ELECTRON IRRADIATION OF THE MATERIAL SAMPLES OF NEW GENERATION NUCLEAR REACTORS IN THE SUPERCRITICAL WATER CONVECTION LOOP

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The design of the Supercritical Water Convection Loop with an irradiation chamber is described [1]. The plant makes possible to carry out simulation corrosion tests of potential structural materials for Generation IV reactors with the Supercritical Water-Cooling under irradiation. Specimens in water flow were irradiated in situ by the 10 MeV/10 kW electron beam of the LAE-10 linear accelerator. The high power relativistic electron-gamma irradiation delivers the absorbed doses sufficient for activation of corrosion and oxidation of material-coolant interfaces. The first results of the electron irradiation of Zr and Inconel 690 samples during 500 hours are gave.

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INTRODUCTION

The Supercritical Water-Cooled Reactor (SCWR) is one of the most promising nuclear technologies identified for R&D under the Generation IV (GenIV) program [2]. At Atomic Energy of Canada Limited (AECL) the SCWR has been recognized as the next evolutionary step of CANDU technology and given a high priority status [3 - 5]. Diverse developments for the next generation SCWRs are being currently carried out in Korea [6-8], the U.S. [9, 10], Japan [11], Russia [12], and China [13]. Along with the Very High-Temperature Reactor (VHTR) and Molten Salt Reactor (MSR) systems of Generation IV roadmap [1], SCWR technologies (either CANDU or Super-WWER-related) may also be considered as possible candidates for GenIV prospects of nuclear power industry of Ukraine.

Different construction materials are being considered as candidates for the SCWR: austenitic and ferritic/martensitic (F/M) stainless steels (SS), Ni-, Zr-, and Ti-based alloys, as well as innovative oxide dispersion strengthened (ODS) steels and alloys. Their corrosion rates and stress corrosion cracking (SCC) in pure SCW are being studied experimentally using SCW circulation loops (SCWCL) without irradiation. A comprehensive survey of current R&D can be found, e.g. [9]. Experiments at test SCWCL facilities [10, 14] provide us with valuable data on temperature and material composition dependencies of corrosion rate [9, 14] for various structural materials, as well as on the impact of SCW chemistry on corrosion kinetics of steels and alloys. However, this experience is insufficient for designing and construction of the SCWR since radiation effects are well known to affect corrosion behavior of materials. It should be emphasized that currently there are no experimental data on the corrosion and SCC of structural materials in the SCW flows under irradiation. The sparse data available [9] are limited to the experiments with pre-irradiated samples. These experiments have definitely shown that the temperature dependent rate of intergranular cracking of 304 and 316L stainless steels in SCW is increased considerably for samples subjected to prior irradiation by protons to the radiation damage dose of several dpa [9]. It strongly motivates further investigations of irradiation impact on SCW induced corrosion.

The corresponding test facilities are currently designed and commissioned, e.g., in the Irradiated Material Testing Laboratory (IMTL) at the University of Michigan (U.S.A.) [15] where SCC of materials irradiated in reactors by neutrons and gamma quanta will be studied in SCW low-oxygen closed-loop circuit designed for 30 MPa/600°C operation. It is worth noting that high radioactivity of neutron irradiated specimens has necessitated the application of automated systems of samples handling and the use of hot cells for sample preparation and post-irradiation SEM tests of microstructure. The radiation safety requirements make these experiments rather complicated and expensive. Nevertheless, since in situ irradiation is out of the present time capabilities of the IMTL facility, the investigations of combined effects of SCW impact and neutron irradiation (that are expected to occur in SCWR environment) still remain impossible within the framework of these important developments.

The irradiation induced impact of SCW radiolysis on flow control and instabilities, incl. transitions from subcritical to supercritical state, is of great interest for SCWR R&D and can also influence the corrosion of structural materials. These issues require thorough studies using dedicated experimental facilities [16] that offer combined exposure of samples to both SCW flow and irradiation.

In 2009 the Canadian government provided funding to support collaborative activities between the NSC Kharkov Institute of Physics and Technology (KIPT) and their colleagues from AECL's Chalk River Laboratories aimed at the development of advanced experimental facilities and methodologies for the assessment of reactor materials recognized as promising candidates for SCW reactors. The advanced skills of KIPT experts in structural materials design and testing, along with experience in simulation of reactor in-pile irradiation using gamma, electron, and ion irradiation, will be employed for SCWR candidate materials characterization.These activities are managed by the Science &

Technology Centre in Ukraine (STCU) within the framework of the Canada-Ukraine partner project STCU #4841 "SCW Convection Loop for Materials Assessment for the Next Generation Reactors". The Project goal is the design and construction of Canada-Ukraine Electron Irradiation Test Facility (CU-EITF) having a specially developed SCWCL with target test cell subjected to electron irradiation. After having been put into operation CU-EITF will be used for collaborative tests of structural materials for GenIII+ and GenIV reactors of the CANDU family (ACR, CANDU SCWCR).

1. THE SUPERCRITICAL WATER CONVECTION LOOP DESIGN

The dimensions of the loop (1.2×1.5 m) and other component parts of SCWCL are essentially determined by the size and arrangement of the KIPT-sited bunker room, which houses the electron accelerator (Fig. 1).

Fig. 1. The placement of the SCWCL in the LAE-10 bunker room

A schematic representation of the major components of the CU-EITF design is shown in Fig. 2. It is conceptually similar to various SCW natural and forced circulation loops that are currently operating and projected (see, e.g., [10, 14, 15]) but notably differs by the incorporation of the electron linac LAE-10 and the appropriate irradiation cell (IC).

All component parts of the SCWCL are designed for safe operation at temperatures up to 450°C (723 K) and pressures up to 25 MPa. The loop is made of stainless steel 12X18H10T and has an external pipe diameter 40 mm and wall thickness of 4 mm. It has two detachable flange connections at the entrances to the irradiation cell and pump.

The water is heated to a temperature above the critical point by the four-section external electric heater with a total power of up to 20 kW. Water circulation in the loop is due to natural convection, and can be adjusted by the influence of the 0.5 kW circulation pump. The upper part of the loop is cooled by a tubular cooler. Two versions of the SCWCL (with detachable flanges and all-welded) have been built. The experiment results with the all-welded SCWCL with four-channel irradiation cell (without recourse to the circulation pump) are given below.

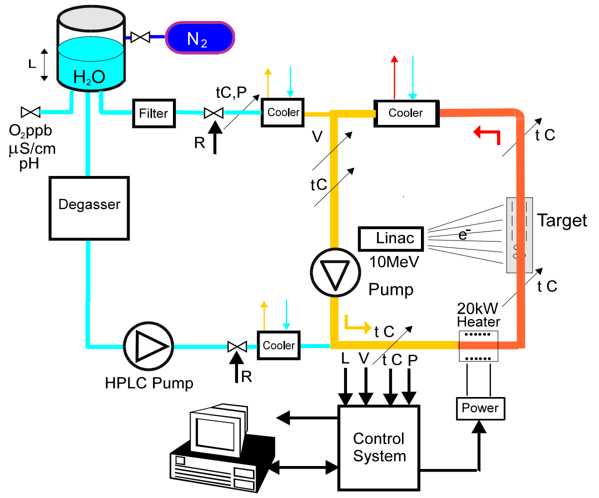

Fig. 2. Schematics of the CU-EITF design

For chemical analysis and degassing, a small portion of water is discharged from the loop through the capillaries, valves and filter into the accumulation tank partially filled with nitrogen. The recirculation of the water back into the loop after degassing is provided by the high-pressure pump (HPLC). The operation of automatic valves and pumps is controlled by an Intel™ CPU based personal computer. For the protection of the elements of the degassing line from overheating, capillaries are cooled with special coolers to a temperature below 100°C.

The control system is designed to provide routine measurements of pressure, flow rate and temperature at several points on the surface of the loop. The system regulates the supply of electric power in the heaters and controls the operation of pumps and valves. In case of an emergency, when the temperature and/or pressure of SCWCL components exceed the specified values, the control system disables the heaters, the linac beam current, and pumps. For extra protection of the loop, a mechanical safety valve that prevents overpressure beyond 27 MPa is installed. The view of the accelerator bunker is made with the help of a video camera.

The irradiation of specimens (as well as additional heating and ionization of the water) occurs in the irradiation cell CU-EITF. The cylindrical irradiation cell (IC) is an integral (but interchangeable) part of the CU-EITF SCWCL. Different ICs can be used in different simulation experiments depending upon the materials under investigation. Three kinds of the cylindrical IC are designed and produced (Figs. 3-6). The longitudinal size IC (310 mm) was determined by scanning system amplitude of the linac on the frontal IC surface.

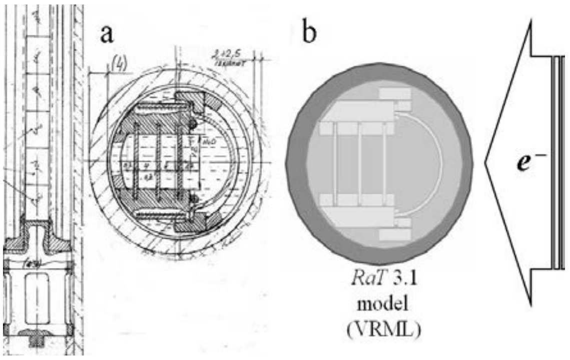

Fig. 3. The design drawings (a) and 3D computer model (b) of the CU-EITF irradiation cell

The thickness of the steel chamber front wall should be no more than 2 mm for the energy losses of the 10 MeV electron beam on the front surface IC were a minimum. At making of the IC cases from the titan alloys which density is twice below of the steel density, the thickness of a face-to-face wall of the chamber should not be more than 4 mm.

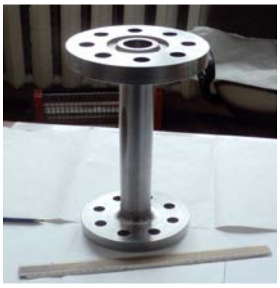

Fig. 4. The view of the one-channel irradiation cell The material cell is an alloy of titan VT22, the thickness of the cell wall – 4 mm, external diameter – 40 mm

Fig. 5. The view of the four-channel irradiation cell. The material cell is stainless steel 12X18H10, the thickness of the cell wall – 2 mm, external pipe diameter – 14 mm

Variant IC (see Fig. 5) with four-channel steel case without flanges (external pipe diameter – 14 mm, a wall thickness – 2 mm) has higher durability in comparison with IC (see Fig. 3). Reduction of the total area of internal section of this IC allows receiving higher linear speeds of water moving along samples (see Fig. 6).

Overlapping of a diagonal of section IC with an axis of a bunch creates the irradiation conditions at which samples in face-to-face tube IC test influence of water, primary beam electrons and gammas, and samples in a back tube practically are not irradiated by primary electrons at the same influence of water and gammas. Thus, such design IC allows estimating the primary electron contribution in destruction of an irradiated sample surface by corrosion (see Fig. 6).

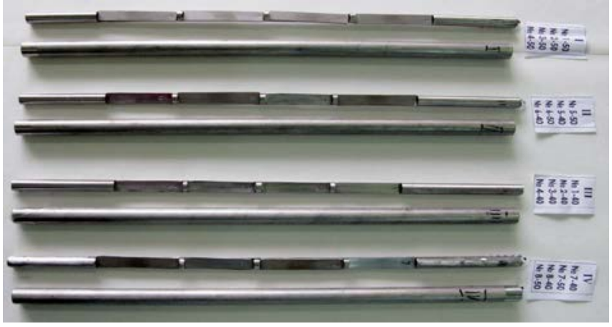

Fig. 6. The view of the consoles with the samples and the pipes of the four-channel irradiation cell

2. THE RESULTS OF THE SAMPLE IRRADIATION

When used the all-welded SCWCL "Loop-1a" with the four-channel irradiation cell, the first time in the world it was possible to receive the irradiation results of the Inconel 690 and Zr samples by electrons. The samples are irradiated at a pressure of 23.5 MPa and a temperature below 380ºC. The total session duration was 574 hours (including 497 hours with the electron beam), the maximum fluence on the irradiation cell surface was 10^{20} el/cm^2. The operating schedule SCWCL "Loop-1a" is shown in Fig. 7.

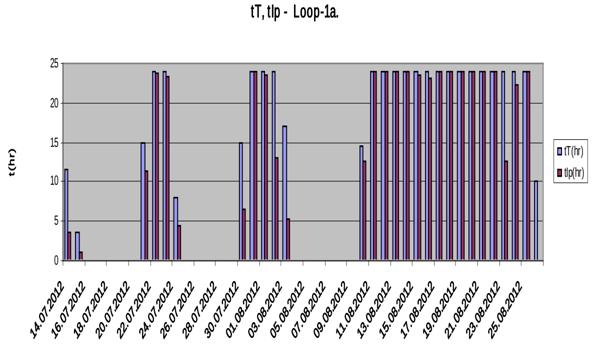

Fig. 7. The total operating time (tT) of the Loop 1a including the time with the electron beam (tIp). July-August, 2012

During the simulation experiments the irradiation cell CU-EITF was exposed to the electron beam of the LAE10 with the energy 10…11 MeV and the scanning frequency along the irradiation cell 3 Hz. The average electron beam current was below 1 mA. The electron beam impulse frequency was 250 Hz. The duration of the impulse was 3.4 μs. Average power of the accelerator electron beam is equal 7 kW at a long operating schedule. Actual parameters of long irradiation sessions can differ a little from designated above nominal parameters, they are checked by the automatic monitoring system of the linear accelerator.

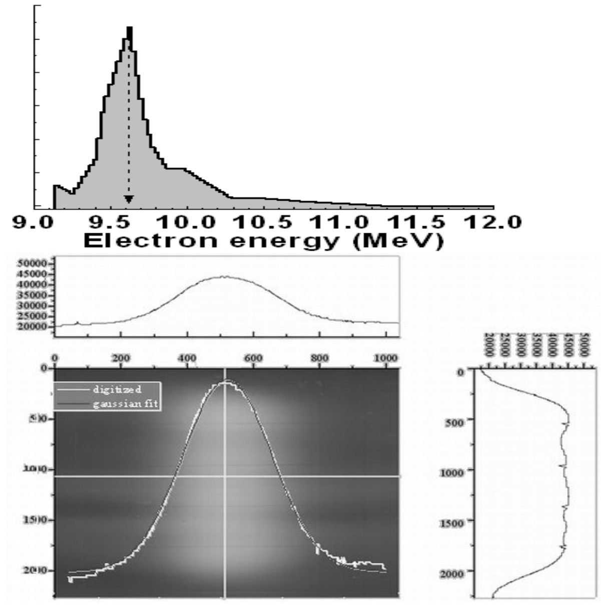

Fig. 8. The characteristic form of a electron energy spectrum of the accelerator LAE-10 in KIPT and the characteristic of distribution density of the scanned electron beam on the surface IC

For example, the 10 MeV energy spectrum, measured during experiment, is represented in a Fig. 8. It has the most probable energy 9.6 MeV with the long tail which has been stretched up to 13 MeV.

The loop was filled with water with average parameters pH = 6.5, conductivity 7…11 μS/cm, oxygen ppb = 0.5. Water from a loop acted with rate of 5 ml/mines. Time of delivery of tests on a 30-meter water-main was 5 hours. Parameters of water were pH = 5.3…6, conductivity 7…23 μS/cm, oxygen ppb = 3…4 on an output from a water-main. After decontamination water came back in the loop. The gain of conductivity was about 0.1 µS/cm at one hour, i.e. the salt amount in water increased. At carrying out of the element chemical analysis of structure of tests of water by means of spectrometer ICPE-9000 of firm “Sumadzo” it is established, that practically at all tests of water from a loop there is a chrome (8…54 µg/l) and there is no zirconium. The isotope structure of samples after an irradiation is resulted in Table.

Samples	Isotops half-life period)
Zr	Zr^{89}(78 h), Zr^{95}(64 d), Nb^{92}(10 d), Nb^{95}(64 d), Nb^{95m}(86 h)
Inconel 690	Cr^{51}(27 d), Mn^{54}(312 d), Co^{57}(273 d), Co^{58}(70 d), Nb^{92}
Steel 12X18H10T cases	Cr^{51}, $Mn^{54,}$ Co^{57}, Co^{58}

CONCLUSIONS

Supercritical Water Convection Loop (SCWCL) with the irradiation cell, connected with the electron accelerator LAE-10 (10 MeV energy, below 10 kW power), was developed specially. Rather short time of the creation of the plant and low cost (in the comparison with nuclear reactors) and an opportunity to investigate the irradiated materials from hot chambers do the offered technique of imitating experiments by very effective tool for obtaining of the necessary data to choose the materials for active zone SCWR.

The basic results:

- For the first time the Supercritical Water Convection Loop with the four-channel irradiation cell from the steel, filled by the samples of two types (from the Inconel 690 alloy with double-layer fusing from the In 52MSS wire and from the Zr alloy), was successfully tested on an electron irradiation. The total session duration was 574 hours (including 497 hours with the electron beam), the maximum fluence on the irradiation cell surface was 10^{20} el/cm^2. Operating mode of a loop – P = 23.5 MPa, the maximal temperature on a irradiation cell surface up to 380ºC.
- During sessions of an irradiation the maximal gain of weight on Zr samples is 0.75 mg/cm^2, and on Inconel 690 samples is 1.5 mg/cm^2 as a result of corrosion.
- During an irradiation parameters of water from a loop changed within the limits of: pH – 5.3…6, conductivity – 7…23 µS/cm, oxygen ppb – 3…4.
- Change of water conductivity in the loop during the irradiation was neaby 0.1 µS/cm at one hour, i.e. the impurity quantity in water was increased.
- In particular the increase Cr amount in water for the same period was about 0.1 µg/l at at one hour.

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ОБЛУЧЕНИЕ ЭЛЕКТРОНАМИ ОБРАЗЦОВ МАТЕРИАЛОВ ЯДЕРНЫХ РЕАКТОРОВ НОВОГО ПОКОЛЕНИЯ В СВЕРХКРИТИЧЕСКОЙ ВОДЯНОЙ КОНВЕКЦИОННОЙ ПЕТЛЕ

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Суперкритический водно-охлаждаемый реактор (SCWR) – одна из самых многообещающих реакторных технологий в программе реакторов IV поколения. С 2009 года в Харьковском физико-техническом институте ведутся работы, направленные на развитие оборудования и методологии для оценки реакторных материалов, предназначенных для реакторов SCWR (проект УНТЦ - P4841). Специально разработанная в ХФТИ суперкритическая водяная конвекционная петля с камерой облучения, связанная с ускорителем электронов ЛУ-10 (8…10 МэВ, до 10 кВт) предоставляет возможность для изучения коррозии и механических повреждений материалов при облучении пучком электронов. Приводятся результаты 500-часового сеанса облучения образцов циркония и инконеля.

ОПРОМІНЕННЯ ЕЛЕКТРОНАМИ ЗРАЗКІВ МАТЕРІАЛІВ ЯДЕРНИХ РЕАКТОРІВ НОВОГО ПОКОЛІННЯ У НАДКРИТИЧНІЙ ВОДЯНІЙ КОНВЕКЦІЙНІЙ ПЕТЛІ

О.С. Бакай, В.М. Борискін, М.І. Братченко, Є.З. Біллер, П.А. Бутенко, В.О. Бочаров, В.М. Верещака, А.М. Довбня, С.В. Дюльдя, Ю.В. Горенко, Г.Г. Ковальов, В.О. Момот, О.О. Репихов, С.К. Романовський, А.М. Савченко, В.В. Селезньов, В.І. Солодовніков, В.І. Тітов, О.В. Торговкін, В.В. Хандак, С.В. Шелепко, Г.М. Цебенко

Надкритичний реактор з водяним охолодженням (SCWR) – одна з самих багатообіцяючих реакторних технологій в програмі реакторів IV покоління. З 2009 року в Харківському фізико-технічному інституті ведуться роботи, спрямовані на розвиток обладнання та методології для оцінки реакторних матеріалів, призначених для реакторів SCWR (проект УНТЦ - P4841). Спеціально розроблена в ХФТІ надкритична водяна конвекційна петля з камерою опромінення, зв’язана з прискорювачем електронів ЛП-10 (8…10 МеВ, до 10 кВт), дає можливість для дослідження корозії та механічних пошкоджень матеріалів після опромінення пучком електронів. Приводяться результати 500-годинного сеансу опромінення зразків цирконію та інконеля.