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Sodium Hideout Study in an Open Tubesheet Crevice

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Abstract

In a locally restricted steam generator (SG) geometry, impurities in the bulk water can be concentrated by boiling process to extreme pH that may then accelerate the corrosion of tubing and adjacent materials. To mitigate the corrosion, the Molar Ratio Control technique is widely implemented with the EPRI initiative. In order to maximize its beneficial effect, the understanding of crevice processes needs to be advanced. To simulate a real SG tubesheet crevice high temperature/high pressure (HT/HP) crevice simulation system has been developed. Primary water with high flow rate flowed through the 3/4" tube and crevice section was made at the outside of the tube. The simulation apparatus was equipped with thermocouples and electrodes for the measurement of oxidation reduction potential (ORP) in crevice and bulk water. In an unpacked tubesheet crevice with 0.15 mm gap and 40 mm depth, axial temperature distribution and Pt potential vs. Ag/AgCl (Water) were measured with time. Also, to find crevice concentration from conductivity data AC impedance technique was used. Secondary pressure was maintained at 805 ± 5 psi. Secondary solution composed of 50 ppm Na and 1.85 ppm H_2 was supplied with the flow rate of about 4 L/hr. NaOH concentration process in crevice was observed from temperature and potential data. Conductivity in crevice was measured intermittently but insufficient to show the concentration process. Experimental data showed a similar behavior compared with calculated results by MULTEQ[®]. From the boiling point elevation data crevice concentration factor was estimated as about 400 for $\Delta T = 14$ °C and about 2000 for $\Delta T = 29$ °C.

1. Introduction

In a Pressurized Water Reactor (PWR), SG tubes were made of alloy 600 or 690 and steam is produced on the outer diameter side of the tubes. The SG reliability has been one of the important issues encountered during the PWR operation. Various corrosion phenomena were observed in the past [1]. In a locally restricted SG geometry, trace impurities in the bulk water can be concentrated by boiling processes to extreme pH. SG tube degradation phenomena result from the concentration of impurities mainly in three locations in the SG: the tube support plate crevice, the tubesheet crevice, and the sludge pile. These locations with concentrated solutions may then develop Outer Diameter Stress Corrosion Cracking/Inter-Granular Attack (ODSCC/IGA), which is the one of the principal degradation mechanisms in recent years [1]. To mitigate the ODSCC/IGA in the restricted geometry and to maintain SG tube reliability, the detailed understanding of boiling crevice phenomena is necessary.

A near-neutral crevice is the environment likely to produce the lowest IGA/SCC crack growth rate [2]. Based on this fact, the Electric Power Research Institute (EPRI) has developed the Molar Ratio Control (MRC) program, of which the goal is to maintain the crevice pH nominally in the range between 5 and 9 at the operating condition [3]. But, within plant operation, no one knows the crevice conditions. Therefore real-time crevice monitoring methods need to be developed.

Baum [4] reviewed, in detail, the thermal-hydraulic and chemical phenomena in restricted regions and the free span area. Early studies about crevice experiment were primarily focused on the characterization of thermal-hydraulic nature. Chemical concentration and chemical or electrochemical measurement in a crevice were studied more recently. Kozawa and Aoki [5] investigated experimentally several characteristics of boiling in a crevice between tube and tubesheet. A flat crevice and a fluid heating method were adopted. Experimental results showed that three kinds of boiling configurations could occur in a crevice.

Baum and Curlee [6] performed tests, which simulated the geometric, thermal, and hydraulic environment found between the tube and the TSP in typical PWR U-tube SGs. It was found that certain tube support configurations could produce a local liquid deficient heat transfer regime, which, in turn, could permit significant chemical concentration. The interrelationship between the heat and the mass transfer processes in the confined geometry was further demonstrated by comparing the results of an analytic model. Sodium hideout studies in SG crevices were carried out systematically by Campan and Shoemaker [7]. A method using Na^{24} as a tracer was developed.

A technique was developed to study electrochemical phenomena in crevices that simulate the geometry in nuclear SGs by Hermer et al. [8]. Electrochemical potentials were measured in TSP crevice geometry. Lumsden et al. [9] constructed a system, which operates with simulated SG crevice thermal conditions. The results obtained for the average boiling point elevation in the crevice, the analysis of the extracted crevice solutions, and the redox potential in the crevice and free span, after equilibrium was attained, agreed well with MULTEQ[®] predictions. The electrochemical noise monitoring technique was evaluated

in a refreshed autoclave system for eventual corrosion monitoring system in SGs [10].

An on-site model boiler facility was constructed in the Kansai Electric Power Company OHI Unit 1 at beginning of 1986 [11]. Corrosion potential monitoring in the bulk secondary water and pH monitoring in simulated SG crevices were carried out using the model boiler [11]. By analyzing directly sampled concentrated solution from heated crevice of an on-site autoclave, SG crevice environment was evaluated [12,13].

Based on earlier work, the objectives of this study are to directly observe the crevice phenomena at HT/HP and to measure chemical, thermohydraulic, and electrochemical parameters in boiling crevice. Single-ended crevice was designed to represent a tubesheet crevice.

2. Experimental

In the earlier works of the authors, electric heater was used to simulate primary heating condition. But the heating method by electric heater caused such problems as the non-uniformity of surface heat flux and difficulty in simulating real SG temperature distribution. Therefore, to simulate a real SG tubesheet crevice HT/HP crevice simulation system has been developed. Fig. 1 shows the schematic diagram of the simulation system. The system is composed of two main systems: primary water system and secondary water system. Primary water with high flow rate of about 2300 L/hr was circulated by HT/HP circulation pump through the 3/4" outer diameter (OD) 316 stainless steel (SS) tube. 1-gallon autoclave was used as the primary heater of which maximum power was 4.8 kW. High purity water purged with 5% hydrogen gas (Nitrogen bal.) was charged by high pressure pump and ejected by back pressure regulator. Primary pressure was maintained at 1985 ± 15 psig.

In Fig. 2 SG simulation vessel is schematically described. The vessel was made of 316 SS. To protect vessel failure by caustic stress corrosion cracking, electroless Ni plating with 10 μm thickness was applied on the whole vessel surface. Secondary water composed of 50 ppm Na and 1.85 ppm H_2 was charged by high pressure pump and ejected by back pressure regulator. Also, secondary water storage tank was made of Ti and purged with 5% hydrogen gas (Nitrogen bal.). The flow rate of secondary system was controlled by high pressure pump between 3 and 4 L/hr. Secondary pressure was adjusted automatically by PID controlled pressure regulator and maintained at 790 ± 5 psig of which equivalent saturation temperature is 271 °C. As described in Fig. 2 to make bottom-closed crevice the electrolytic Ni plating was applied on the crevice section of the 3/4" OD SS tube, and the plated section was machined as the required dimension.

The SG tubesheet crevice simulation vessel was equipped with thermocouples and electrodes for the measurement of ORP in crevice and bulk water. Fig. 2 shows the location of sensors installed in crevice and bulk region. Two thermocouples were installed at bulk region and five thermocouples were installed at crevice region. And primary inlet and outlet temperatures were monitored. Ag/AgCl (Water) electrode was used as reference electrode and two Pt electrodes were installed. In both crevice and bulk region,

reference and Pt electrode had the same axial location. In an unpacked tubesheet crevice with 0.15 mm gap and 40 mm depth, axial temperature distribution and Pt potential vs. Ag/AgCl (Water) were measured with time. Also, to find crevice concentration from conductivity data AC impedance technique was used. The voltage was measured by HP 34401A multimeter through the HP 3495A scanner. And AC impedance was measured by Solartron 1260 impedance/gain-phase analyzer.

3. Results & Discussion

Fig. 3 shows the temperature and pressure variation with time at the condition of $T_p=285$ °C. At the elapsed time of 114 hours secondary water flow rate was increased from 3 L/hr to 4 L/hr. As the flow rate was increased the crevice and bulk temperatures were suddenly decreased and gradually increased by Na concentration process. After about 5 hours crevice temperature became constant. 'T/C #5' located in the lowest level indicated that the boiling point elevation at that point compared with saturation temperature was about 2 °C. Fig. 4 shows the crevice and bulk potential variation with time at the same condition as Fig. 3. Bulk potential remained -0.9 V vs. SHE(T) but crevice potential showed fluctuation from -1.0 V to -0.7 V. 'T/C #3' located in the same level as Pt and reference electrode indicated that the solution temperature at that point was under the saturation temperature and Na concentration didn't occur. If boiling didn't occur in crevice the crevice potential had to be the same as the bulk potential. But the reason why the crevice potential showed slightly higher value than the bulk potential is not clear. Fig. 5 shows the temperature and pressure variation with time at the condition of $T_p=299-300$ °C. Primary pressure data showed the stable behavior even at the temperature transient condition. Secondary pressure data also showed very stable behavior. Crevice temperatures were dependent on primary temperature variation at the period of 135-145 hours. After the primary temperature became constant all of the crevice temperatures increased gradually beyond the saturation temperature. Crevice temperature ramp rate got higher as the position of measuring temperature got deeper into the crevice. 'T/C #5' located in the lowest level indicated that the boiling point elevation at the condition of $T_p=299$ °C compared with saturation temperature was about 15 °C. At the elapsed time of 160 hours primary temperature increased to 300 °C. 'T/C #5' and 'T/C #4' showed no significant change but 'T/C #3', 'T/C #2', and 'T/C #1' indicated additional boiling point elevation. Fig. 6 and Fig. 7 shows the crevice and bulk potential variation with time at the same condition as described in Fig. 5. In Fig. 6 crevice potential data showed the Na concentration in crevice at the period of 140-165 hours. In case of 'Pt #1', the potential vs. SHE(T) was varied from -0.9 V to -0.8 V at first and decreased to -1.1 V. Although some difference was in the time scale 'Pt #2' showed the similar behavior. The potential signal of 'Pt #1' showed the dependency on the crevice temperature variation at the period of 158-165 hours. In Fig. 7 bulk potential became very unstable as the primary temperature increased to 299 °C. This is probably because the bulk temperature came close to the saturation temperature and started boiling. To suppress the bulk water boiling higher flow rate is needed. Fig. 8 shows the axial temperature distribution according to the primary water

temperature variation.

By using MULTEQ-REDOX® Ver.2.22 high temperature solution pH, boiling point elevation, and ORP were calculated [14]. System temperature was set to be 270 °C and flowing option was used. Fig. 9 shows the MULTEQ® calculated results about pH and boiling point elevation as a function of crevice concentration factor. And Fig. 10 shows the MULTEQ® calculated results about pH and ORP as a function of crevice concentration factor. As described in Fig. 9 the boiling point elevation becomes significant only just when the concentration factor exceeds 100. As the concentration factor is 10^4 the boiling point elevation becomes more than 50 °C. In Fig. 10 ORP is about -0.9 V vs. SHE(T) when concentration does not occur. But as a little concentration occurs ORP goes up to -0.75 V and then decreased gradually. Hydrogen has very high distribution coefficient and by boiling process hydrogen escapes from the crevice solution. This is the reason of the initial jump of ORP. In this experiment the bulk potential was maintained at -1.0 V vs. Ag/AgCl (Water) and from Fig. 10 the bulk potential calculated by MULTEQ is -0.9 V vs. SHE(T). From these results it was deduced that Ag/AgCl (Water) vs. SHE(T) at 270 °C was +0.1 V. By using this conversion value the potential measured against Ag/AgCl (Water) was converted to the potential against SHE(T). In Fig. 11 experimental results were compared with MULTEQ® calculated results. Experimental data showed a similar behavior compared with calculated results by MULTEQ® although the absolute values showed some discrepancy. From the boiling point elevation data concentration factor in an open tubesheet crevice was estimated as about 400 for $\Delta T=14$ °C and about 2000 for $\Delta T=29$ °C. The real components of impedance data measured at the elapsed time of 116 hours are plotted in Fig. 12. Impedance in crevice and bulk was measured intermittently but insufficient to show the concentration process. It is needed to show the reasonable relationship between temperatures, potential, and conductivity data measured in crevice and bulk.

4. Summary & Conclusions

From the experiment and analysis for the HT/HP open tubesheet crevice following conclusions are made.

- NaOH concentration process in an open tubesheet crevice was confirmed from temperature and potential measurement data.
- From the boiling point elevation data, 0.15 mm open tubesheet crevice concentration factor was estimated as about 400 for $\Delta T=14$ °C and about 2000 for $\Delta T=29$ °C.
- Conductivity in crevice was measured intermittently by AC impedance technique but insufficient to show the concentration process.
- By using MULTEQ-REDOX® Ver.2.22 high temperature solution pH, boiling point elevation, and ORP were calculated. Experimental data showed a similar behavior compared with calculated results by MULTEQ®.
- It is needed to show the reasonable relationship between temperatures, potential, and

conductivity data measured in crevice and bulk.

Acknowledgement

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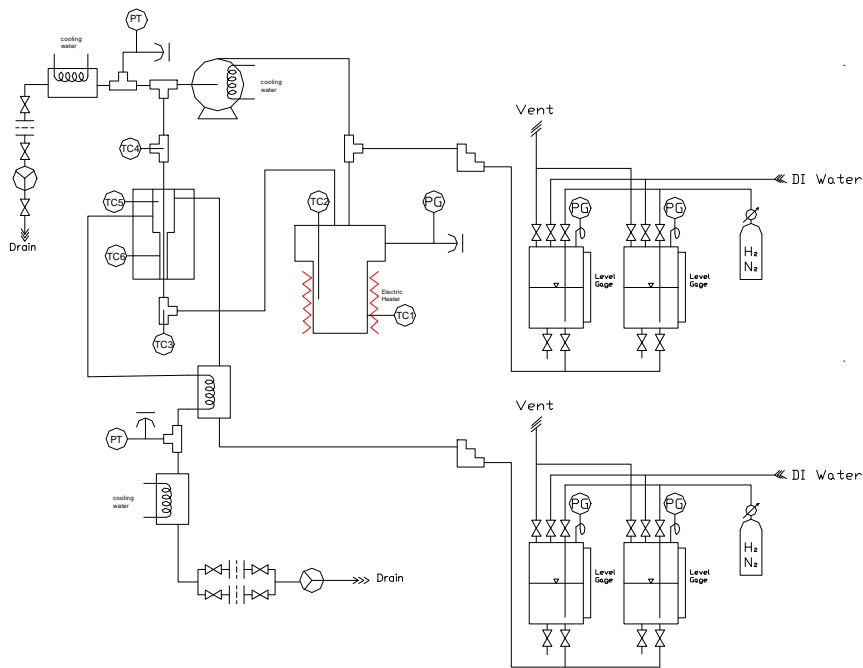


Figure 1. Schematic diagram of SG crevice simulation system.

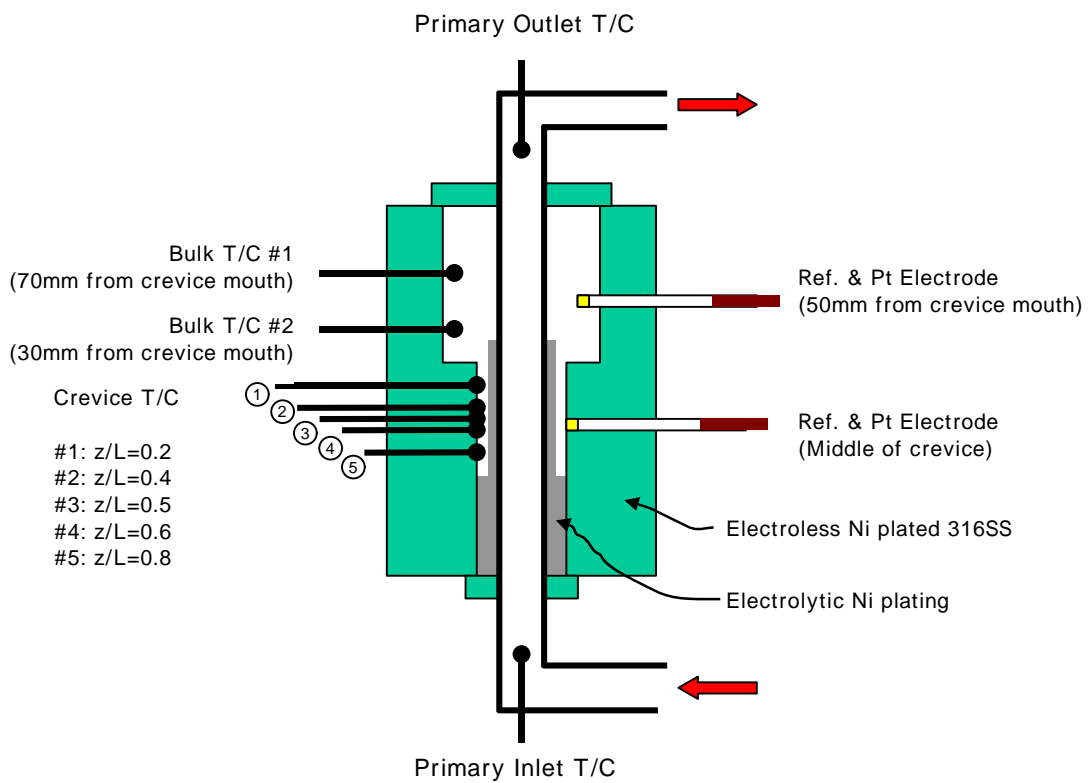


Figure 2. Schematic diagram describing location of sensors installed in crevice and bulk region.

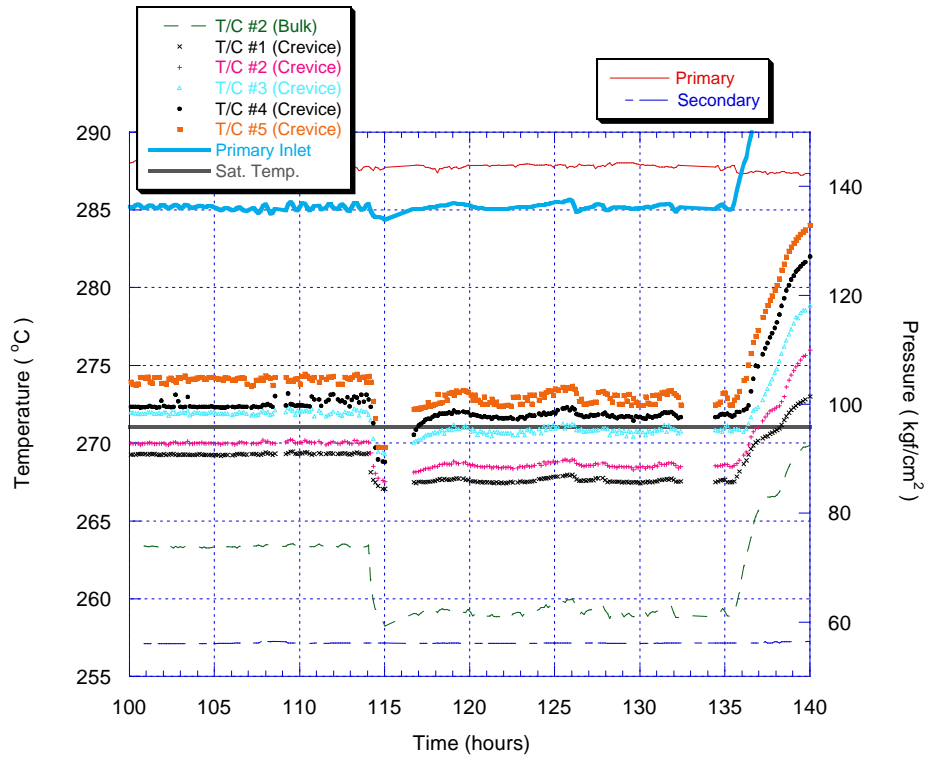


Figure 3. Temperature and pressure variation with time at the condition of $T_p=285$ °C.

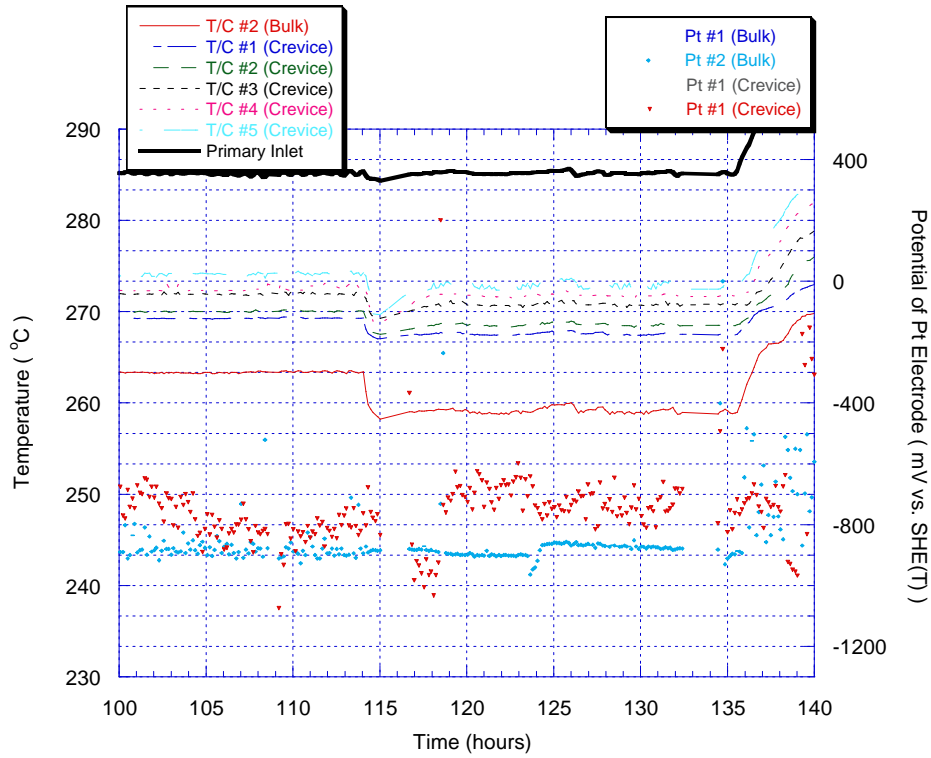


Figure 4. Crevice and bulk potential variation with time at the condition of $T_p=285$ °C.

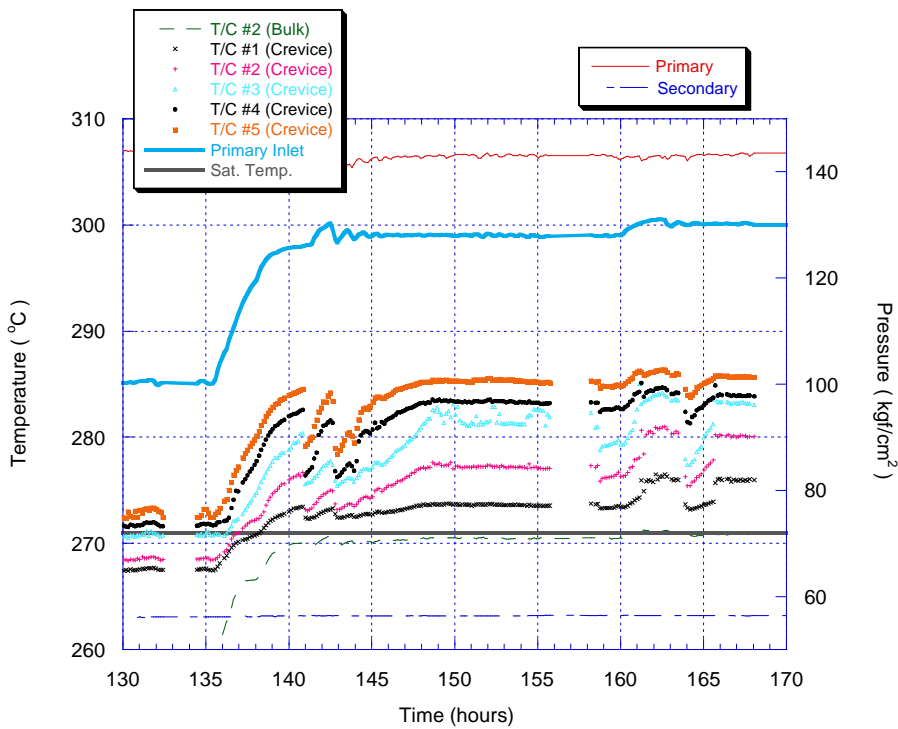


Figure 5. Temperature and pressure variation with time at the condition of $T_p=299-300$ °C.

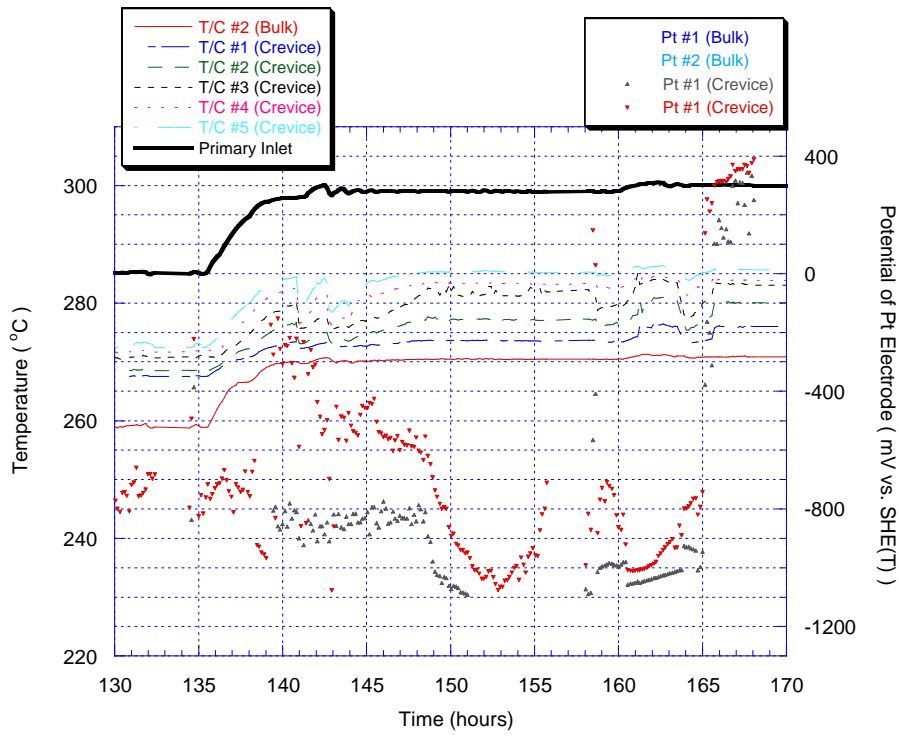


Figure 6. Crevice potential variation with time at the condition of $T_p=299-300$ °C.

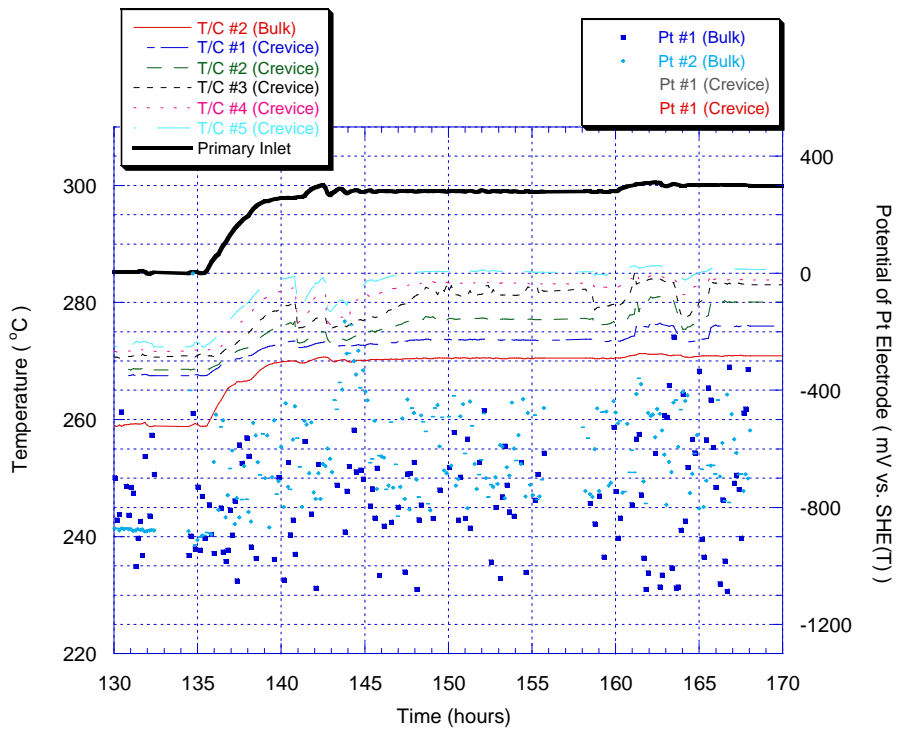


Figure 7. Bulk potential variation with time at the condition of $T_p=299-300$ °C.

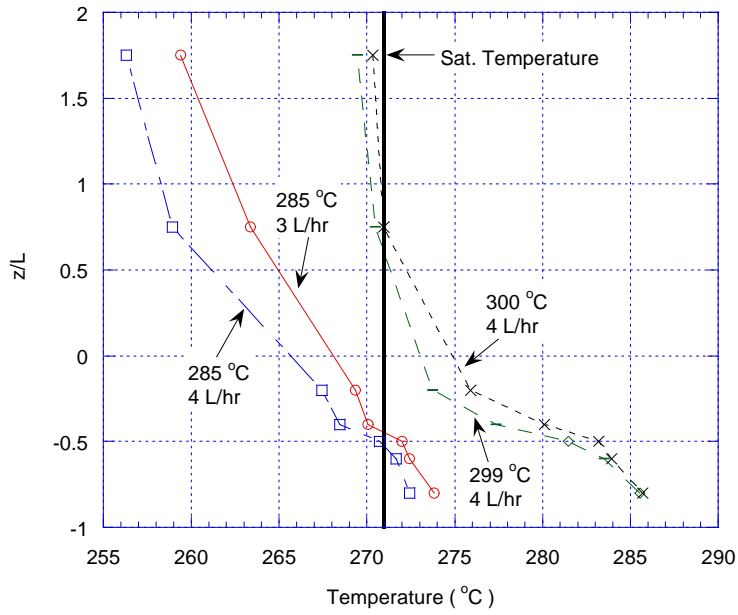


Figure 8. Axial temperature distribution according to the primary temperature variation.

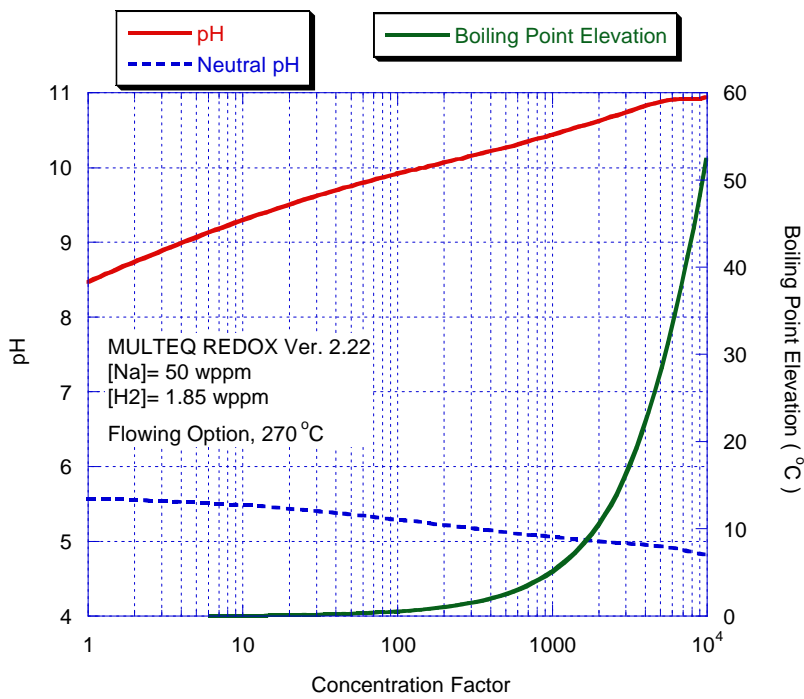


Figure 9. MULTEQ® calculated results about pH and boiling point elevation as a function of crevice concentration factor.

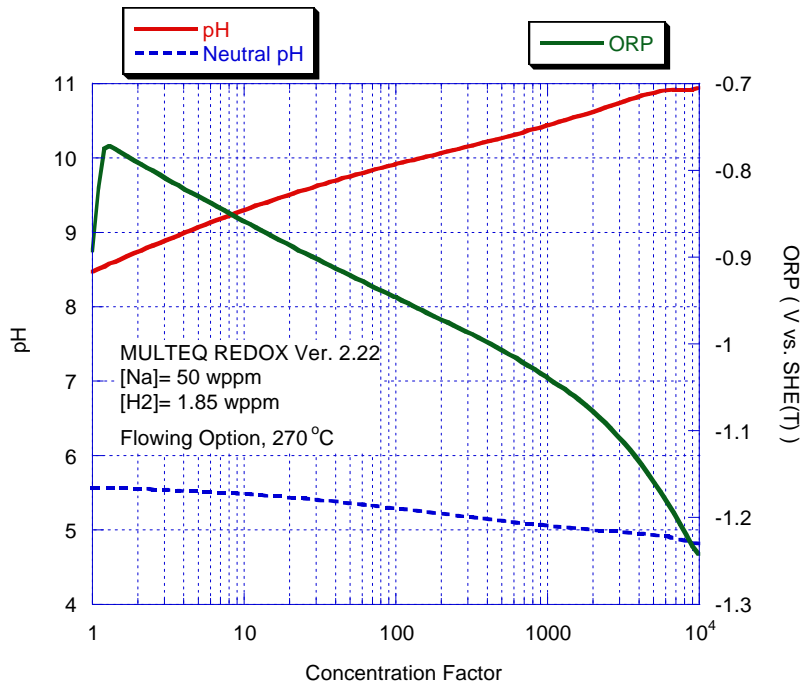


Figure 10. MULTEQ[®] calculated results about pH and ORP as a function of crevice concentration factor.

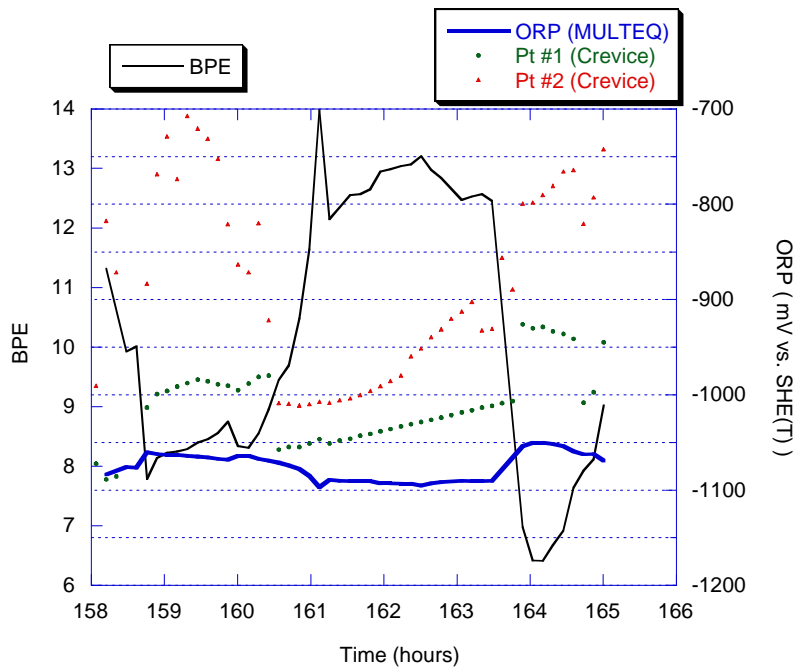


Figure 11. Comparison experimental results with MULTEQ[®] calculated results.

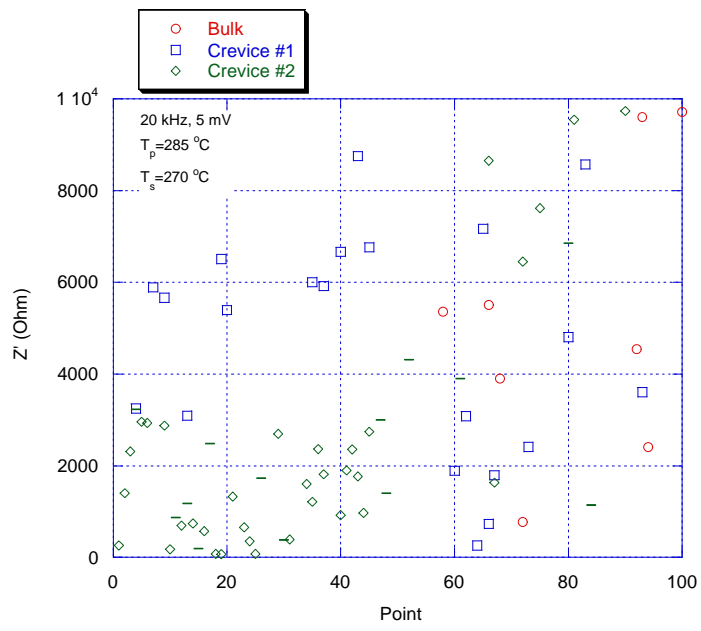


Figure 12. Real components of impedance data measured at the elapsed time of 116 hours.