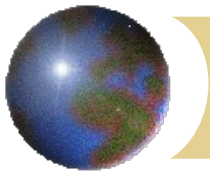


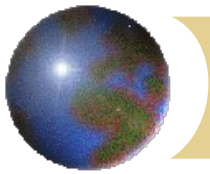
*Results of ISTC Project #2680 - MATINE .
Status of ISTC Project #3608 - MATINE2 .*

CEG Annual Meeting , January 22 –23, 2007
Brussels, BELGIUM



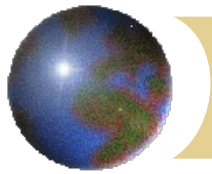
*“MATINE-Study of Minor Actinide Transmutation in Nitrides:
Modeling and Measurements of Out-of-pile Properties“- #2680*

- On September 30, 2006 the Project has been completed (after 2.5 years of activity)
- Objective - to identify the optimum fuel pin design that would perform well under irradiation in ADS fast neutron spectrum
Fuel (Pu,Am,Cm,Zr)N (with ZrN=50-65at%,
Pu/Am/Cm=40/50/10)
- Three Institutes –IPPE (Obninsk), VNIINM (Moscow), RIAR (Dimitrovgrad) - participations. CEA (France) and KTH (Sweden) - collaborators.



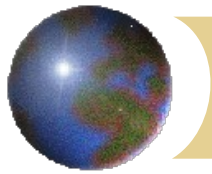
MATINE Scope of activity

- Task 1:** Compilation and analysis of literature data on nitride fuels and on MA fuels. The objective is to perform input data for calculation modeling of fuel performance.
- Task 2:** Experimental study of out-of-pile PuZrN properties: high temperature stability, thermal conductivity, high temperature creep, linear expansion.
- Task 3:** Modeling of (Pu,Am,Cm, Zr)N behavior under irradiation up to 35at% in ADS with Pb-Bi coolant (pellet for helium, sodium and lead-bismuth bonded fuel pin, vibro for helium-bonded pins only).
- Task 4:** Technical – economical assessments of fabrication feasibility of (Pu,Am,Cm, Zr)N with (60 ± 10) at% ZrN and with 10 at% Cm at RIAR site.



TASK 1 (IPPE, VNIINM, RIAR)

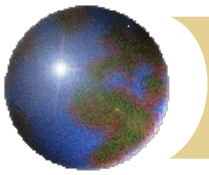
- ⊕ Review of literature data on (UN, UPuN, UN+ZrN, PuN+ZrN) properties and on fuel with MA. Few data for ZrN-based fuel.
- ⊕ Empirical relationships on (Pu,Am,Cm,Zr)N properties are recommended: maximum permissible temperature; Young, Poisson modula; thermal expansion coefficient; creep rate; thermal conductivity.
- ⊕ Absence of necessary data complicates correct prediction of ADS fuel performance. First of all, it is He release under cladding and its retention in fuel matrix that determines fuel swelling which, in turn, determines fuel pin life time for dense fuel.
- ⊕ Experimental data of ZrN fuel swelling are practically absent. We used model of spherical cells (SC) developed in IPPE for dispersion fuel compositions and model of spherical gas pores (SGP) usually used for dense fuels. The last one is adapted for our fuel on base of UN irradiation in various experimental reactors (see Task 3).
- ⊕ Fuel swelling depends essentially on fuel creep. Fuel creep has also determining influence on FCMI as well. There are no creep data of ZrN based fuel. The original data on ZrN-based fuel creep and thermal expansion have been received in Task 2. The recalculation was done using new results on ZrN fuel creep received in Task 2.



TASK 2 (VNIINM) –Fuel fabrication

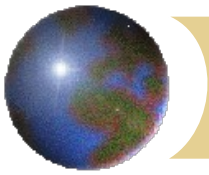
(Pu,Zr)N samples with ZrN=60mol% of 85%, 93% density are fabricated by mixing of PuN and ZrN powders by patented electro-vortex blending method (rotating ferro-magnetic needles) - **solid solution received**. PuN powder was made using hydriding-nitriding method, ZrN - commercial powder .

At initial stage the fabrication technology was finalized using zirconium nitride as a dummy.

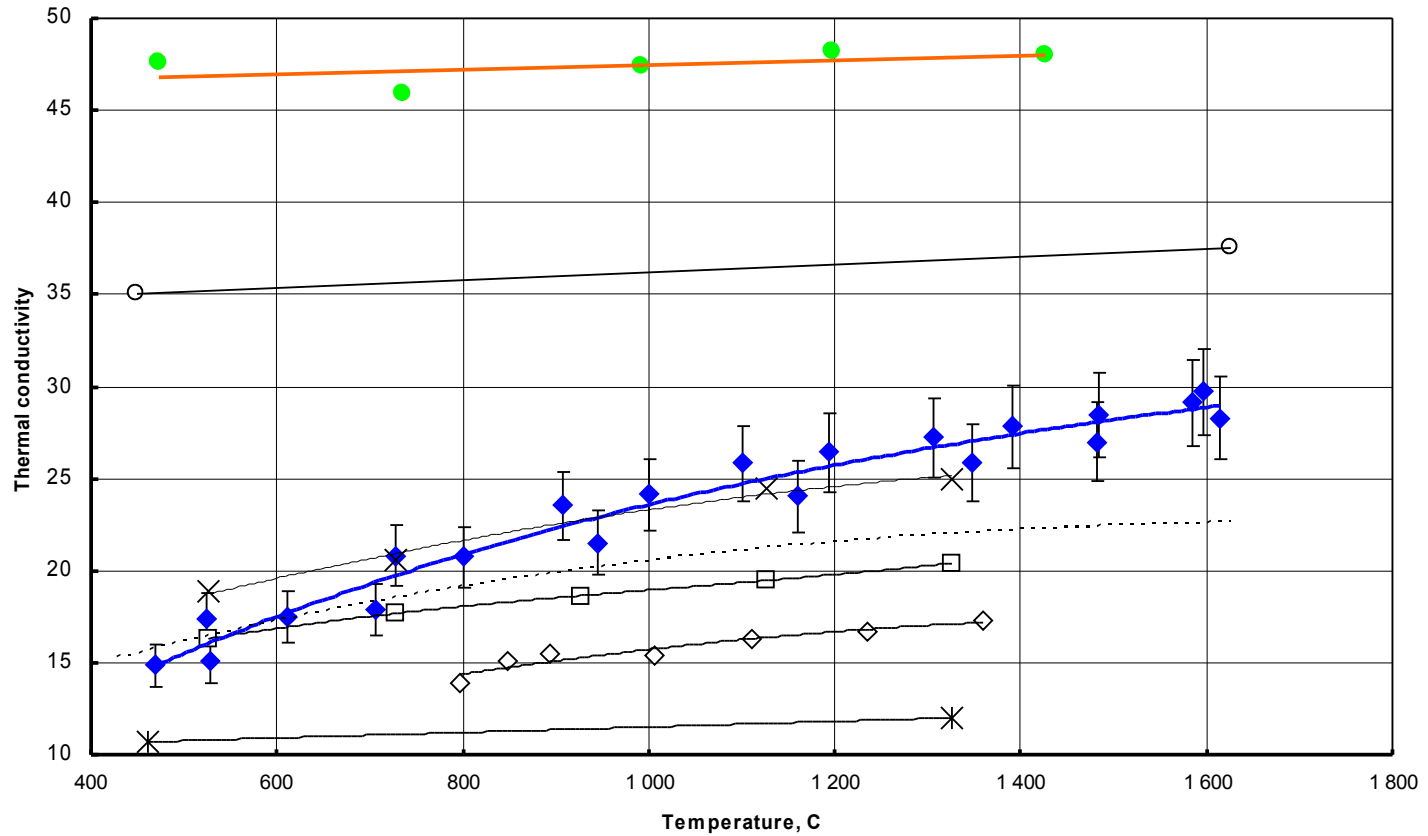


Task 2 - PuZrN Thermal conductivity

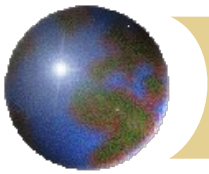
- ⊕ Measurements of PuZrN thermal conductivity has been done by laser flash method in vacuum (at least $1 \cdot 10^{-5}$ mm Hg) at temperature range of 400°C to 1600°C
- ⊕ Certain discrepancies between thermal conductivity of PuZrN investigated in this research and respective foreign data is received (/4,9/ - V. Basini et al; /11/ Y.Suzuki). This can be explained by different physico-chemical and structural properties of the samples due to difference of sample fabrication processes.



Task 2 - PuZrN Thermal conductivity



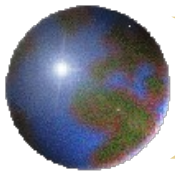
- ◆ - $(\text{Pu}_{0,4}\text{Zr}_{0,6})\text{N}$, (in the present work); □ - $(\text{U}_{0,8}\text{Pu}_{0,2})\text{N}$, (work [8]); ○ - ZrN, (work [9]);
× - UN, (work [8]); * - PuN, (work [10]); ◇ - $(\text{Pu}_{0,36}\text{Zr}_{0,64})\text{N}$, (work [11]);
● - ZrN, (in the present work); — $(\text{Pu}_{0,25}\text{Zr}_{0,75})\text{N}$, (work [4]).



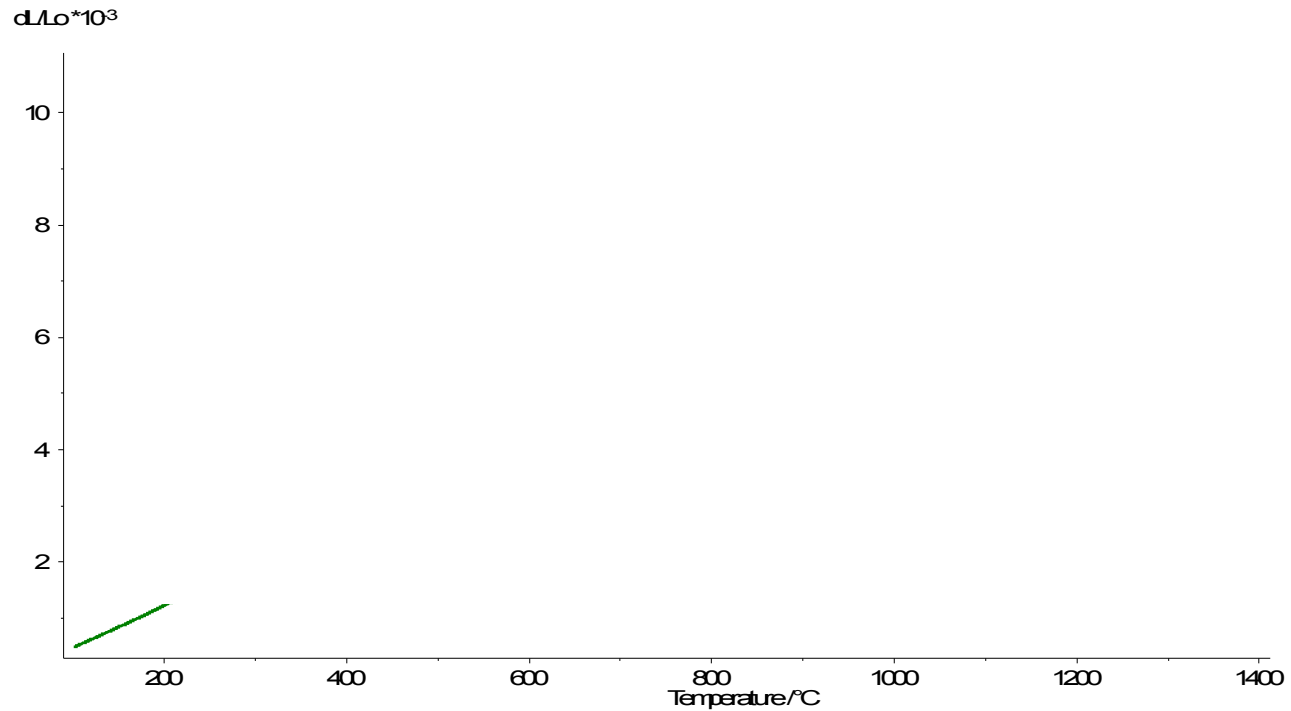
Task 2 - High-temperature stability and linear expansion

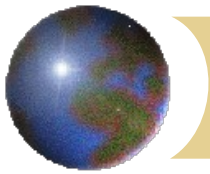
- ⊕ **High-temperature creep** - method of uni-axial compression at fixed temperatures of 1250°C to 1450°C in very-high-purity argon. The experimental data at steady-state phase depending on temperature are satisfactorily described by the Arrhenius equation. The creep rate increases linearly with load increasing. It is about one order lower than for UPuN.

- ⊕ **High-temperature stability** study revealed differences in behavior of (Pu,Zr)N at temperatures from 2200°C to 2300°C in various environments:
 - in vacuum fuel stability was the lowest. Presumably, Pu evaporated through matrix open porosity ;
 - in argon and nitrogen the samples revealed high stability, however, separation of phases with high Pu concentration was observed.



Coefficient of thermal expansion (CTE) was measured using Netzsch DIL 402C dilatometric system in argon + hydrogen. Maximum temperature – 1400 °C. Good agreement with literature data for ZrN.

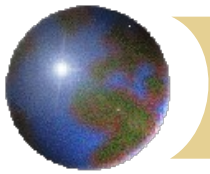




TASK 3 –IPPE, Obninsk.

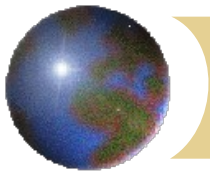
Modeling of He- bonded pin performance with pellet fuel

- ✦ Calculations are done by **DRAKON-3D** code developed in IPPE on the basis of KONDOR -2D code. For calculation of ADS fuel pins with (PuAmCmZr)N fuel - code modernization. New version - DRAKON - 3M.
- ✦ Modernization: calculation scheme improving (automatic transition from w/o contact loading scheme to a contact one), in development of temperatures calculation module, module of fuel swelling and fission gas release.



Task3 - Fuel swelling models

- ✦ For calculation of (PuAmCmZr)N pellets swelling two models are used: model of spherical cells (SC) and model of spherical gas pores (SGP).
- ✦ Model of SC is usually used for dispersion fuels swelling estimation. It is supposed, that fuel consists of identical cells, regularly located on fuel volume. Each cell is thick-walled spherical cladding made of matrix material with spherical grain of fissile material inside.
- ✦ Model of SGP - fuel (mixture of matrix and fissile part) consists of identical spherical cells with gas pore inside.
- ✦ Calculations results for SGP model were compared with nitrides PIE data on swelling and gas release. It was concluded that (PuAmCmZr)N swelling is correctly described by semi-empirical SGP model. Additional verification was done using PIE of PuZrN fuel irradiated in BOR-60 in frame of CEA (France)-Rosatom experiment BORA-BORA.



Nitride swelling

△ UN, max fuel temperature 1675 K, density 93%, ○ UN, max fuel temperature 1460 K, density 95%, ◇ UN, max fuel temperature 1173 K, density 84-94%,
Calculation at max fuel temperature 1675 K - ▲, at 1460 K - ●, at 1173 K - ◆.

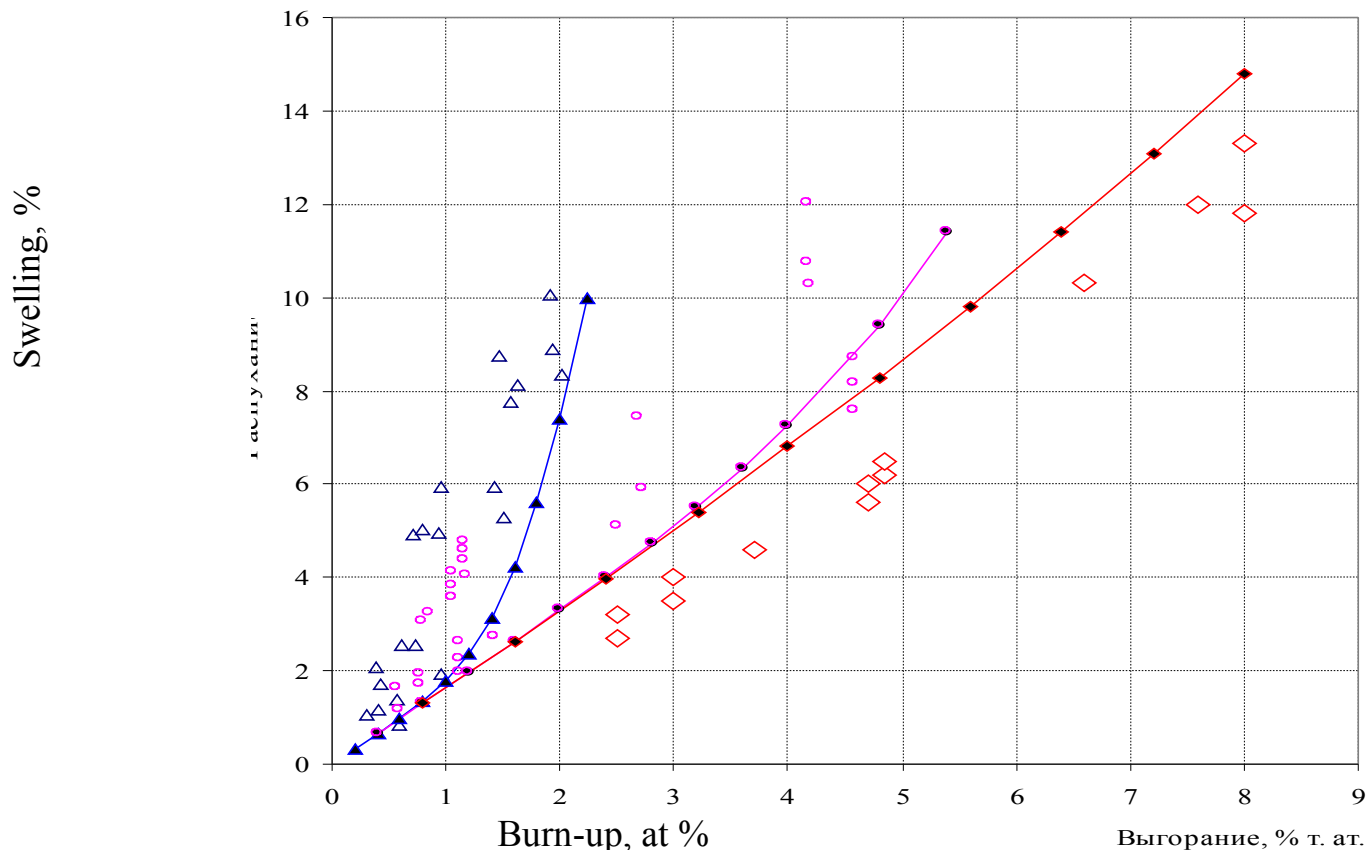
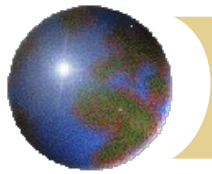


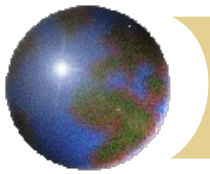
Рис. 4.2. Распухание нитридного топлива

△ UN, макс. температура топлива 1675 K, плотность 93 %, исслед. реактор [5]

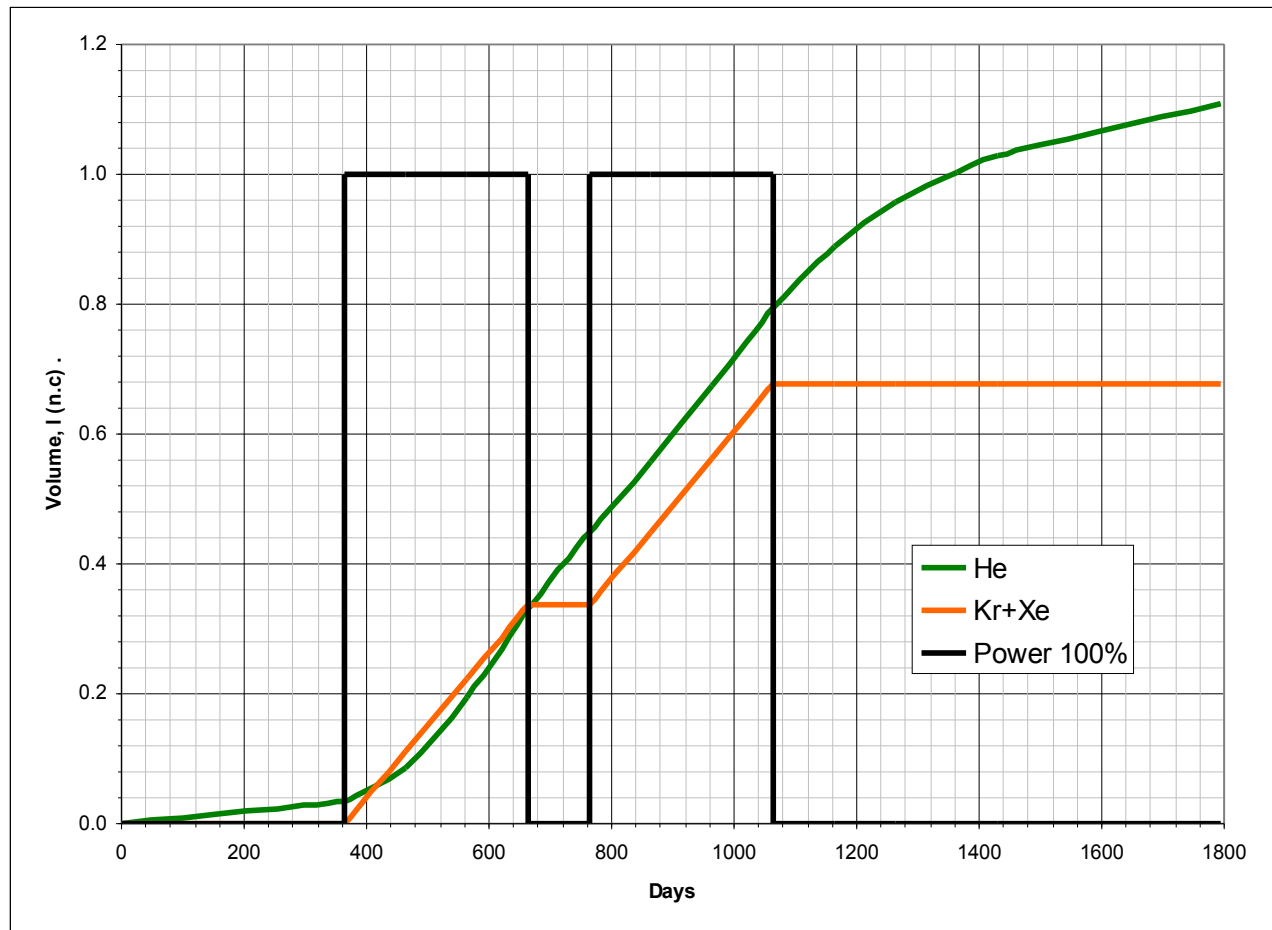


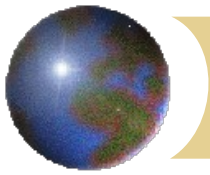
Task 3- Preparation of initial data

- ✦ Initial data (fuel pin and assembly sizes, core lay-out, irradiation history, content and density of fuel) have been prepared by Jan Wallenius (KTH, Sweden - Project collaborator). Its discussion have been held on December 15-18, 2003 at 14-th meeting of working group №1 (WG1) in Cadarache.
- ✦ With purpose of specification of fuel burn - up value and also for estimation of fission gas and He production values in ADS pin with (PuAmCmZr)N neutronic calculations by CARE code are done.
It is shown that He production starts before fuel core loading and proceeds after its unloading. At EOL (33.5at%) He content almost on 20 % exceeds fission gas amount.



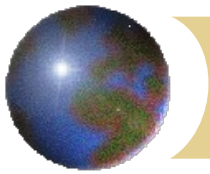
He production for most hot ADS fuel pin with (Pu,Am,Cm,Zr)¹⁵N fuel - inner zone fuel pin with ZrN=65 %, Pu/Am/Cm=40/50/10.





Task 3 - Preparation of initial data.

- ⊕ EP-823 ferritic-martensitic steel - cladding material.
- ⊕ On base of domestic experience on EP-823 steel corrosion behavior in Pb-Bi and also on PIE data of ferritic-martensitic steels the following requirements for ADS fuel irradiation parameters have been taken:
 - inlet coolant temperature $\geq 330-350$ C,
 - maximum cladding temperature (internal surface) ≤ 550 C,
 - coolant rate ≤ 2.5 m/c.



Task 3- Thermohydraulic calculations

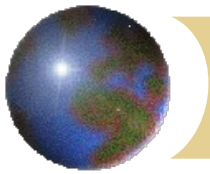
Calculations have been carried out by **FACIC code** developed in given Project.

Calculation showed that optimization of pin bundle geometry is effective means of decreasing of max cladding temperature and elementary cells heat-up. The following ways of optimization are considered:

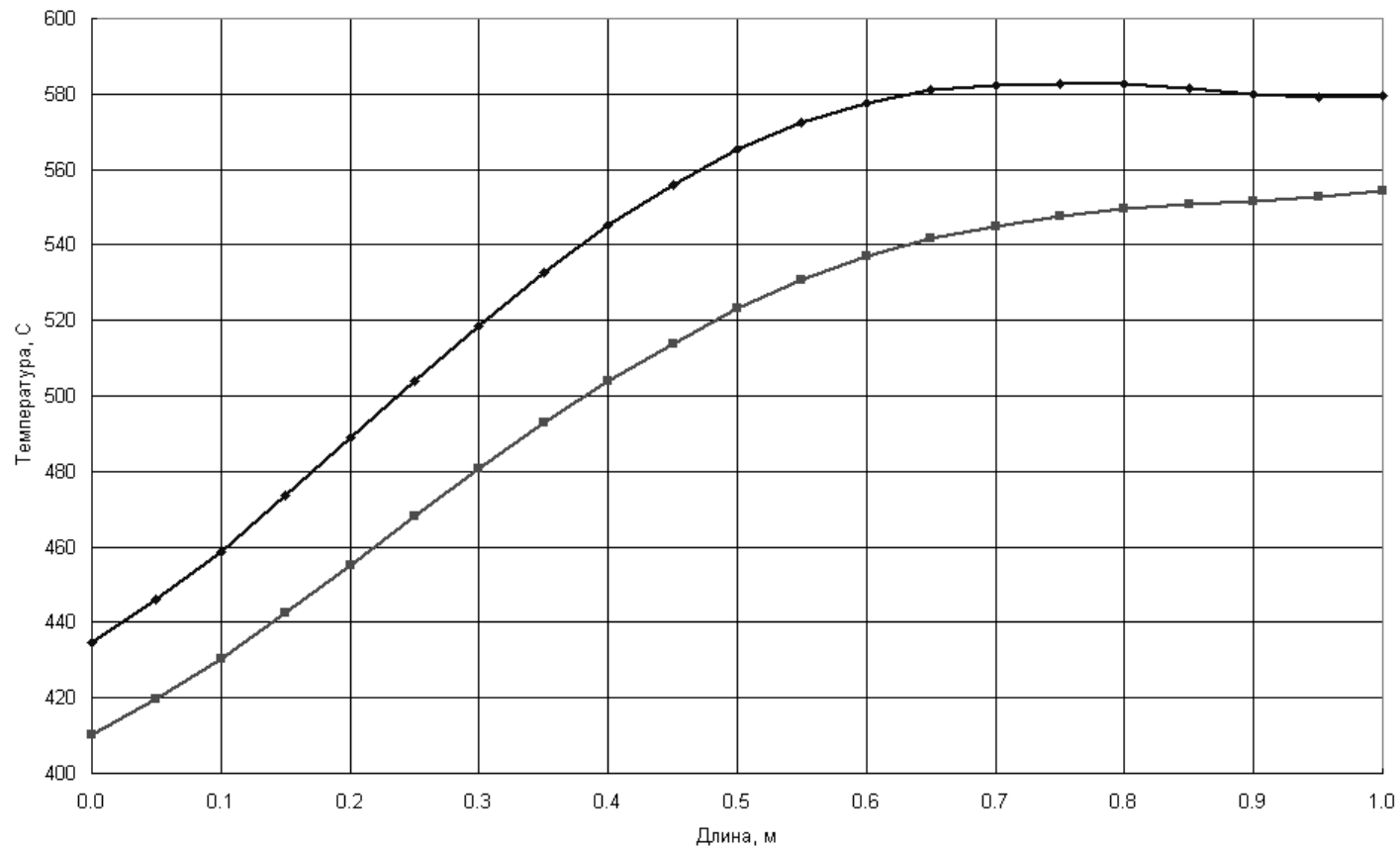
- ⊕ changing flat-to-flat wrapper size at constant pin pitch value equal to 1.75;
- ⊕ changing relative pin pitch at constant internal wrapper size equal to 96 mm.

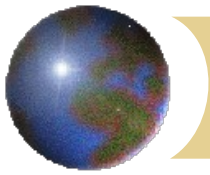
For further researches it was supposed to consider FA design with pin pitch equal to 1.75.

It was shown that max temperature of inner cladding surface does not exceed 590 C. The maximum temperature of outer cladding surface is insignificantly more than 550C (on 5 – 12 C in dependence on pin diameter: 5.7 mm, 6.3mm, 6.7mm).



Temperature distribution along core height (inner and outer cladding surfaces), cladding diameter $\varnothing 5.7$ mm





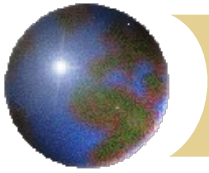
Task 3- Modeling of performance of ADS He-bonded pin with pellet (Pu,Am,Cm,Zr)N fuel

Parametrical researches of most “hot” He-bonded fuel pin of internal zone of ADS with pellet fuel (PuAmCmZr)N:

- ⊕ Influence of two fuel swelling models.
- ⊕ Influence of He release law from fuel matrix for each swelling model: 1) 100 % release, 2) He release by the law of fission gas release.
- ⊕ Influence of two fuel creep laws :

I-st stage - dependence recommended by results of literature review of Task 1

II-nd stage - dependence on PuZrN creep rate received in Task 2 was used.

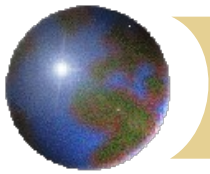


Criteria of fuel performance

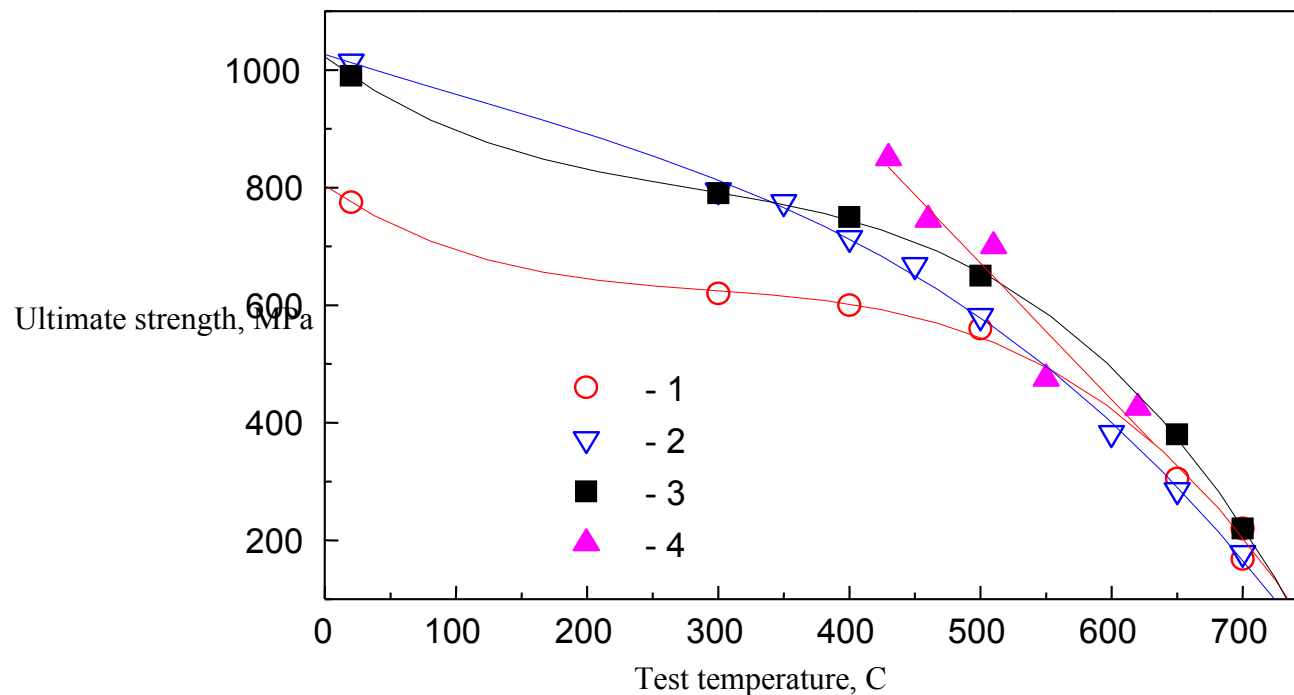
⊕ Maximum permissible fuel temperature
($T=2200\text{K}$, there is a danger of fuel decomposition at higher temperature);

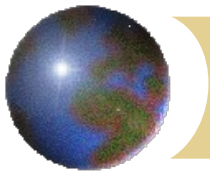
⊕ Maximum permissible cladding stress ; $[\sigma] = \frac{\sigma_{\epsilon}}{K_{\sigma}}$,

⊕ Cladding high temperature strength (CDF –criterion). $\omega_{\sigma}(t) = \int_0^t \frac{dt}{t_r} \leq [\omega]$



***Dependence of ultimate strength of EP-823 steel on tests temperature
1,2 - initial, 3 - irradiated, 50 dpa, 490°C, BN-350,
4 - irradiated, 44-62 dpa, $T_{irr} = T_{test}$, BOR- 60 .***





Task 3- Results of ADS He-bonded pin irradiation behavior modeling.

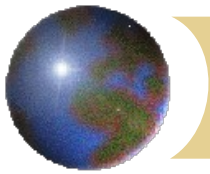
Pin of initial geometry

- ⊕ Hypothesis on He release from fuel matrix essentially influences on stress-strain state of fuel pin.

For all variants with assumption of He release as fission gas, fuel pins **performance is provided neither in cold, nor in hot sections. It is recommended to accept a hypothesis on helium release as fission gas release in fuel swelling model for carrying out estimations with margin.**

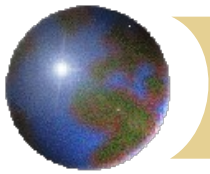
Analysis of PIE data on He and fission gas release from nitrides is done, including BORA-BORA calculation by CARE code. It is shown, that for higher-temperature fuel the higher relative helium release in comparison with fission gas is observed, but essentially lower than 100 %.

- ⊕ **In order to get the results with a margin it is necessary to use SGP model.** SGP model apparently more real when (PuAmCmZr)N fuel is considered as homogeneous mechanical mixture of ZrN matrix and fission materials



Task 3 - Results of ADS He-bonded pin irradiation behavior modeling

- ⊕ For SGP swelling model and hypothesis on helium release as fission gas release the performance of fuel pin of internal zone with pellet (PuAmCmZr)N fuel is provided only up to burn-up of about 19.0at%, that is a little more than half of required value.
- ⊕ Increase of initial gap between fuel and cladding up to 0.2 mm leads to essential decrease in cladding stresses and the required life time is provided according to all criteria. But fuel temperature increases at BOL. Maximum temperature is equal to 1725C that is permissible value according to taken criterion of maximum allowable fuel temperature.



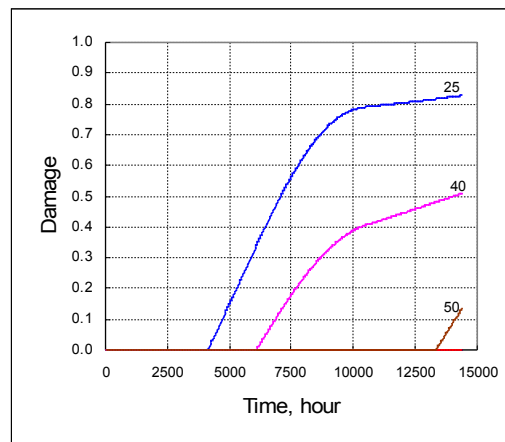
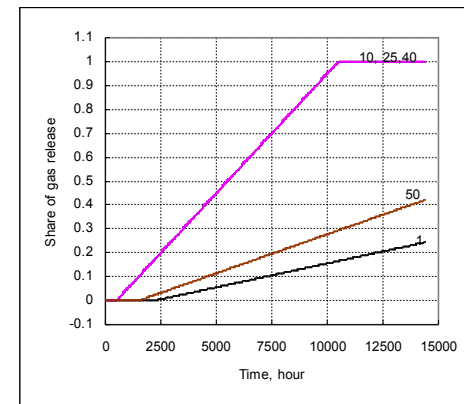
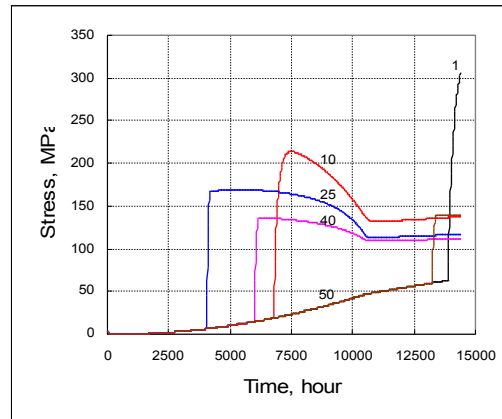
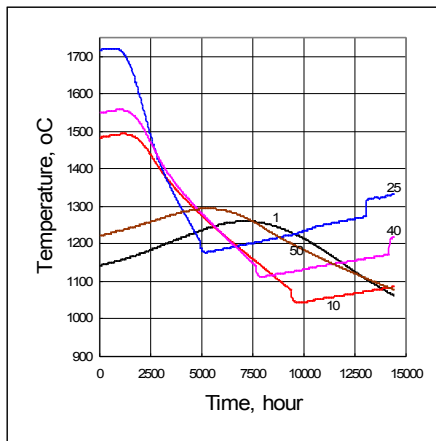
He-bonded pin (gap 0.2mm)

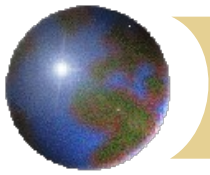
Fig.1. Max fuel temperature in 5 core sections

Fig.2. Clad hoop stresses in 5 sections EOL (33.5at%)

Fig.3. Fission gas release fraction in 5 sections

Fig.4. Clad damage in 3 sections





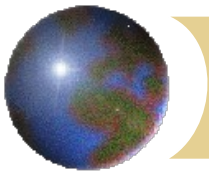
Task 3 -Researches of Na-and Pb-Bi-bonded fuel pin performance

- ⊕ High thermal conductivity of Na and Pb-Bi allows to increase fuel-cladding gap value w/o fuel temperature increase and to avoid cladding stresses from FCMI.

- ⊕ On literature review base the maximum permissible fuel-cladding gap value has been proposed:

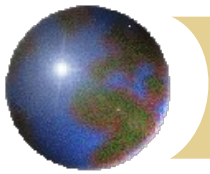
For ADS fuel pin with pellet diameter of 4.9mm this value is equal 0.7mm (cladding geometry 6.3 mm x 0.35 mm).

This pin geometry meets the permissible irradiation conditions according to FACIC code calculations (part.3).



Task 3-Researches of Na-and Pb-Bi-bonded fuel pin performance

- ✦ The performance of liquid metals bonded ADS pins with pellet (PuAmCmZr)N fuel is provided for required irradiation parameters at condition of some modification of pin design, namely, increasing of the initial fuel-cladding gap value.
- ✦ For Na-bonded pin the gap value equal to 0.45mm is sufficient in order to avoid FCMI almost to EOL. Outer cladding diameter - 6.05 mm. For Pb-Bi –bonded pin the gap should be increased to 0,5mm. Outer cladding diameter - 6.1mm.



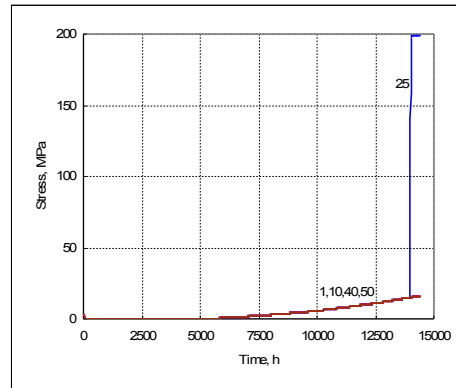
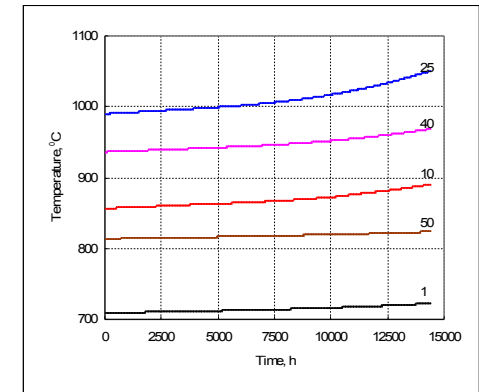
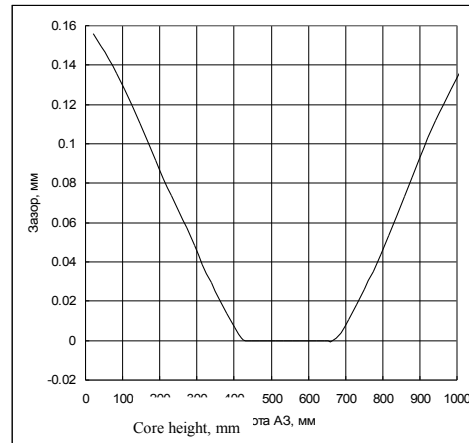
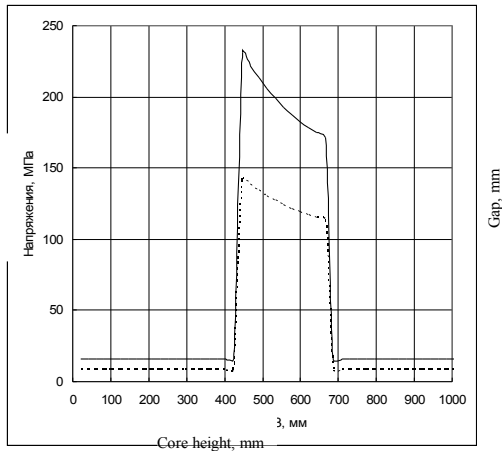
Na-bonded pin (gap 0.45mm)

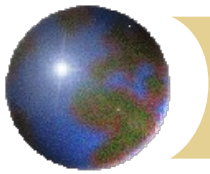
Fig.1. Clad hoop and axial stresses axial distribution at EOL (33.5at%)

Fig.2. Fuel-clad gap axial distribution

Fig.3. Max fuel temperature in 5 sections

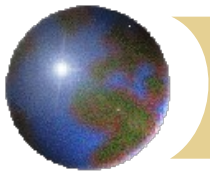
Fig. 4. Clad hoop stresses in 5 sections.





Task 3- Cladding stability under coolant pressure

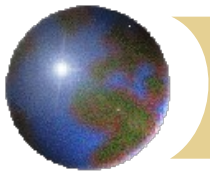
- ⊕ Estimation of ADS cladding stability under the action of outer pressure of Pb-Bi coolant for the case of initial cladding ovality is done by ELLIPS code.
- ⊕ According to thermal-hydraulic calculations results, maximum coolant pressure at bundle inlet is less than 1.2MPa. The results show that for ADS core operation parameters the pin stability is provided even for the case of initial cladding ovality up to 10% at the temperatures of 585-600 C. Only at higher cladding temperature (680C) the stability failure is possible closer to EOL (after 12500 h of irradiation).



Task 3 –RIAR, Dimitrovgrad

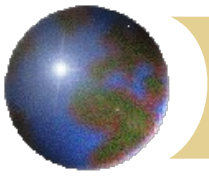
Modeling of pin performance with vibropacked fuel

- ⊕ Calculations have been carried out by VIKOND – 2D code developed in RIAR on basis of KONDOR -2D code. Code VIKOND was developed to calculate and predict performance of vibropacked oxide fuel pins. For calculation of ADS fuel pins with VIPAC (PuAmCmZr)N fuel code some modernization is done.
- ⊕ Input data on fuel geometry, irradiation duration, steel and basic fuel properties are the same as for pellet fuel. The principal difference is twice lower fuel thermal expansion coefficient, constant fuel swelling rate (0.35%/1at%) and gas release (100%).



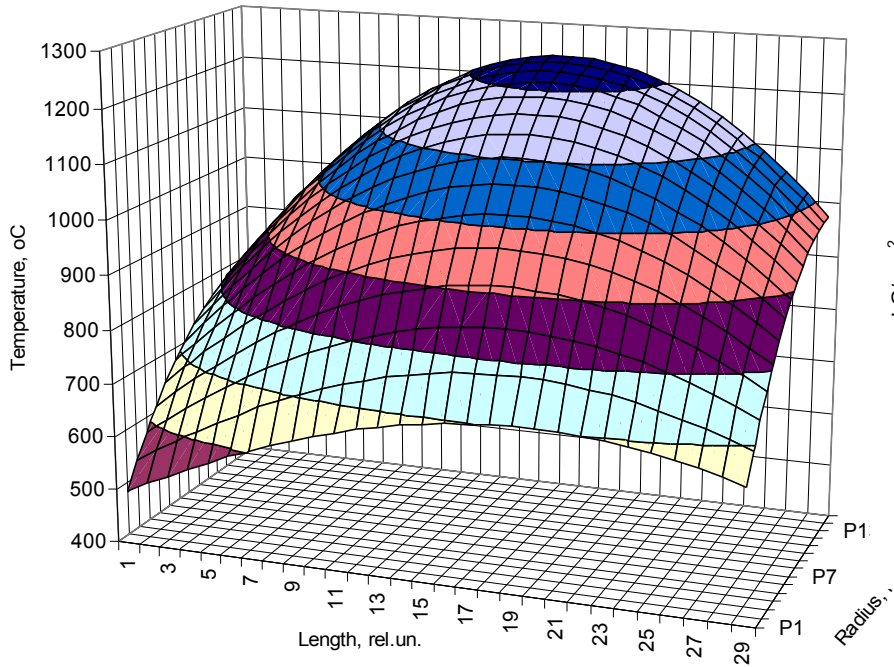
Task 3 – Modeling of pin performance with vibropacked fuel

- ⊕ Calculation results have shown that for taken fuel properties the cladding stresses at EOL are within EP-823 steel strength limit. The calculated time till cladding rupture is much higher than fuel planned lifetime.
- ⊕ Additional investigations are required in order to prove the used data on vibropacked fuel properties.
- ⊕ It is shown, that mass transfer in vibropacked nitride fuel pins at set operating parameters is absent (max fuel temperature less 1300C). The temperature of fuel mass transfer starting exceeds 1500 C.

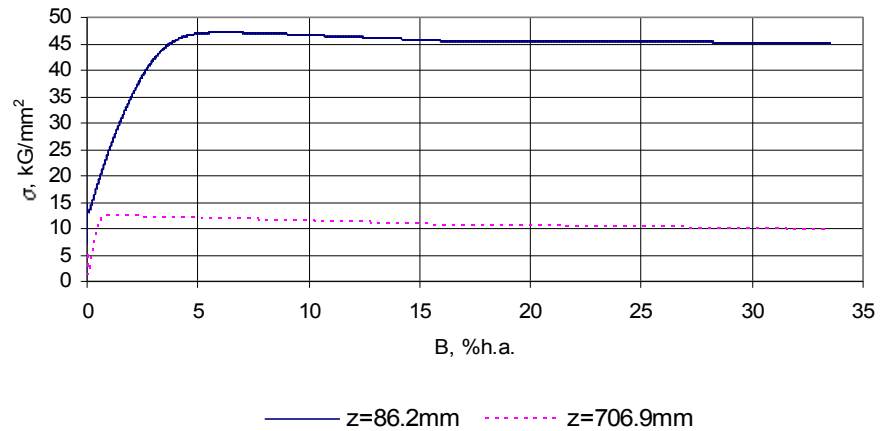


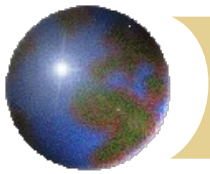
VIKOND code calculation results

Temperature distribution in fuel column



Dependence of cladding hoop stresses in internal zone fuel pin on burnup

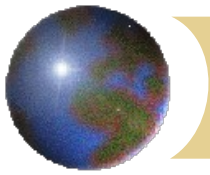




TASK 4. Technical – economical assessments of fabricating feasibility of (PuAmCmZr)N vipac fuel with 60 ± 10 at %ZrN and up to 10 at% Cm

When selecting the procedure of fuel production and equipment provision the following criteria were taken into consideration:

- ⊕ feasibility of fuel fabrication process from technological viewpoint;
- ⊕ technological readiness (availability of pilot equipment and procedures);
- ⊕ cost effectiveness;
- ⊕ technological effectiveness (absence of excess operations during reprocessing, an optimal organization of production cycle).



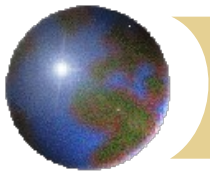
Task 4 - Fabrication technique

The possibility was considered of fabrication on RIAR site of (Pu, Am, Cm, Zr)N vibropacked fuel, (60±10)at% ZrN and 10at% Cm sufficient for manufacturing of 2 BOR-60 fuel pins- ~0.6 kg.

On base of literature analysis and own RIAR experience the electrolytic refining in molten chlorides on liquid metal cathode is proposed as main flow sheet for nitride manufacturing.

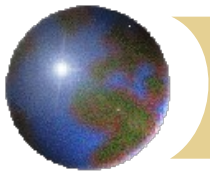
Fuel pins manufacturing comprises the following main stages:

- ⊕ chlorination of the initial components;
- ⊕ production of MA nitrides;
- ⊕ (Pu Am Cm,Zr)N fuel production;
- ⊕ manufacturing of fuel pins.



Task 4- Vibropacked (Pu Am Cm,Zr)N production

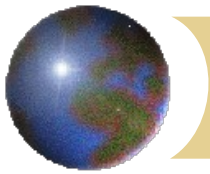
- ❖ Powder of zirconium nitride is pressed and extruded to form press-pellets.
- ❖ Then powder of plutonium, americium and curium nitrides obtained by precipitating MA nitrides by Li_3N is added to 5-th fraction of ZrN granulate.
- ❖ Prepared fuel is transferred to area of fuel pin fabrication. Fuel pins are fabricated under standard conditions with application of existing standard equipment.



Task 4 – Provisions for fabrication cost estimations of vibro PuAmCmZr N fuel pin

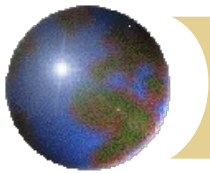
In order to assess fabrication cost of nitride fuel pins for further BOR-60 reactor irradiation the following provisions were agreed:

- ⊕ initial materials : Cm-244, Pu, Am-241 and ZrN,
- ⊕ nitride fuel in powder form is produced in hot cell of bld. 120 by using customized and existing equipment,
- ⊕ pellets from nitride powder are produced at bld.118,
- ⊕ fuel pins are fabricated by vibropacking process at facility located in bld.180,
- ⊕ the components are supplied by Rosatom enterprises.



Task 4 – Fabrication cost estimations

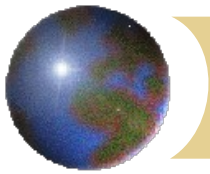
- ⊕ High cost of full-scale pin fabrication (**885 000 Euro**) is completely determined by Cm cost.
- ⊕ Cost of one experimental micro -fuel pin fabrication (curium is included in cost of the experimental fuel pin at full price) – **214 442 Euro**
- ⊕ Cost of one experimental micro-fuel pin fabrication (curium is considered to be made on commission, and the fuel pin cost includes irreparable losses 10% during fabrication process) – **69 722 Euro**



Principal results of MATINE Project

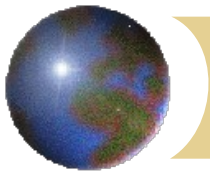
- ⊕ Original data on thermal conductivity, thermal creep, thermal expansion and high temperature stability of $\text{Pu}_{0.4}\text{Zr}_{0.6}\text{N}$
- ⊕ New codes versions (DRAKON, FACIC)
- ⊕ Recommendations on best designs solutions for He-, Na-, Pb-Bi- bonded ADS fuel pin and FA
- ⊕ Demonstration of technical possibility of fabrication at RIAR site of $(\text{PuAmCmZr})\text{N}$ vipac fuel pin with (10at% Cm) and its cost estimation.

9 reports are given to CEA and KTH.



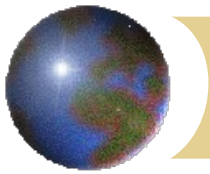
MATINE results presentation

- ⊕ Topical Meeting /2006 ANS Annual Meeting “Nuclear Fuel and Structural Materials for the Next Generation Nuclear Reactors”, June 4-8, 2006, Reno, Nevada, US
- ⊕ 1st annual meeting with KTH, June 23-24, 2005, Stockholm, SWEDEN.
- ⊕ 2nd annual meeting with KTH, December 5-6, 2006, Obninsk, RUSSIA.
- ⊕ 15th, 17th WG1 meetings (CEA-ROSATOM), 2004, 2006, Dimitrovgrad, RUSSIA.
- ⊕ 16th WG1 meeting (CEA-ROSATOM), 2005, Cadarache, FRANCE.



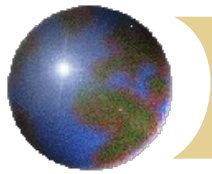
MATINE works prolongation - New ISTC Project Proposal

- ⊕ Results of in-pile performance calculation made in MATINE have shown that liquid metal bonded fuels may perform well up to the target burn-up of 33.5at%. He- bonded fuels however may exhibit problems at high burn-ups in ADS core due to severe FCMI because of fuel swelling under high linear ratings and low fuel creep rate. Since He - bonding is required to remain compatible with aqueous reprocessing options, alternative fuel compositions are of interest.
- ⊕ One of the possible options are the composite materials based on porous matrix made of refractory compounds, like Zr carbide, with heavy nuclides incorporated into the pores. The fuel is developed in VNIINM, Moscow.



Porous matrix fuel

- ⊕ Relative simple fabrication technique, including the absence of dust operations with high radioactive materials, since the last ones are introduced into matrix pores as liquid solutions. Thermal destruction process, mixed oxides formation.
- ⊕ Porous ZrC matrix of required diameter and length is fabricated prior to the filling by actinides. All basic stages of technological route may be robotized.
- ⊕ Formation of mixed oxides even of those nuclides that don't form solid solution in melt (e.g., Np+Am). The possible solid solutions: Pu-Am-Np-Cm, U-Np-Am-Cm, Np-Am-Cm-Th, Pu-U, U-Th etc.
- ⊕ Irradiated fuel reprocessing may be carried out using traditional routes of radiochemical facilities.

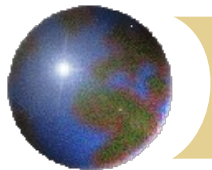


New ISTC Project Proposal #3608

"MATINE -2: Study of MA Transmutation in INert-matrices fuels: modeling, fabrication, and measurements of out-of-pile properties"

The objective of the Project - identification of uranium-free fuel that would perform well under irradiation to high burn-up in ADS core. The possibility to use ZrC porous matrix fuel is proposed for investigation.

- ⊕ The neutronics and thermo-hydraulics calculations will optimize ADS core parameters for effective actinides transmutation. The basic result – substantiation of PuAmCmO₂-ZrC fuel pins performance - **IPPE**
- ⊕ Fabrication and measurement of PuO₂-ZrC thermal conductivity, creep, linear thermal expansion - **VNIINM**
- ⊕ On the base of MATINE-1 estimations vipac PuAmZrN granulate and CmN powder will be fabricated. Thermal stability will be measured - **RIAR**



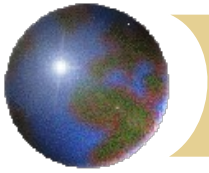
MATINE 2 – Project #3608

Project collaborators

- ✦ Kungliga Tekniska Högskolan, Department of Nuclear and Reactor Physics, SWEDEN
- ✦ The Commissariat à l'Énergie Atomique (CEA), SACLAY, Direction de l'Énergie Nucléaire, FRANCE
- ✦ Nexia Solutions Limited, UK

Supported organizations

- ✦ Serco Assurance, UK
- ✦ JOINT RESEARCH CENTRE Institute for Transuranium Elements, GERMANY



MATINE 2 – Project #3608

- ⊕ The proposal was considered by GB on December 2006.
No final decision about funding.
- ⊕ Additional consideration –March 2007.