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NANOSTRUCTURAL MATERIALS IN THE NUCLEAR ENGINEERING

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A brief review of research findings, pilot projects and application of nanomaterials in the nuclear power industry and engineering is presented. Examined are the prospects of creation of nanostructural materials and coatings for building blocks in NPPs and future fission reactors with a view to improving mechanical properties, increasing radiation resistance. Also, investigated are the main problems arising from development of methods for nuclear fuel modification, development of dispersion-hardened steels.
KEY WORDS: nanomaterials, nanotechnologies, nuclear fuel cycle, radiation resistance and corrosion resistance

НАНОСТРУКТУРНЫЕ МАТЕРИАЛЫ В ЯДЕРНОЙ ЭНЕРГЕТИКЕ

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Представлен краткий обзор результатов исследований наноматериалов для ядерной энергетики. Рассмотрены перспективы применения наноматериалов в ядерной энергетике, связанные с созданием наноструктурных материалов и покрытий конструкционных элементов АЭС и будущих ТЯР для увеличения показателей твердости и прочности, повышения радиационной стойкости. Также рассмотрены основные проблемы, возникающие при разработке методов модифицирования ядерного топлива, разработке дисперсно-упрочненных сталей.
КЛЮЧЕВЫЕ СЛОВА: наноматериалы, нанотехнология, ядерный топливный цикл, радиационная и коррозионная стойкость

НАНОСТРУКТУРНІ МАТЕРІАЛИ В ЯДЕРНІЙ ЕНЕРГЕТИЦІ

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Представлено короткий огляд результатів досліджень наноматеріалів для ядерний енергетиці. Розглянуто перспективи використовування наноматеріалів в ядерний енергетиці, які пов'язано з створенням наноструктурних матеріалів і покриттів конструкційних елементів АЕС і наступних ТЯР для збільшення показників твердості та міцності, підвищення та радіаційної стійкості. Також розглянуто основні проблеми які виникають при розробці методів модифікування ядерного палива, розробці дисперсно-зміцнених оксидами сталей.
КЛЮЧОВІ СЛОВА: наноматеріали, нанотехнології, ядерний паливний цикл, радіаційна і корозійна стійкість

Nuclear power is a major source of electric and heat energy in Ukraine. In Ukraine, many nuclear fuel cycle (NFC) elements are already available, and for their further successful operation and development, it is necessary to carry out intensive fundamental and applied research [1, 2]. In nuclear power engineering, the issue of upgrading fuel and constructional materials for the reactor core is of critical importance. The fuel materials include a wide range of uranium and transuranium elements and their compounds. The nuclear reactor constructional materials include austenic, ferritic, ferritic-martensitic and other steels, graphite and carbon materials, zirconium alloys and various ceramic materials.

Therefore, one of the tasks is to develop basic and applied research in the field of radiation materials science and radiation technologies, creation of new fuel and constructional/engineering/structural materials, as well as new methods for analysis and control of materials. Over the last decades nanotechnologies have been used practically in all areas of advanced technologies, and in fact have turned into an interdisciplinary field of science and engineering. In the nuclear sector, nanotechnologies had been used even before the word 'nano' became popular with researchers because newly created heat and structural materials were largely based on qualitative changes in materials properties which occurred when going down to the nanometric size range [3-5].

The purpose of this paper is to present a summary of outcomes of the last research and developments in the area of application of construction and functional nanostructural materials in the nuclear power industry and engineering.

STATUS OF NANOMATERIALS IN THE NUCLEAR POWER INDUSTRY

Nanomaterials in nuclear power engineering

Among the top priorities in R&D support for creation of NFC constructional materials one can note research works aimed to ensure design and beyond-design (30-60 years) operational life of nuclear units, as well as development of improved constructional/engineering/structural materials for thermal neutron reactors, increase of the depth of nuclear fuel burnup. The areas of application of nanotechnologies in the nuclear power industry are rather diverse and cover practically the whole range of problems relating to the nuclear fuel cycle and created fusion fuel cycle [6-33]:

- Creation of a new high-density nuclear fuel with nano-sized additives, fuel compositions for fuel assemblies of the NPP reactor core (active zone of nuclear reactors). Creation of a new class of mixed uranium-plutonium mixed oxide (MOX) fuel. Development of the torium-uranium cycle [7].
- Creation of nano-disperse construction and functional materials (zirconium alloys for nuclear fuel elements, oxide dispersed strengthened (ODS) ferritic-martensitic steels or nano-dispersed ODS steels.
- Research and development of materials for quick neutron reactors and future Generation IV reactors. Research of irradiation-induced microstructure. Microstructure grounds for a possibility to extend operational life of reactor elements: vessel excluding swelling.
- Development and production of fast-quenched solder alloys for high-temperature flux-free soldering of zirconium alloys, corrosion-resistant steels, transition joints of steel-zirconium and steel-titanium, and other dissimilar materials providing a preset level of radiation and corrosion resistance of soldered joints.
- Creation of nano-membranes and nano-filters for spent nuclear fuel (SNF) and radioactive waste (RAW) conversion technologies, ceramic materials for afterburning of radiolytic hydrogen.
- Development of metrological assurance for application of constructional and functional units based on nanomaterials for nuclear facilities.
- Multi-scale simulation of nanostructures, materials and processes.
- Nanogages and nanosensors for NPP management and control systems.
- Research and development of materials for future nuclear fusion reactors. Nanostructural materials for the ITER blanket and first wall. Nanostructural low- and high-temperature superconductors for ITER magnets.

In the process of solution of the above-listed tasks, experimental-industrial technologies are developed to obtain functional materials and products with application of nanotechnologies and nanomaterials for nuclear, thermonuclear, hydrogen and conventional fuel power production, as well as pharmaceutical drugs. Let us look at some of the results obtained in the course of research and development of nanostructural materials in the nuclear power industry.

Nuclear fuel with nanometric additives

The Ukrainian energy strategy stipulates gradual introduction of new MOX fuel nuclear power technologies with nuclear fuel cycle closure. Further development of the nuclear power industry requires inclusion of fast neutron reactors into the NPP capacity structure. Out of all types of such reactors only sodium loop reactors (BN) are commercially manufactured. The BN-600 reactor, which has been working at the Beloyarsk NPP (Russia) since 1980, may serve as an example of such reactors. In 2012, there is a plan to launch the 4-th power generating unit of the Beloyarsk NPP with a BN-800 reactor, set up MOX fuel production and implement a closed fuel cycle based on this reactor. One of the pre-requisites for higher NPP efficiency is an increase of the depth of nuclear fuel burnup. To ensure a deep fuel burnup, there is a need to create nuclear fuel macrocrystalline structures with controlled porosity. Nuclear fuel macrocrystalline structures confine gaseous and volatile fission products, prevent transportation of fission fragments on grain boundaries towards the fuel element jacket, which results in considerably lower level of damageability of the fuel element jacket. To obtain a macrocrystalline state, when the fuel ceramics is being pressed from UO_2, (U, Pu)O_2, (U, Pu)N) it is being added ultradispersed powder UO_2 with nano-crystals of a size of ~40 nm. As a result, the fuel ceramics sintering temperature goes down to ~ 200 degrees, ceramics density increases, and the grain size increases up to 35÷40 µm without degradation of other characteristics. Activation of the sintering process due to nano-additives may become one of the trends in creation of technologies for new types of uranium-plutonium oxides and nitrides for the nuclear fuel of the fast reactors nuclear power industry (Fig. 1).

These trends are as follows:

- modification of nuclear fuel;
- creation of complex carbonitride doped with nanodiamond UZrCN;
- fine tuning and modernization of the pilot plant project for production of MOX fuel for BN-800;
- development of a model of solid structures formation on the solid-liquid boundary (solid-melt interface) of reactor materials;
- investigation of interaction of prototypes of melted materials in the reactor core with the reactor vessel;
- study of the mechanism of explosion of liquid metal drops;
- large-scale simulation of reactor core (RC) materials.

Paradoxically, the process of application of ultradisperse additives in fuel ceramics manufacturing lies in utilization of nano-additives to produce a structure which is close to monocrystalline (syngle-crystalline).

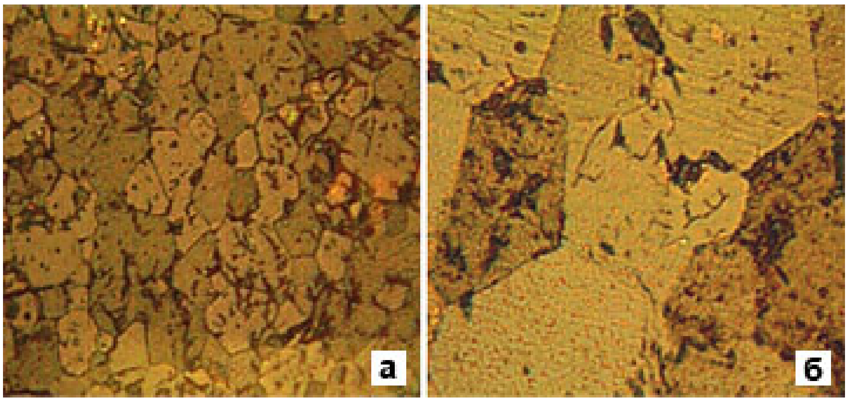


Fig. 1. Microstructure of nuclear fuel.
a - standard microstructure, b - microstructure obtained with application of nano-additives

Oxide dispersion strengthened ferritic-martensitic steels. Nano-dispersed ODS steels

Application of nanomaterials and nanostructures becomes more wide and efficient, particularly in development of ODS steels. ODS steels are various chromiferous steels of the ferritic-martensitic class, strengthened with nano-sized oxide particles. The chromium content in steels is in the range of 9÷18 wt. %, while content of other alloying elements (Al, W, Mo, Nb, Ti, Zr etc.) may be at a level of a few percents and less. The main strengthening oxide particle with a size of <10 nm is Y_2O_3 (down at 0.5 wt. %). Along with yttrium oxides, also used are oxides of aluminum, titanium and other metals. Increase in performance efficiency and length of operational life of prospective fast neutrons reactors requires first of all increase in burnup fraction up to 18-20% without degradation in performance of heat-carrying medium. Solution of these problems is inseparable from development of radiation resistant constructional/engineering/structural materials. These materials are to be exploited in fast neutron reactor cores (E > 0.1 MeV) in fluencies up to $2{\cdot}10^{16}$ n/(sm^{2}s) with damaging doses 160…180 dpa at temperatures 370...710 °C. Radiation resistance of constructional materials in fast neutrons reactors is defined by swelling, creep, high- and low-temperature embrittlement, as well as stability of the material structure properties in the neutron irradiation field. Similar problems arise in creation of radiation resistant constructional/engineering/structural materials for the first wall and blanket of the ITER (International Thermonuclear Experimental Reactor). Solution of this problem requires creation of a new class of radiation resistant metallic materials strengthened with nanoparticles of metal oxides. These materials should have the following properties [14-19]:

- low creep at temperatures up to 970 K and dimensional stability, long operational life ~9 years;
- high radiation resistance to neutron irradiation with irradiation doses ~250 dpa;
- combination of radiation resistance and increased heat resistance;
- high mechanical properties: high rupture resistance of >300 MPa at 970 K, creep rupture strength of >120 Mpa at 10^4 hrs at 970 K, percentage of elongation of >1%;
- high corrosion resistance compared to heat-carrying medium under an elevated temperature and chemical compatibility with the fuel;
- high chemical compatibility when in contact with the fuel and sodium flow.

One of the ways to solve this problem is to create and use a new class of ferritic-martensitic radiation resistant steels strengthened with nano-sized oxide particles (ODS steels).

One should note that this task is an element of the global development of constructional/engineering/structural materials strengthened with high-disperse non-metallic particles (nanoparticles) and designed for reactor core elements in prospective nuclear reactors. This line of research is being intensively developed in countries which have developed nuclear power ITER structures (Russia, USA, Japan, China, France, Ukraine) [20-29]. Radiation swelling is the main criterion for selection of prospective nuclear reactor constructional/engineering/structural materials. Dose dependencies of swelling for materials of austenite and ferritic-martensitic classes are considerably different. Swelling and creep strains of EP-450 cladding from high burn-up fuel assemblies in BOR-60 fre presented on Fig. 2. Currently available data to ~163 dpa imply that the void swelling rate of EP-450 is very low. It is reasonable to assume that the swelling rate will always be low, especially at dose levels not yet reached in reactors. Ion bombardment can be used to explore swelling at very high dpa levels. A dpa rate of 10^{-2} dpa/sec is available on accelerator.

At present there is a great variety of ODS steel grades and types grouped according to their producer countries:
Belgium: DT2906 (Fe - 13Cr – 1.5Mo – 2.9Ti – 0.6 O_2); DT2203Y05 (Fe – 13Cr – 1.5Mo – 2.2Ti – 0.3O – 0.5 Y_2O_3
USA: MA957 (Fe – 14Cr – 0.9Ti – 0.3Mo – 0.25Y2O3); 12CrY1 (Fe – 12.4Cr – 0.25Y_2O_3); 12CrYWTi (Fe – 12.3Cr – 3W – 0.39Ti – 0.25 Y_2O_3)
Europe: ODS EUROFER (Fe – 9Cr – 1.1W – 0.2V – 0.14Ta – 0.3 (0.5) Y_2O_3)

China: K7 (Fe – 13Cr – 1.1Ti – 0.2Mo – 2W – 0.39Y_2O_3)
Japan: Fe – 9Cr – 0.12C – 2W – 0.20Ti – 0.35Y_2O_3
Fe – 12Cr – 0.3C – 2W – 0.3Ti – 0.23Y_2O_3

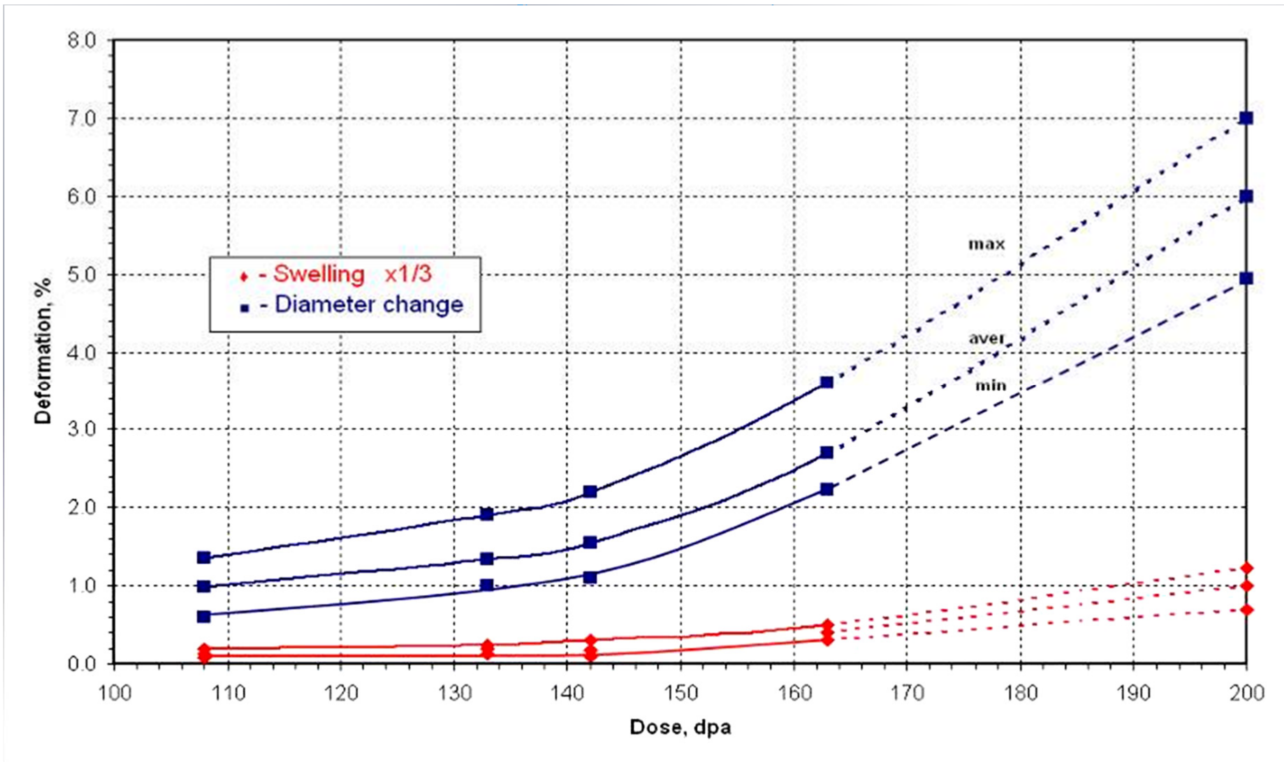


Fig. 2. Dose dependencies of for different swelling. Swelling and creep strains of EP-450 cladding from high burn-up fuel assemblies in BOR-60. Povstyanko et al., 2011 [33]

On this background, stand out chrome-rich (up to 22%) steels with Al additives (up to 4.5 %). 9Cr-ODS steels developed for sodium-cooled fast neutron reactor shells turned out to be unpromising for super critical water cooled fast reactors (SCWFR) and liquid bithmuth energetic fast reactors (LBEFR) due to high corrosion levels.

Table.

OD Steels of Russia			
Steel	Y_2O_3 Content (wt. %)	Ti Content (wt. %)	$MgAl_2O_4$ Content (wt. %)
EP450	0.5	-	-
EP450	0.25	0.3	-
EP450	-	-	1
F1 (18%Cr)	0.25	0.3	-
EK181	1	1	-
EK181	0.25	0.3	-
C+S68	0.25	0.3	-
AISI 316 (waterdispersed)	0.25	0.3	-

The structure of ODS steel is characterized by precipitation of oxide phase particles of various sizes, composition and distribution density against the matrix background (Fig. 4).

Presence of nano-oxide particles substantially improves mechanical properties, e.g. significantly changes thermal creep parameters (Fig. 4). The main results of earlier studies of oxide dispersion strengthened ferritic steel [27-29] and developed powder ferritic steels (based on Cr13 steel for the whole class of ferritic stainless steels) [30] after strengthening them with high-dispersion oxides drive us to the following conclusion: under all irradiation conditions (in a heavy-iron accelerator, in VVR-M and BN-600 reactors) embrittlement of material is not observed. In fluencies of up to $2.6 \cdot 10^{23}$ n/sm^2 swelling of material makes 0.25%, residual plasticity is 3…4%.

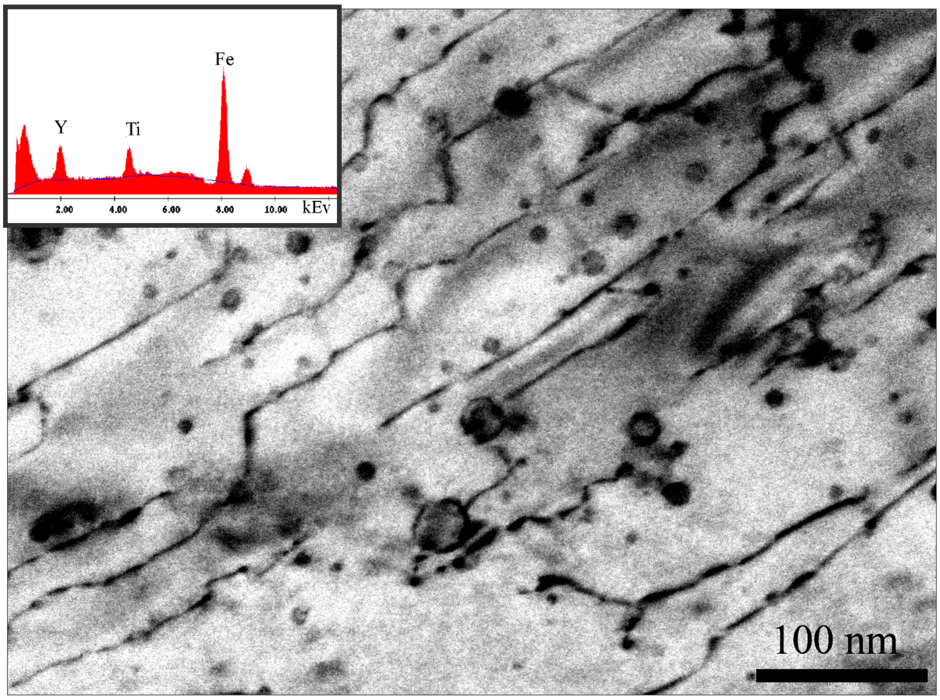


Fig. 3. Nano-oxides in steel EP450 ODS

Corrosion resistance of dispersion strengthened steels in liquid lithium, lithium-lead eutectics, cesium and tellurium vapors is equal or somewhat higher than it is for the best industrial reactor steels. The steel has a sufficient heat resistance at 970 K, high radiation and corrosion resistance in liquid metal environments and fission products. In spite of the above named facts, powder technology does not ensure sufficient homogeneity of products. Over the last years, Russian researchers have developed a number of ODS ferritic-martensitic steels for fast neutron nuclear reactors, including those based on steel EP450 (Fe-13Cr-2Mo-Nb-V-B-O, 12C) which is used as operational material of BN-600 reactor fuel assembly [14, 33].

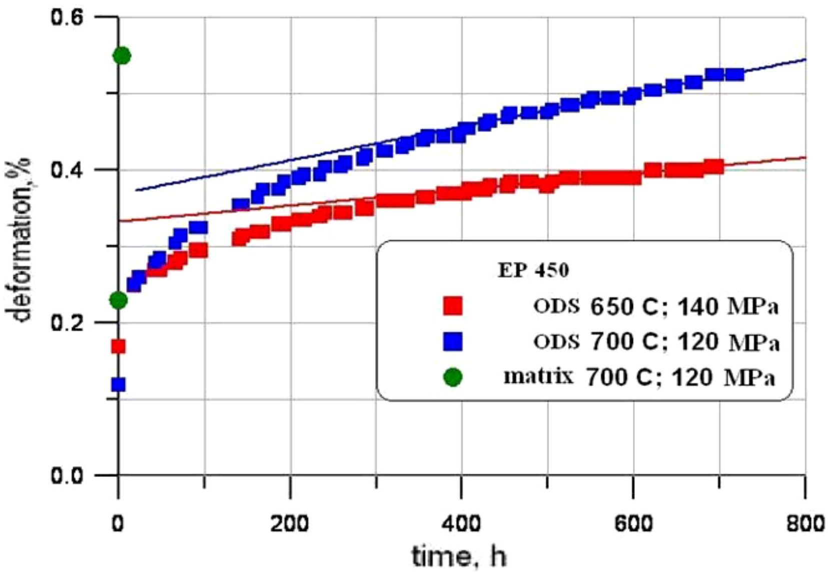


Fig. 4. Thermal creep of EP450 (matrix) and EP450 ODS steels.

Fig. 5 shows that in transition from scale 50 nm to scale 5 nm the particles structure displays an order with a hexagonal or octaedric symmetry (Fig. 6).

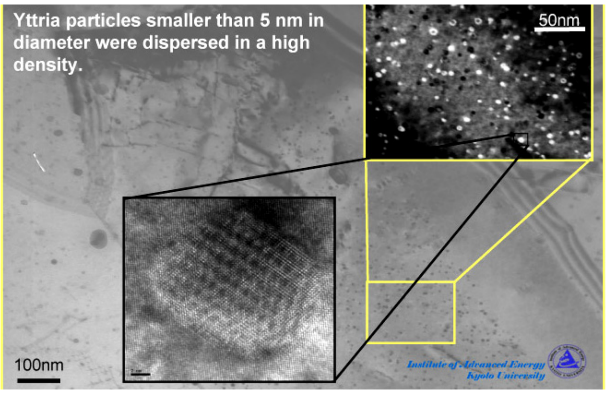


Fig. 5. Nanoscale oxide dispersion: Y_2O_3 particles have the size of less than 5 nm in diameter and are disperse distributed with high density (bottom left corner). (Data according to Kyoto University)

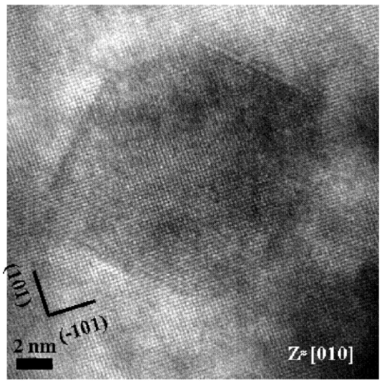
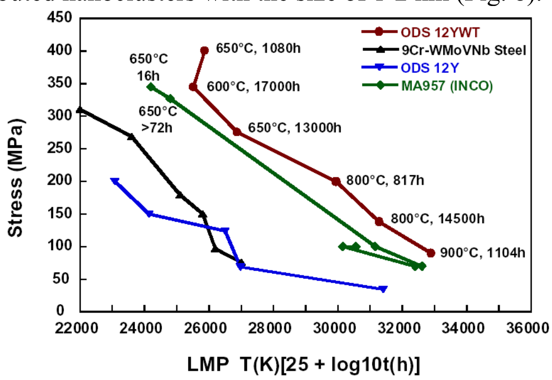


Fig. 6. Structure of precipitates in SOC steels

The best creep resistance characteristics (Fig. 7) are obtained in formation of the maximum amount of evenly distributed nanoclusters with the size of 1-2 nm (Fig. 8).



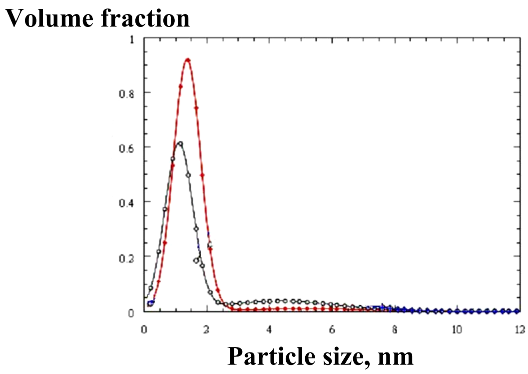
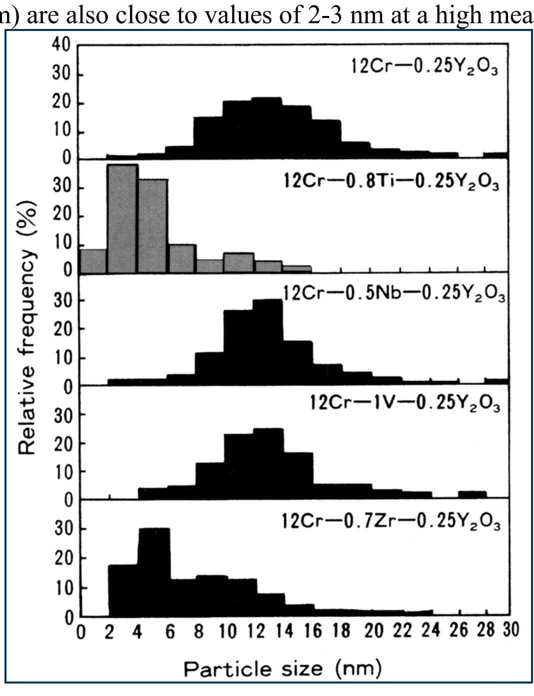


Fig. 7. Creep resistance characteristics of ODS steels

Fig. 8. Distribution of nanoparticles in the strongest steels (Fig. 5).

In the process of additional alloying with zirconium, dispersion of oxide yttrium particles shifts towards smaller sizes, close to optimal (Fig. 9). High density of particles enriched in Ti, Y and O (Fig. 10) is observed. The mean size and concentration of oxide particles in initial and annealed samples of 12YW and 12YWT ODS steels (alloyed with titanium) are also close to values of 2-3 nm at a high mean concentration.



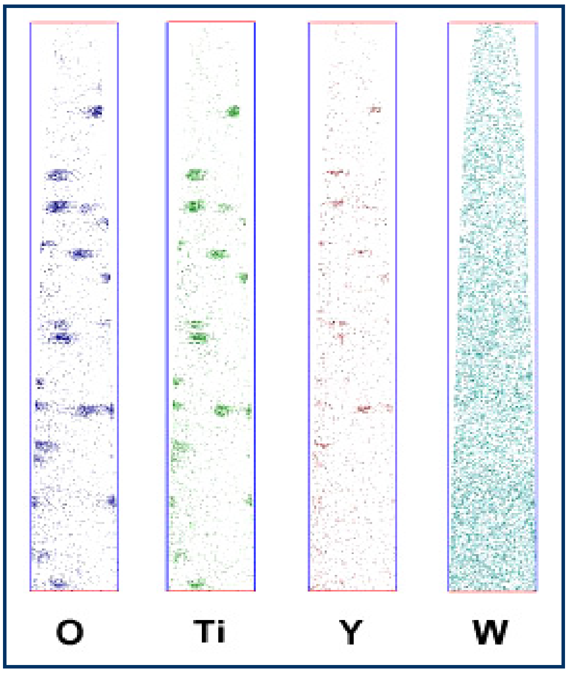
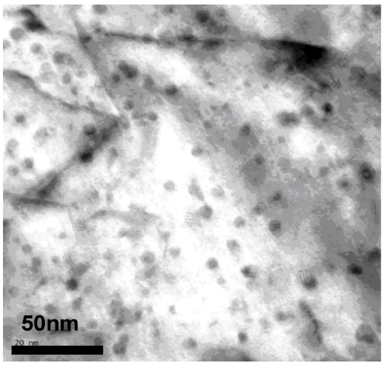


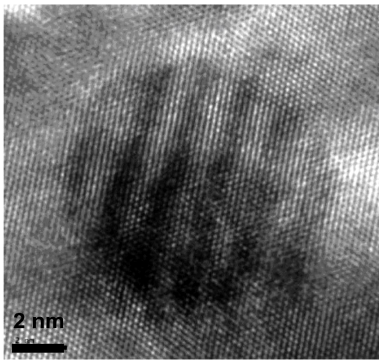
Fig. 9. Size distribution of Y_2O_3 particles in ODS ferritic steel alloyed with Ti, Nb, V and Zr.

Fig. 10. Position of atoms in the sample of 12YWT steel annealed at 1300°C.

Besides, MA K3-ODS steel contains mostly $Y_4Al_2O_9$ (YAM) particles with a monocline structure (Fig. 11a). Fig. 11b shows another type of ordered oxide structure at a scale of 2 nm.



a



b

Fig. 11. Disperse oxide particles in SOC-1 steel without irradiation Z=[111]
a - general view of the microstructure, b - nanostructure of ordered oxide particle

ODS STEELS: IRRADIATION

Radiation swelling of ODS steels of different compositions was intensely investigated in neutron irradiation, as well as in one-beam, two-beam (metal, gas) and three-beam irradiation. When analyzing irradiation results, various models of oxide nanoparticle formation mechanism are used.

Formation of nanoparticles is done in three stages:

1) Fragmentation of moving Y_2O_3 particles at the early stages of globe mill processing.
2) Clasterization and solid-state amorphization of Y_2O_3 fragments mixed with matrix components at later stages of globe mill processing.
3) Clystallization of amorphous agglomerates of a size less than 2 nm to form oxide nanoparticles with a nuclear shell structure at the steel consolidation stage.

Nanoclasters of a size of less than 2 nm remain amorphous (disordered) or have a crystal structure similar to the matrix. These nanoclusters play a critical role in ODS steel radiation resistance.

The irradiation results given below (Fig. 12) show that a substantial change in the steel structure takes place with doses of 65 dpa and at T=700 C.

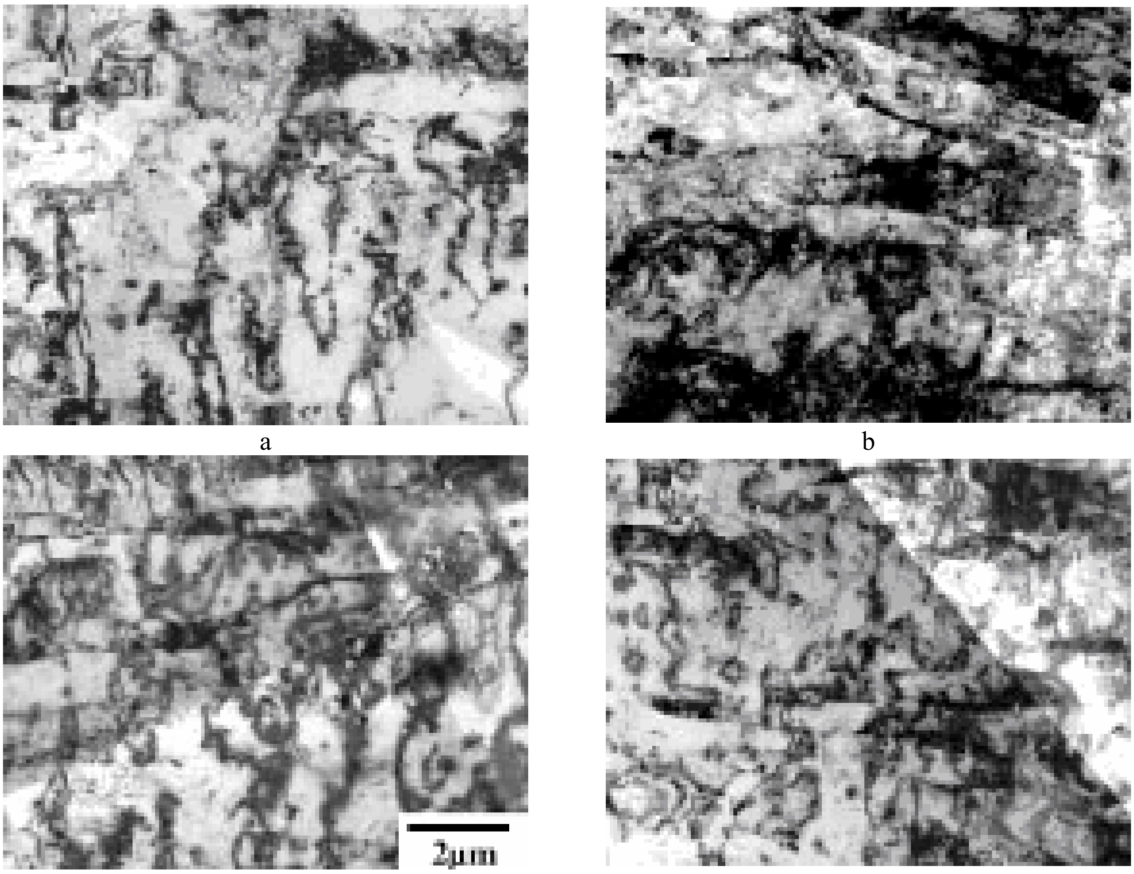
On the other hand, an important element which characterizes irradiated ODS steels is formation of voids in nanoclusters. Electron microscope investigations allowed to study the crystal structure and density of oxide particles in the ODS ferritic steel, investigate the density and distribution of radiation-induced voids in original steel Fe-14Cr (without oxide strengthening) and in ODS steel Fe-16Cr-4Al-2W-0.3Ti-0.3Y_2O_3. Partial inputs of the crystal matrix and oxide nanoparticles nucleus/shell structure have been defined.

Preliminary results have shown that the swelling process slows down due to increase of oxide particles density in ODS steels. Complex oxides ensure stability of the structure after ion irradiation of up to 60 dpa at 650 °C. Given below is an example of an irradiated area in SOCP-1 steel, 300 °C, 60 dpa. An evident contrast of damage caused by ion irradiation as well as dense dislocations in irradiated and non-irradiated areas are observed. Synergy effect of He and H was clearly demonstrated in triple ion beam (Fe3+ He+ H+) irradiation. Average swelling in F82H steel was considerably accelerated under the influence of triple ion beam irradiation. To create more sophisticated irradiated conditions, researchers of the NSC KIPT (Kharkiv, Ukraine) apply irradiation in a triple ion beam mode on the basis of the ESUVI accelerator with Cr, Cr+He, Cr+H, Cr+He+H ions.

The study of irradiated K3-ODS steels shows that: there are voids formed in nanoclusters; a homogeneous distribution of voids filled with helium in the matrix of K3-ODS steel similar to high-density disperse nanoclusters is observed; nanoclusters enhance heterogeneous bubble nucleation near the clusters which results in retardation of transformation of bubbles into pores.

Analysis of chemical properties of nanoclusters in ODS steels is an important issue. Experiments on homogeneous saturation of ODS materials with He atoms have been conducted at different temperatures on the RRC KI cyclotron. Fast helium ions stop in targets forming profiles of radiation-induced damage, degradation and development of which result in homogeneous saturation of ODS materials with helium. Mechanical tests of irradiated and non-irradiated ODS materials have shown presence of helium embrittlement in irradiated ODS steels at T=450°C. Helium bubbles are observed in irradiated ODS materials at C_{He} = 500 appm at 300°C and 450°C.

The structure and composition of oxide/matrix boundaries which depend on the size of $Y_4Al_2O_9$ particle for Super ODS steels are shown. Incoherent particles beecome larger much quicker due to the higher boundary energy leading to changes in the strength of flows which take up pin holes and helium bubbles. A large precipitate (of size > 15 nm) is characterized by an incoherent boundary with the matrix associated with/ the spherical -shaped shell. In The small precipitates (of size < 10 nm) with a semi-coherent boundary associated with/ the facets.



a b c d

Fig. 12. Structure of MA957 (Fe -14Cr – 1 Ti - 0.3 Mo – 0.25 % Y_2O_3) after irradiation in reactor JOYO
a - without irradiation, b - 65 dpa, 500°C; c - 101 dpa 710°C, d - 101 dpa, 500°C

CONCLUSIONS

One can see from the quoted results that nano-structural materials take on an important role in the nuclear power industry as construction and functional materials which are being used in practically all stages of the nuclear fuel cycle. Extremely important is formation in irradiated nano-structural materials of an ordered nanostructure composed of new phases with a period of a few nanometers. This structure facilitates retention of materials properties in high-dose irradiation. The discovered phenomenon may initiate development of a new trend in the radiation materials science – creation of structural materials "positively" responding to radiation exposure. The prospect of application of nanotechnologies in the nuclear power industry is related to the opportunity of creation of structural and functional elements of nuclear and thermonuclear facilities with the required complex of mechanical, corrosion and radiation resistance properties. Impressive characteristics of ODS steels make it possible to plan application of this kind of materials in the thermonuclear power industry as materials for first wall and blanket. Transition to nanostructural materials will make it possible to create materials with new, game-changing properties for the nuclear power industry and set up create new departures in development of power generating equipment. This is why the crucial task is to accelerate development of projects in the field of nanotechnologies and nanomaterials to ensure economic stability and innovative industrial transformations.

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