### COMPARISON OF THE SPECIAL PROPERTIES OF SUPERCRITICAL WATER BY THERMAL AND COLD DYNAMIC NEUTRON RADIOGRAPHY

# M. Balaskó<sup>1</sup>, L. Horváth<sup>1</sup>, Á. Horváth<sup>1</sup>, P. Tóth<sup>1</sup> B. Schillinger<sup>2</sup> and M. Mühlbauer<sup>2</sup>

<sup>1</sup> MTA KFKI Atomic Energy Research Institute, Budapest, Hungary <sup>2</sup> Technische Universität München, Garching, Germany

#### Abstract

The supercritical-water-cooled reactor will work at high temperature and high pressure above the thermodynamic critical point of water. It is very important task to gain an understanding of the fluid mechanic behavior of the supercritical water. Dynamic neutron radiography is able to provide visual information about the motion of hydrogen containing materials (e.g. heavy and light water). A special sample holder family and a special measuring frame were developed at the Budapest research reactor to study the properties of water at high temperature and at high pressure by thermal neutrons. These investigations were extended using cold neutrons of FRM II. Some subtleties of water's thermo-hydraulic behavior near the critical point were observed.

### 1. Introduction

The supercritical-water-cooled reactor will work at high temperature and high pressure above the thermodynamic critical point of the water. However, little is known about the thermo-hydraulic behavior of water under extreme conditions, although water at temperatures beyond the critical point ( $T_c \sim 374$  °C,  $p_c \sim 221$  bar,  $\rho_c \sim 0.32$  g/cm<sup>3</sup>) is known to exhibit many peculiar properties. For example its density can be controlled between gas like and liquid like values by varying its pressure and temperature. Dynamic neutron radiography (DNR) gives a possibility to observe the change in density of the water under extreme conditions. The basic phenomena of the changing density of the water depending on the temperature and the pressure were investigated by DNR at the Dynamic Radiography Station (DRS) of the Budapest research reactor (10 MW) [1, 2]. Neutron radiation passing through the object is converted into visible light by a scintillator screen. The light is detected by a low light level (LLL) television camera. Radiography images are displayed on a monitor, stored by S-VHS and DVD recorders for further analysis. The aim of the DNR investigation was to give visual information about the behavior of the two phases of the water in the sample holder. The phase changing of the water was (near to supercritical state) visible by DNR. The real time pictures contained many disturbing artifacts by the intensive background of scattered thermal neutrons. As a result, the fine details of the events may be hidden.

We had the opportunity to extend our results at a high level neutron radiography station at the ANTARES at FRM II (20 MW) research reactor [3]. This neutron beam line is directed towards a liquid deuterium cold source inside the moderator tank and the majority of its neutrons have very low energy. We were able to see many details of the streaming inside the titanium alloy sample

holder because the interaction between the neutron radiation and sample environment did not produce as many scattered neutrons as were detected at DRS.

### 2. Experimental procedure

### 2.1 Experimental facilities

The investigation was performed at the DRS of the Budapest research reactor (10 MW) and the ANTARES facility at the FRM II (20 MW) research reactor. The main characteristics of the DRS neutron beam are the following: the neutron flux is  $8X10^7$  n cm<sup>-2</sup> sec<sup>-1</sup>, the gamma dose rate is 8.5 Sv/h, the Cd ratio is 8 and the L/D ratio is 170. The same parameters of the ANTARES are the following: the neutron flux is 9.4X10<sup>7</sup> n cm<sup>-2</sup> sec<sup>-1</sup>, the gamma dose rate is 0.69 Sv/h, the Cd ratio is 15 and the L/D ratio is 400. Neutron radiation passing through the object is converted into light by a scintillator screen, and the light is detected by a LLL tv camera. Radiography images are displayed on a monitor and stored by S-VHS and DVD recorders for further analysis. At the DRS the sensor of the LLL tv camera is a vidicon tube with built in light-intensifier. The type of the camera is TV1122. The single frame exposure time of the camera is 40 msec and its sensitivity is  $10^{-4}$  lux. At this registration speed and sensitivity are available to study the medium speed motion of the water in the sample holder. At the ANTARES facility the sensor of the LLL tv camera is a CCD chip, with resolution of 2048X2048 pixel and 16 bit dynamics. The type of the camera is ANDOR. The camera is able to change its picture time between 100 msec and 6 sec, thus changing the resolution of the pictures. Naturally, these exposure times are suitable to detect very slow motion of the water in the sample holder only.

## 2.2 Investigated object

The study of water in the supercritical regime places heavy requirements on the material of the sample holder. Materials with good corrosion resistance and of high strength at high temperature (above 400  $^{0}$ C) were selected, as it published in [2, 4]. The safe measurement conditions and the integrity of the sample holders were verified by a stress analysis calculation based on the equivalent tensile stress, or Huber-Mises-Hencky yield criterion. Details can be found in [5]. The sample holder was made of GR5 type titanium alloy. The application of the titanium alloy gave two advantages. The first was that the higher strength allowed for thinner wall thickness which resulted in better quality of the radiography pictures. The second was that the total macroscopic cross section of titanium is only half of carbon steel or stainless steel. This also means a better quality neutron radiography picture. The sample holder applied a flange style closure fitting with 8 x M8 screws (GR-5H). It has 7 mm wall thickness and it contains a hole with 20 mm diameter. The GR-5H sample holder was measured by three "K" type thermocouples

in the middle and at the ends (3-4  $^{0}$ C variation). Likewise the parameters outside the sample holder were measured by three "J" type thermocouples positioned in the middle and at the ends again (3-4  $^{0}$ C variation). One version of the closure fitting has a special inner heating coil for simulating the core of SCWR. The sample holder was placed in an alumina cylindrical temperature reflector with a diameter of 90 mm, its length is 150 mm and its wall thickness of 0.3 mm. The temperature-increasing step took ~ 60 minutes from room temperature to 400 °C. A neutron radiography picture of GR-5H filled with 13 cm<sup>3</sup> of heavy water ( $T_c \sim 371$  °C,  $p_c \sim 220$  bar,  $\rho_c \sim 0.36$  g/cm<sup>3</sup>) at room temperature is shown in the Fig. 1. This picture was recorded by the ANDOR CCD camera (exp. time: 100 msec).



Fig. 1. Cold neutron radiography picture of GR-5H with 13 cm<sup>3</sup> heavy water at room temperature by ANDOR CCD camera

## 2.3 Experimental technology

The aim of the DNR investigation was to give visual information about the behavior of the two phases of the water in the sample holder. The whole test was recorded on S-VHS video and on DVD. Simultaneously, the temperature was monitored and recorded at 8 positions in the arrangement, together with the pressure.

To confirm repeatability of the events, a special measuring frame was designed and built by us. This arrangement is able to reproduce the starting conditions of the whole measurement procedure. A block diagram of the apparatus is shown in Fig. 2. It has a water cooled double Peltier block to freeze the water during the evacuation of the sample holder above the water level. After the evacuation, the hermetic valve is closed and water is injected into the high pressure area. This area contains a pressure transmitter for measuring the pressure while the temperature is increased above 200 °C. Above this temperature the hermetical valve is opened. It is situated in the high pressure area. This remote control ventilating valve is able to stabilize the pressure independent from temperature. In addition, this element can create a situation for simulating a loss-of-coolant type accident (LOCA). Naturally the Peltier block is removed before the heating procedure is started.



Fig. 2. Functional diagram of the measurement

### 3. Results

Thermalhydraulicists are interested in the starting point of the measurement procedure and the details of LOCA simulation. Both of these events contain large scale motions of the water, and high pressure vapour streaming. Many interesting details were observed during the investigation. The first period of the measurement procedure (between 45 °C and 110 °C) gave the most spectacular events, such as bubble generation, plug motion as a piston, and whirlpool motion. Many new subtle differences were observed by the high quality cold neutron beam and the two kinds of LLL cameras on the ANTARES. In this paper, we present some results about the initial period of the measurement test. Figs. 3a. and 3b. were recorded by the ANDOR CCD camera while Figs. 4a and 4b. were recorded by the vidicon camera at the ANTARES using heavy water as working fluid. The exposure times in Fig. 3a. and 3b. were 100 msec (binning 6X6; 118X284 pixel; 16 bit; L/D = 400), and were 40 msec (193X494 pixel; 8 bit; L/D = 400) in Figs 4a and 4b. A large bubble was generated at 45 °C as shown in Fig. 3a. A large expanding water plug moved upward at 60 °C as is visible in Fig.3b. The motion of a large bubble at 50 °C led to a depleted area on the right side of the sample holder as shown in Fig. 4a. The position of a water plug (like a piston) is visible at 65 °C in Fig. 4b.



Fig. 3a. Large bubble at 45 °C with heavy water using ANDOR CCD camera (exp. time: 100 msec) at the ANTARES



Fig. 3b. Water plug at 60 °C with heavy water using ANDOR CCD camera (exp. time: 100 msec) at the ANTARES





Water plug

Fig. 4a. Large bubble at 50 °C with heavy water using vidicon camera (exp. time: 40 msec) at the ANTARES.

Fig. 4b. Water plug at 65 °C with heavy water using vidicon camera (exp. time: 40 msec) at the ANTARES.

The exposure times in Figs. 5a. and 5b. were 40 msec using the vidicon camera at the DRS where the working fluid was light water. A large boiling effect is visible in Fig. 5a. at 45 °C. A water wave front coming down on the left side of the sample holder at 50 °C is observable in Fig.5b.(347X494 pixel, 8 bit, L/D = 170).



Fig. 5.a. Boiling of the light water at 45 °C on DRS (exp. time: 40 msec)



Wave front Fig. 5. b. Light water wave front is coming down at 50 °C on DRS (exp. time: 40 msec)

It seems the biggest difference between the character of the cold neutron (Figs 3.a., 3.b., 4.a. and 4.b.) and the thermal neutron (Figs. 5.a. and 5.b.) radiography pictures are that the cold neutron radiography pictures show more details of the thermalhydraulic characteristics of heavy water but hardly see the contour of the sample holder which is made from GR5 type titanium alloy, while the thermal neutron radiography gives more information about the metal sample holder but it can not show the details of the heavy water streaming, and we had to substitute it by light water.

#### 4. Conclusion

Cold and thermal DNR is available to study the behavior of the water from room temperature to supercritical temperatures and at high pressure. A special sample holder GR-5H and measuring frame were developed for the success of the investigation. The first period of the measurement procedure (between 45 °C and 110 °C) gave many attractive events, like bubble generation, plug motion as a piston, thermalhydraulic features and whirlpool style motion, which were observed by DNR. Many new subtle of were observed using the cold neutron beam and two kinds of LLL cameras. Their superior resolution and shorter exposure times gave complementary results. The thermal DNR provided plenty of information about the thermalhydraulic behavior of light water in the sample holder.

#### 5. Acknowledgment

The authors wish to thank Dr. Gábor Házi for valuable discussions. This work was supported by the National Office for Research and Technology in the NUKENERG grant and by the European Commission under the 6<sup>th</sup> Framework Program through the Key Action: Strengthening the European Research Area, Research Infrastructures, Contract no: RII3-CT-2003-505925.

#### 6. References

- [1] M. Balaskó, E. Sváb, "Dynamic neutron radiography instrumentation and applications in Central Europe" *Nucl. Instr. and Meth.* A 377 (1996) pp 140-146.
- [2] M. Balaskó, L. Horváth, Á. Horváth, L. Kammel, P. Tóth, G. Endrőczi, "Investigation of the behaviour of supercritical water in the model of 4<sup>th</sup> generation nuclear reactor by combined nde methods"*EUR* 23356 EN- 2008, 441- 448,Proc. Sixth International Conference on NDE in Relation to Structural Integrity for Nuclear and Pressurised Components,8-10 October 2007, Budapest, Hungary, *EUR* 23356 [3] E. Calzada, B. Schillinger, F. Grünauer, "Cold neutron radiography at the FMR research reactor", *Nucl. Inst. and Meth.* A542 (2005) pp 38-44.
- [4] M.C. Bellisent-Funnel, "The structure of supercritical heavy water as studied by neutron diffraction", *J.Chem.Phys.*, 107(8) (1997) pp 2942-2949.
- [5] J.W.Murzewski, "Calculation and modeling of pressurized vessels" Int. Jour. of Damage Mech. (2006) 15, pp 69-87.