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Summary of Works Over MARIA Reactor Core Conversion From HEU to LEU Fuel

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ABSTRACT

Conversion of MARIA reactor core from HEU to LEU fuel has imposed on performance a number of serious challenges. The list of those works enclosed a number of consecutive projects to be developed (project of INVAP, 2 projects of CERCA – AREVA), execution of calculations and safety analyses which exhibited among others, the necessity for reactor pumping system upgrading, carrying out the test irradiation of new fuel and conducting post-irradiation examination of this fuel. The positive results of test fuel elements made possible to start full core conversion with LEU fuel.

1. Introduction – brief information on MARIA reactor

Reactor was put into operation at the first time on December 18th 1974. MARIA reactor is pool type reactor with pressurized fuel channels. Water and beryllium are used in reactor as moderator and graphite is used as a neutron reflector.

Current basic characteristics and data for MARIA reactor:

•	rated power	30 MW
•	maximum thermal neutron flux	$3 \cdot 10^{14} \text{ n/cm}^2 \cdot \text{s}$
•	moderator	H_2O , Be
•	fuel cooling system	pressurized channels
•	type of fuel element (MR)	- materialUO ₂ -Al.
		- enrichment 36%
		- cladding Al
		- shape: concentric tubes
		- length of fuel region 1000 mm

The MARIA reactor started its operation on high-enriched fuel (80%). During its history the reactor underwent a number of fuel conversions resulting in enrichment reducing. In Table 1 are enclosed the most characteristic features of the conversions.

Туре	Operation period	Material	Enrichment %	Mass of ²³⁵ U	Density of U g/cm ³	Cladding mm
MR	1974÷1999	UAl _x	80	350	1.2	0.8
MR	1999÷2005	UO ₂	36	540	2.3	0.6
MR	since 2005	UO_2	36	430	2.8	0.75
мс	since 2012	U ₃ Si ₂	19.75	485	4.8	0.6
_	(tests 2009÷2011)	5 4				
MR	since 2014	UO_2	19.7	485	3.8	0.6
	(tests since 2012)	- 2				

Table 1. Fuel conversions in MARIA reactor

The way of reactor fuel cooling is a feature of fundamental importance for the conversion process. The diagram for cooling the fuel channels and coolant distribution between fuel tubes are shown in Fig. 1. When assuming that fuel conversion proceeds gradually, i.e. the fuel elements of old type are replaced successively by the new fuel elements the parallel layout of fuel channels forces preserving the hydraulic resistances for both fuel types.

The mode for fuel element cooling (downwards flow between outer fuel tubes and upwards flow between the inner tubes) is imposing the demand to maintain the leak-tightness of the flow separation tube.



Fig.1. Schematic diagram of fuel channel cooling

2. Low enriched fuel (LEU) for MARIA reactor

The works over reactor conversion on LEU fuel were launched in 2005. Preliminary physical calculations unveiled that to maintaining physical features for the core with 20% enrichment it is necessary to include in fuel element 485 g of 235 U. In association with necessity to preserve hydraulic characteristic of fuel element it brought about to substantial increase of uranium density in the fuel element. The most suitable option was to apply the silicide fuel with uranium density of 4.8 g/cm³.

The company Areva (CERCA) took up production of silicide fuel for MARIA reactor.

Characteristic of MC fuel (CERCA)

The fuel to be used in MC fuel elements is a uranium silicide dispersion U_3Si_2 (uranium/silicon alloy) in aluminum matrix. The fuel enrichment in ²³⁵U is 19,75%. The MC fuel element consists of 15 curved fuel plates connected in 5 fuel tubes bundle (the numbering of the tubes is identical as for the MR fuel; the tube #1 has been in the MC fuel left out). The plates composing the fourth tube (separation tube) are longitudinally welded. As regards the remaining tubes the assemblies of three bended fuel tubes they are longitudinally kneaded on special connectors. There are two independent sets of connectors: inner - biding the tubes #2 and #3 and outer - binding the tubes #5 and #6. The missing fuel tube #1 in MR fuel is substituted in the MC fuel element by aluminum tube fulfilling the role of filler.

Discontinuity of fuel layer in tubes, lack of fuel tube #1 and attendance of linking connectors (stiffeners) along the whole length of fuel element is responsible for substantial differences in hydraulic and thermal characteristics of MR and MC fuel (Table 2).

Parameter	MR		МС	
Water gaps	Inner	Outer	Inner	Outer
Hydraulic diameter [mm]	5	5	4.49 ÷ 4.71	4.77 ÷ 4.82
Water gaps surface [mm ²]	1005	1496	724	1361
Flow rate through the FA [m ³ /h]	25		30	
Average water velocity [m/s]	↑ 6.8	↓ 4.7	↑ 9.2	↓ 5.1
Heat exchange area [m ²]	1.72		1.29	

Table 2. Essential characteristics of the MR and MC fuel elements

Construction of the MC fuel in the form of bent plates connected with stiffeners creates a source of thermo-mechanical stresses. Calculations made in NRG (Petten, the Netherlands) by means of CFD code (Fluent Ver. 6.2) shown that the critical stresses are arisen in upper part of fuel plates' joints with linking connectors (Fig. 2). The results of these calculations made possible to develop a new project assuming a mechanical separation of linking connectors of inner tubes (#2, #3) from outer tubes (#5,#6) in the final version for MC fuel.





Fig. 2. Stresses [Pa] in upper part of the fuel element (the first version of MC fuel)

3. Irradiation and Post Irradiation Examination of the Lead Test Assemblies (LTA)

Course of irradiation of the LTAs

In accordance with the approved procedure the irradiation of 2 MC test fuel elements (LTAs) was initiated in August 2009 and pursued until the preset burnups levels was achieved. The MC001 fuel element was irradiated until it achieved the burnup of 5899 MWh i.e. by 21/08/2011, whilst the MC002 fuel element till 17/08/2010, i.e. after achieving the burnup of 4025 MWh.

On the basis of measuring results, pursued during the test irradiation of LTAs one can deduce the following conclusions:

- Technological measurements conducted by means of the technological control system (SAREMA) didn't show any abnormalities. It has not been any other effects which could indirectly indicate an abnormal behavior of any fuel element during the tests (it refers to all fuel elements and not only to investigated MC). Among other it pertains to such phenomena as: power fluctuations, unstable operation of automatic power regulation of the system etc.
- Indication of the Fuel Integrity Monitoring System (FEIMS) were substantially lower than the limit value and they didn't depart from the indications for the remaining fuel elements of the MR type.
- Spectrometric measurements of water contamination in the fuel channel cooling circuit unveiled little level of fission product contamination, however of two orders smaller than in the years 2000 ÷ 2004, when it was used the MR-6/540 fuel was being used.

Visual inspection of irradiated LTAs

On accomplishing the irradiation of the test fuel elements and their cooling period the visual control of both MC fuel elements was performed in April 2011. To achieve this target a special out of reactor stand was constructed in the MARIA reactor spent fuel storage pool. This stand enabled to carry out a remote observation and recording of the fuel element surface. To make a comparison the burnt MR fuel element was subjected to observation too.

The tube surface of the MC002 fuel element, which achieved the burnup of 4025 MWh, is covered by visible and well preserved oxide layer of differentiated thickness. The MC001 fuel element was subjected to significantly greater burnup (~5900 MWh) which caused an oxide layer breakaway on a significant part of fuel side surface (Fig. 3). The separation of the oxide layer occurs at the central part of the tube (photography b), where during the fuel element irradiation appeared the greatest wall temperatures: on the upper and the lower edge of the fuel tube the oxide layer has been well preserved (photography a). Apart from the separated oxide layer on the external surface of the MC001 element fuel tube (it refers surely to the remaining, invisible surfaces) there have not been noticed any deformations or other changes.



Fig.3. Photographs of external side of the MC001 fuel element

An accumulation and separation of the oxide layer on cladding surface it has been also observed in the MR fuel. Similarly as in the MC fuel this effect appears in the central part of the fuel in which the cladding temperatures are pretty high. In the upper and lower part of the external fuel tube the oxide layer remains intact.

It has been also carried out the observations of the test MC fuel elements from the top. This observation was aimed to check if there have not appeared noticeable shape deformations of the fuel tubes and water gaps between the tubes. These observations were difficult to be realized due to the presence of intensive Czerenkov's radiation, which makes impossible to have a look into the intertube space on a larger depth. It has not been observed any noticeable shape deformations of the fuel tubes and water gaps between the tubes.

Oxide layer on fuel cladding surface

Pending the reactor operation a layer of hydrated aluminum oxide of low thermal conductivity is formed on the fuel elements' cladding surface.

The samples of oxide layer were taken from the MC001 fuel element (Fig. 4). Approximate thicknesses of these samples are within the range $30 \div 50 \ \mu m$, which confirms that the detachment of oxide layer takes place on achieving by these layers the thickness layer are of order of several tens μm .



Fig. 4. Sample of oxide layer

The investigation of structural phase of oxide sample taken from the MC001 fuel element surface performed by means of X-ray diffraction exhibited that the layer composition includes: bayerite

 α -Al(OH)₃ (63.8%), gibbsite α -Al(OH)₃ (15.6%) and boehmite α -Al(OH) (2.8%). This oxide layer accrues gradually with time and for a certain time it adheres to the cladding. On achieving a certain thickness (several tens of μ m) this oxide layer breaks away from the cladding, whilst the accretion process of oxide layer is starting anew.

There have been also conducted the measurements of oxide layer thickness on outer tubes of LTAs by means of eddy current method. The gauge to be used for these measurements required preparing of the calibration standards fabricated from aluminum tubes with overall dimensions of the fuel tube #6. Preparation of these standards was quite difficult to be manufactured due to necessity of forming thick oxide coatings as well as bearing in mind the standard curvature. In Fig. 5 are depicted a vertical distributions of oxide layer thickness of LTAs. For comparison the theoretical thicknesses of oxide layers (solid lines) are also shown.



Fig. 5. Axial oxide layer distributions on LTAs

An impact of oxide layer on heat removal conditions in the fuel is a subject of separate presentation (S. Suchcicki at al.: Investigation of Oxide Layer Impact on Heat Transfer in MARIA Reactor Fuel Elements).

4. Modernization of the fuel channel cooling circuit

Hydraulic measurements of the MC fuel performed on out-of-reactor stand proved that the hydraulic resistance for the new fuel is considerably larger than the flow resistance in MR fuel. This effect is even more highlighted because of demand to increasing the coolant flow rate (up to $30 \text{ m}^3/\text{h}$) through the MC fuel elements due to smaller heat exchange surface compared to MR fuel.

The pumping system currently operating doesn't provide the possibility to cool the MARIA reactor core with the MC fuel and hence the need of its modernization. The modernized cooling system will be subjected to substantial qualitative upgrading to be aimed on improving reactor safety and system operational reliability. The basic parameters of the new pumping system is collated in Table 3.

Deremeter	Pumps			
Parameter	Main units	Shutdown units		
Number of installed assemblies	4	3		
Number of operating assemblies	2	2		
Flow rate [m ³ /h]	400	70		
Pressure head [H ₂ O m.col.]	128	12		
Demand of power supply [kW]	179	3.1		
Motor power [kW]	200	4		

Table 3. Parameters of the pump assemblies of the MARIA reactor fuel channel cooling circuit to be upgraded.

Modernization of the fuel channel cooling circuit leads, among others, to increasing of coolant flow intensity in the circuit. One can anticipate that this change will cause changes in mechanical vibrations of the circuit pipelines. The tubes vibration can be also affected by changes of the pump system, thereby an introduction of new tube joint to link the shutdown pumps with the rest of the circuit. An assessment of modernization influence on piping vibrations requires to defining a pattern of tubes' vibrations before the modernization. In order to attain this there have been appointed the reference points on the delivery collector in which the vibration measurements were made.

5. Safety analyses

Neutronic calculations

Neutronic calculations of the MARIA reactor core conversion campaign from the MR fuel into low enriched MC fuel were carried out in two stages: in the first stage the first consecutive steps of the campaign were performed, in the second one the detailed calculations of the kinetics parameters, reactivity temperature coefficients and efficiencies of absorbing rods were accomplished.

On the stage of the successive conversion steps the following assumptions were taken into consideration:

- The fuel loading and shuffling operations in the core have to be scheduled in such a way that the reactivity excess in the core by the end of an operation cycle (after 3 days from the reactor start-up) will be within the ranges $1.5 \div 2.0$ \$.

- Each fourth reactor operation cycle is to be called the molybdenum cycle, i.e. in the core there are 2 channels (positions f7 and i6) with uranium plates for ⁹⁹Mo production.
- Maximum thermal power of the most thermally loaded fuel channel is 1.8 MW in molybdenum cycle and 1.6 MW in the remaining cycles.

Neutronic calculation were performed by means of the diffusion code REBUS with library of cross-sections to be calculated by means of the WIMS-ANL code. In calculations the burnup and poisoning of the fuel as well as the poisoning effect of beryllium were taken into consideration.

Thermal-hydraulic calculations at steady-states

The calculations of the steady-states have been performed for various thermal powers and coolant flow rates. There were being assessed the relations of channel thermal power as a function of coolant flow rate preserving the two criteria:

- 1. exclusion of the film boiling on cladding or
- 2. parameter ONBR = 1.2,

and for three sets of parameters: nominal ones, with accounting for the uncertainty factors by means of the statistical method and the method of cumulating the uncertainties.

Analysis of unsteady states

Among reactor unsteady states to be analyzed in connection with conversion of the core from MR fuel on MC fuel of significant importance are those states in which the specifics of MC fuel, i.e. the terms for heat exchange and balance of fission products activity is manifested. It is also necessary to take into account the modification of the fuel channels' cooling circuit. Due to that the verification of analyses is needed for the following groups of unsteady states:

- decay of coolant flow rate in the channel circuit,
- blocking of flow rate through the LTA fuel channel and in a consequence its burnout,
- loss of tightness of the fuel channel cooling circuit (LOCA),
- insertion of the positive reactivity and relevant to this the changes of power during reactor start-up and at operation on full power.

Neutronic calculations pursued for conversion process unveil that the basic neutron parameters of the MARIA reactor don't undergo any significant variations in comparison with the MR fuel. In case of temperature coefficients of void and fuel where exhibit a distinct tendency to increasing the values of these coefficients that correspond to the core with MR fuel.

In comparison with analyses having been conducted so far for unsteady states for reactor with MR fuel, the analyses for reactor with MC fuel and upgraded pumping system for fuel channel cooling circuit don't lead to the significant quantitative and qualitative changes.

Exception are as follows:

- the transient caused by loss of coolant flow (LOFA, disengagement of the main pumps due to loss of main power supply),
- the transient to be caused by leakage of coolant from the fuel cooling circuit (LOCA).

6. Conversion procedure

The conversion process from HEU (MR) to LEU (MC) fuel launched in the MARIA reactor on 24th September 2012. In the conversion procedure the following assumption were taken into considerations:

- MR and MC refueling will be proceeded gradually. From the moment of commencing the conversion, the burned MR fuel will be unloaded from the core whereas the core will be refueled with a fresh MC fuel.
- It is to be admitted to have in the core the two LEU fuel elements MR-6/485 of Russian production during the core conversion. From the reactivity point of view there are to be equivalent to the MC fuel elements, however, from the heat transfer view they correspond to the standard MR fuel elements.
- There will be met all criteria for the safe reactor operation, particularly reactivity, thermal-hydraulic, and radiological limits. It is to be allowed to diversify coolant flow rates for the MR and MC fuel elements; the rated coolant flow rates through the MR fuel channel are to be lower than 25 m³/h and in the MC fuel 30 m³/h.
- While doing the core conversion, the reactor will pursue the normal investigation and production activity. Among others there will be irradiated uranium plates to be used for production of molybdenum-99.
- Procedure for core conversion from the MR fuel onto the MC fuel have been modeled by calculations. In calculation was assumed, that every fourth cycle of operation will be used as a cycle with irradiation of uranium plates for production of ⁹⁹Mo. The full core conversion has been achieved within the span of 43 operation cycles.
- Commencement of the conversion process from the MR fuel into the MC fuel can be launched with an unmodernized pump system. As the experiment with the test fuel elements has unveiled it is possible with presence of at least two MC fuel elements in the core.
- Each fresh MC fuel element to be loaded into the core will be monitored in a continuous way by means of the system FEIMS during the first operation cycle. The remaining fuel elements will be monitored by the system FEIMS according to the standard procedure.
- For the cores with 11 (approximately half load) and 21 (full load) MC fuel elements the calibration of absorbing rods PK, PAR and PB will be performed.