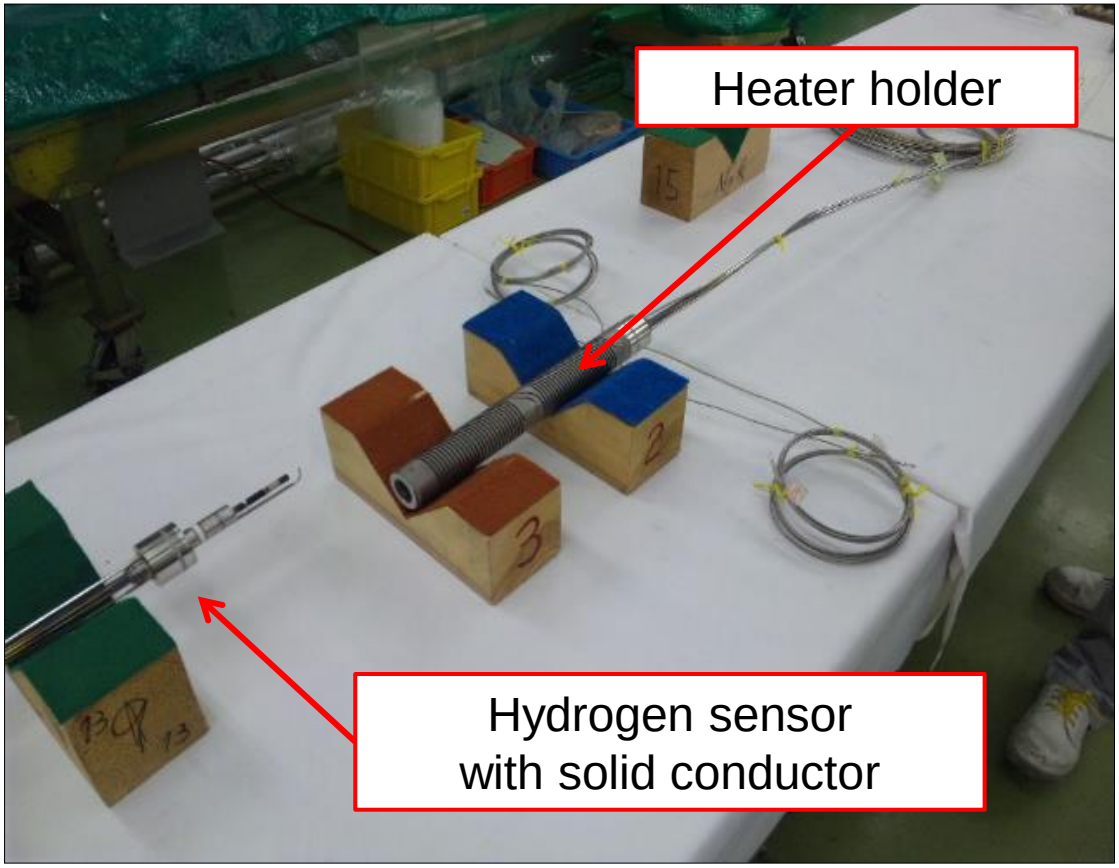
	Heater (W/cm)	environment
□	0	2/5 He
●	0	He
◇	150	2/5 He

No	Parts
①	Hydrogen sensor
②	Heater holder
③	Spacer
④	Inner tube
⑤	Outer tube

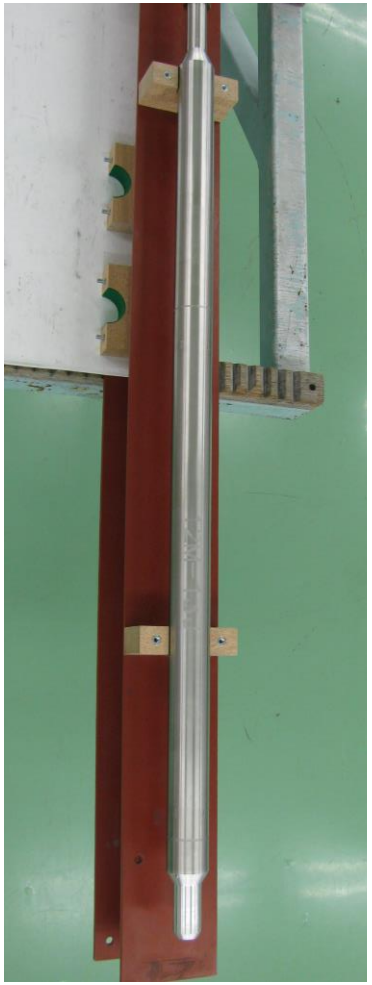
The fabricated irradiation capsule is able to control temperature of hydrogen sensor from 400 to 600℃.

Preparation for irradiation test after JMTR re-operation

Lording of Hydrogen sensor to irradiation capsule



Appearance of Irradiation capsule



CONCLUSION

1. Out-pile test of Hydrogen Sensor with Solid conductor

- The hydrogen sensor was able to measure H_2 concentration with high accuracy in N_2 and He base gas conditions.

2. Fabrication of irradiation capsule for the hydrogen sensor

- The capsule for irradiation test in JMTR was confirmed by GENTC code to be able to control temperature of the hydrogen sensor from 400℃ to 600℃.

In future, we plan to research the characteristic of the hydrogen sensor in irradiation test after JMTR re-operation.