THE APPLICATIONS OF MICRO-NANO-CER-LIQ MATERIALS IN NUCLEAR POWER

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Abstract

Modern nuclear fuels exploit the unique features of nano-structures. With proper engineering of fuel micro-nano-structure, fission products can, in principle, be separated from the fuel in-situ, and further be separated by the transmutation products. Fuel entities which are about 10 to 30 microns in diameter can contain the fuel ions in the particles while allowing the fission fragments to escape into the surrounding matrix, by virtue of the difference in travel distance. The matrix is chemically engineered to form stable phases with the most common fission products so that these products do not diffuse back into the fuel particles. This nano-structure engineering of fuels can be done with virtually any existing fuel, including oxides or metals for both thermal and fast reactors. A variant of this idea is to use a flowing liquid that may be continually cleaned, in order to remove the fission products, leaving a fuel which is clean and long-lived, or which may be sealed inside the pellet cladding improving the fuel performances and its reprocessing technology. The applications are in Light Water reactors, Fast breeder Reactors and Traveling singular wave reactor, making possible the increase in their burnup, life-time, performances.

Keywords: Microstructure, nanostructure, nuclear fuel, fission products, separation

1. Introduction

The present work is a part from a larger work of multi-scale/ multi-dimension / multi-phenomena and multi-application development started by 1980 in Romania, inside special applications research having as initiators Prof. Dr. Ioan Purica [1] and Prof. Ioan Ursu [2], Dr. Geavidt Musa [3] and members of the Romanian Military Research [4].

In Figure 1 is briefly shown a spiral of evolution [5] from the femto-level of the nuclear reaction to the pico-level of the atomic structure, nano-level of the molecular and grain structure to micro & mezo-level of the functional material up to application macro level.

In the center in the red rectangle there is an ideographic presentation of a generic neutron - induced nuclear reaction having as opened reaction channels the scattering, absorption and transmutation with decay or fission. Nuclear dimensions are in tenth of fm (femto-meters) while the time scale of the primary interactions is in pico-seconds and lower.

The kinematics of this reaction is immediately transmitted at the atomic and molecular level where there are about 3-5 atoms per nm, and about 1-2 molecules per/nm.

At this level starts the buildup of the nano-engineered functional materials.

The page is divided in 4 and in center it has the basics of the development – the nuclear reaction in interaction with matter at atomic, molecular, cluster level, to mezzo- and macro-scale driving to engineering applications [6].

The ideograms surrounding the red rectangle show elementary cell of the nano-engineered functional material, which are specific nuclear materials that are further driving to applicative mezzo-scale structures near the blue rectangle, and further to new nuclear structures towards the border of the outer pink rectangle.
These structures are leading to novel nuclear reactor generations “G” followed by a Latin number; this is just giving an idea relative to the potential of novel nuclear reactor generations as a development from the actual Generation 4 [7], while the numbers followed by a “Y” represents the years until the first prototype might be delivered if the research is launched and funded properly [8].

The left-upper corner represents the spring off micro-hetero-structures for Generation 5 that is mainly characterized by a fuel that self-separates from fission products, allowing easy fuel reprocessing and near-perfect burnup reducing the need for enrichment.

The right-upper corner shows applications of the engineered nano-clustered enhanced transmutation for Generation 6 and the schematics of a “Candle” type reactor [9, 10] with high burnup using $^{239}$Pu or $^{233}$U as a criticality enhancer [11], with easy separation of the transmutation breeding products, no matter if they are actinides or other radioisotopes.

In the left-lower corner is generically presented a super-capacitor structure that charges from the energy of nuclear particles and discharges as electricity, also referred as a direct nuclear energy conversion nano-hetero structures, that drives to Generation 7 of nuclear reactors that occupies a tenths of the space of the actual nuclear reactor for ten times higher power, missing the thermo-mecano-electric infrastructure.

In the right-down corner is presented the radiation nano-guiding NEMS that are driving to electronic control of nuclear reactors and ultra-light shielding, making possible very compact, mobile nuclear power sources, called Generation 8.

From this ideographic presentation are missing the micro-nano-fractal engineered materials which can self repair and recover from radiation damage and are mainly used as structural materials, with properties invariable with radiation dose, used in all these generations.
Fission products

Similar to classical energy production inside the thermo-electric power plant where the energy comes from oxidizing carbon present in various fuel, in thermo nuclear power plants the energy comes from decomposing actinides in medium elements, called fission products (FPs) and releasing a part of the nuclear binding energy as kinetic energy of the fission products that is transferred to the fuel lattice finally becoming heat [12].

In order to understand the nuclear reaction, one has to look for all the nuclear reaction channels and their occurrence probabilities [13]. In Figure 2, it is shown an incident particle, where we have chosen a neutron that collides with an atomic nucleus. It may open the scattering reaction channel where the incident particle scatters elastically under the angles $\sigma_1$ or $\sigma_2$ or non-elastically, under a different angle, depending on the amount of energy absorbed in internal processes, while the nucleus recoils under the angle $\theta$, with different recoil energies, given by the conservation of momentum and energy principles [14].

Another option is to open the neutron absorption channel, plastically colliding with the nucleus and creating a compound nucleus that recoils forward [15]. This is what is usually called a transmutation [16]. The compound nucleus results usually in an excited state, gaining energy is excess and gets rid of that by nuclear decay, where it expels small particles as electrons, protons, neutrons or alpha. Sometimes this is not enough, and the nucleus emits larger particles as $^{12}$C or bigger [17], case that is conventionally called fission.

All the expelled particles share the energy in excess, and the process continues until all the energy is released, and particles become stable.

Figure 2a is referring to the Fission products occurrence probability and its dependence on the atomic mass [19, 20]. The curves presented in Figure 2a, represent the yield after a time of operation [21-23], and that may vary with the nuclear fuel type and isotope, neutron energy or spectrum and the time the statistical measurement (tally) was made [24].

Inside matter lattice, the excess of energy that is released as kinetic or photon energy.

As for the record, the images of the nuclei are just ideograms, it is believed that inside nucleus there is a “soup of quarks” [18] and the probabilities are applied due to our lack of control over the quantum states of the interacting assembly. If quantum environment where the interaction takes place might be controlled, by coherence or entanglement, the desired quantum states transitions might be obtained and the result of the reaction will be predictable and controllable.

In the actual nuclear physics, we use the notion of reaction cross section, in order to quantitatively describe the output of each type of reaction, but for a singular interaction the outcome is unpredictable.

In Figure 2b, one can observe a symmetry from the reactants half of the total mass, with a dip in the middle, and a rise of the middle mass probability and tail heavy masses with the neutron energy, and over 20 MeV the fission gradually changes into spallation, increasing the probability of occurrence of the high mass elements.

In Figure 2c, it is given the yield in “%” in a linear scale for the high mass elements, and these are the elements we mainly consider in a 5% approximation; no element with mass >155, or with mass between 115-125, or <85 amu [25-27].

As one can see in Figure 3 some anomalies from continuous energy decrease with mass are experimentally obtained. It is also easy to observe that the fission products behaves similar with medium mass ion beam from a particle accelerator, and that means it behaves as a charged particle in interaction with matter or the nuclear fuel surrounding a fission spot, starting at few nm distance from the fissioned nucleus, where the impact of the local atomic implosion due to fission act is fading fast enough [28]. From this point of view one may distinguish three distinct areas:
1 – Fission locus vicinity comprising a 5-10 atoms range around the fissioned nucleus that is affected by the atom collapse where the nucleus disappeared into fission products suddenly accelerated to a top speed, taking with them some of the electrons, shooting EM radiation in the form of prompt gamma, neutrons and neutrinos in all direction as a skew 3D wave for few picoseconds, followed by a high vacuum, and collapse or implosion, that activates modal resonance oscillations in the lattice;

2 – Ionization based fission product stopping range, representing up to 80% of the total range in the material of a fission product because the speed of the dynamically charged particle is high enough and the short duration electric field interaction is displacing only the electrons in the atomic orbitals, and is not long or strong enough to expel the nuclei, but only to give them a shake. This range can be seen in Figure 4 – left under the pointer “Trajectory”, up to “Stopping range” and the energy deposition by ionization of the surrounding material, in our case urania (Uranium oxide) fuel, is seen in Figure 4 - right under ionization, down to “increased damage area” zone.
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3 – Bragg peak end of stooping range, where nuclear recoils dominate. The particle slows down, the electric field interaction time becomes longer enough to make the nuclei in the lattice move far enough, as to become interstitial defects. An important amount of energy is deposited in Bragg peak zone where increased damage occurs, in Figure 4-right, and all the fission products come to rest in the “stopping range” in Figure 4-left [29, 30].

As one can see, because fission products behave like an accelerated heavy ion their track in matter may be simulated using SRIM Monte Carlo code, in 2D giving reliable intuitive information on what may happen in 3D where the interaction cross section decreases with the distance from origin, because of quasi-spherical characteristic of the fission product’s associated wave, up to hundred of inter-atomic distances.
where the associated wave becomes quasi-planar, and match the code assumptions.

**Micro-nano-engineered nuclear fuel**

According to SRIM simulations, the range the fission products travel is about 15 microns in most of the fuel materials and about 8-10 microns in LBE (Lead Bismuth Eutectic).

It is known that the actual nuclear fuel is badly damaged by fission products, which make it brittle and swallow, and when it is warmed up and cooled down it cracks, mainly because fission products deposit their energy as heat towards the center of the fuel pellet, making the fuel expand more in the middle than on its borders, and ceramics does not resist to shear or stretching efforts [31].

Actually, we like the fission products power deposition as heat, but we like to avoid the Bragg peak, which transforms the fuel material ceramic structure into a few cubic microns plasma, with many dislocations per atom (DPA), and many remnant structural defects [32].

The fuel dispersion concept is used since 1956 [33-35], and was based on making fuel grains small and mix them inside an inert matrix more resilient to Bragg peak damage.

The question: “What may happen if the fuel bead is shorter than fission product range?”, remained unanswered until the recent years. The answer to this question is twofold: if the fuel bead is embedded in an inert matrix as YSZr [36]; the result is well predicted by the dispersion theory [25] and many experimental results, but if this micro-bead is immersed in a liquid metal [5, 29, 30, 37], and the Bragg peak also called fission product spike takes place outside the bead in the liquid, almost no remnant damage is produced [38]. The liquid has no structure, and the fission spike creates a pressure burst, a few cubic-micron plasma and an implosion, similar to cavitation process [39], and after that the liquid comes back occupying all the space, trapping the fission product.

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**Figure 5. Fission products path in a micro-bead material called “Cer-Liq-Mesh”**
The beaded fuel structure

In Figure 6, a new micro-nano engineered fuel concept also called “Cer-Liq-Mesh” is presented, with the meaning that Ceramic fuel beads, as made of oxides like urania, plutonia, are stabilized on a 3D elastic Mesh, made of tungsten-Rhenium or stainless steel, nickel, titanium or Moly and is immersed into a Liquid metal that can be silver, mercury, lead, bismuth, sodium, potassium or mixtures.

Figure 6 is rather an ideogram of an elementary cell, than the real 3D simulation, which looks more complex, and shows the principle of operation of such a fuel. It is a functional material, because it uses the nuclear reaction’s kinematics to self-separate the fission products from the fuel, leaving the fuel beads almost free of fission products.

The radial distribution of the fission products is shown in the upper right chart, in the situation when the liquid is frozen and allows their accumulation in place at the end of the stopping range. On the right side chart is shown the radial temperature distribution that is milder than what is today got in urania fuel pellets and is a goof fit for the structure.

A nano-layer for chemical stabilization to coat the bead and wire is recommended.

Beads need to be stabilized from reacting with the immersion liquid, and the usage of carbon based materials prohibits the usage of oxides, where the oxygen has the smallest neutron absorption cross section, but carbides or nitrides with a little higher n absorption, are allowed.

Maintaining the normal properties at high temperature [40] is important but for the moment operation temperatures over 1200 C seem unreasonable because this is the maximum temperature accepted by the present gas turbines [41-43].

Making the beads smaller than the fission product range is shown in Figure 6 ideograms, but the space between the beads have to be large enough to prevent the implantation of the fission products originating from a bead into the neighbor bead, with the associated end of range Bragg peak.
Using the elastic properties of the stabilizing wires in a 2D fuel bead mesh that may be profiled to fill a conic surface where a number of beads are connected to the central rod and the peripheral beads are connected to some wires touching the cladding [44], an interesting stretchable structure may be obtained as shown in Figure 7-right, bottom, which shows a transversal cut through the mesh planes of the conical structure.

This structure has the mesh made of carbon fiber wires with the Uranium carbide deposited on wire cross creating a bead of about 15 microns diameter placed at about 20µm apart, on 5µm thick carbon wires. At the beginning, the wires will be stretched for a double interstice or distance between fuel beads, but with burnup will be relaxed and beads will come closer, modifying fuel reactivity.

The fission products released in the drain liquid, may move inside based on their own buoyancy if the fluid is static, and higher density molecules will sink while lighter molecules will rise gently among the beads up to the top of the fuel tube. In the situation the drain fluid is moving in an open fuel tube circuit, the fission products will be continuously removed from the nuclear reactor’s core, somewhere outside into a separator unit. In Figure 7-lower-left is shown the case of $^{135}$Xe buildup in a new reactor core, made with the actual pellets, compared with what might happen in a “cer-liq-mesh” flowing drain liquid core, where the Xe peak is obtained outside the reactor core and will not capture one extra neutron to further transmutation, and this way its neutron poisoning effect will be drastically dimmed.
As it was previously shown, there are several constructive versions for the “Cer-Liq-Mesh” fuel, by making the right choices for the fuel, liquid, coating and wire mesh. Figure 8 shows the potential improvement in pellet’s structure thermal conductivity various combinations may bring. The red curve shows the variation of thermal conductivity with temperature for urania (UO\(_2\)) [45]. The light-blue curve, above, shows the thermal conductivity variation with temperature for Lead-Bismuth-Eutectic (LBE) that stops at about 1900 K, the boiling point. The light-green curve shows the effective thermal conductivity of a mixture of 50% LBE in Urania, and it is seen that it is 4-5 times higher than the pure Urania’s thermal conductivity [37]. A 20 times improvement in thermal conductivity may be obtained if Sodium (Na) is used, which is much lighter. Uranium nitride by itself has the thermal conductivity higher but closer to LBE, and no significant increase might be achieved, while metallic Uranium is by a factor of 10 better than urania with regard to thermal conductivity [46].

Finally, one may observe that a “Cer-Liq-Mesh”, structure has the temperature range of a ceramic material and the thermal conductivity similar to a metallic fuel.

**Fuel compression to adjust criticality variation**

As previously described, the beads are placed on a wire mesh, where wire exhibits elastic properties. In a 3D structure, say, a cube, inside the space remained free from fuel bead and wire, the drain liquid is inserted, as shown in Figure 9 lower-right corner, forming an elementary cell C4, where the 4 beads, approximated with spheres may touch each other or may sit at a distance. The dimension L and D may vary simultaneously or independently, giving various parallelepipeds. Let’s suppose that the beads having a radius “R”...
form a cube with the lateral “L”, from corner to corner placed in the center of the bead. In the initial position, the beads sit apart, at a distance L, forming a cube cell with volume L³, where $4 \pi \frac{R^3}{3}$ is occupied by the fuel and $3\pi r^2(L-R)$ is occupied by connecting wire. The volume remained to be occupied by the drain liquid is $L^3 - 4 \pi \frac{R^3}{3} - 3\pi r^2(L-R)$, and the only variable is L, that varies as the upper-left ideogram shows, from a distance down to very near where the beads touch each other.

The ratio between drain fluid and fuel was traced, showing the dilution factor that determines the nuclear reactivity of the fuel that is able to make the reactor critical.

It is some point on the curve where the reactor is critical, at a certain power density and operation parameters, and that is the point to start, and when compress the fuel harder it becomes super-critical. Burnup reduces the fuel and criticality, producing fission products that absorb neutrons, reducing even more the reactivity, therefore in order to keep the structure critical one needs to eliminate the fission products and compress a little bit the structure, in order that the dilution for criticality to be maintained constant, or gently reduced. As was previously mentioned, the minimum distance between beads has to be greater than the range of the fission products, in order to avoid implantation on the opposite beads, and a fuel contamination [47].

There is possible to use the “Cer-Liq-Mesh” structure in sealed cladding tubes, simulating the actual pellets, without compression mechanism, where the fission products will be deposited on the bottom or top of the pellet cylinder, or in open fuel tubes where the drain liquid is slowly flowing, taking away the fission products, as shown in Figure 10.

The compression of the fuel structure
Cubical geometry volumetric dilution variation
The new reactor concept has a strong impact on fuel cycle, because by continuously eliminating the fission products while maintaining the transmutation products, also called breeding products inside the bead, and using gentle compression, it drives to a breed and burn scheme where most of the fertile $^{238}$U is burned out, reducing the need for enrichment, and drastically reducing the proliferation risks.

Proliferation and terrorism are complex hot subjects, being sources of good paid jobs for a class of personnel that are unqualified for other more productive jobs, and in some countries is exaggerated over the limits of paranoia, in most of the cases ending in a 3G approach (Gates, Guards and Guns), that are obstacles for collaboration and R&D.

This is also related to the self-trust of each nation in its people in charge, and local grown terrorism, which is a real danger for developed states, due to inappropriate social policies, disgruntling their own people and turn them in hateful terrorists.

The concept presented here has this good feature of bringing no Uranium or Plutonium outside the nuclear reactor structure, delivering out, via a separator unit, the fission products, in small amounts that have the risk of being used in little dirty bombs, which is discouraged, because after separation and partitioning, the available small amounts will do minimum harm and require a lot of skill and effort which makes this type of actions to be highly unpractical, for any type of unbalanced individual, but has a sensibility to criticality accidents, and if the fuel is compressed over criticality position it becomes supercritical, driving to higher neutron fluxes, power and temperature and a fail safe system has to be in place that to poison the reactor and shut down the nuclear chain reaction, that to double defense in depth preparedness based on negative worth of temperature, and liquid voids.
Online separation purification unit relies on the transport of the fission products by the drain liquid, liquid, which improves fuel’s cooling capability, but does not replace the nuclear reactor cooling agent. Inside the drain, fluid takes place a micro-flow, based on micro-hydraulics parameters that are different from the parameters used in cooling agent circuit. In the micro-flow, the interface liquid-fuel forces play an important role, and at limit, these forces are responsible if fission products may stick on the fuel's coating surface or may stay stable inside the drain liquid, and be carried out into the separator unit. As shown in Figure 11, the separation unit, starts with a drain fluid accumulator, which makes a voxel of drain fluid spend about 1 week and cool down from 5% of the operating power to about 0.1% by allowing short lives fission products decay inside a shielded cooled area, outside the nuclear reactor core, preventing them from increasing the neutron absorption cross section, and poisoning the nuclear reaction.

After this time, a centrifugal separator is used, allowing the light and heavy fission products concentrate and be separated from the drain fluid and be partitioned on types of materials, later stabilized and transported for dispositioning into national fission products storages. These materials are not waste or trash, as considered today, but future ore, for novel radioisotope-based applications in the future, and is better to store them than dump into an underground geological repository.
Physical properties of the fission products

The cleaner drain fluid is passed through high extraction power units, able to purify the drain fluid under ppb (parts per Billion) level, and further reintroduced in the circuit.

Once the nuclear reaction kinematics was used to extract fission products from the fuel, there is no need to use chemical processes based on urex or purex methods, which require to dissolve the entire nuclear material into hard, aggressive acids as HCl-HNO$_3$ etc., and use expensive ion exchanger resins.

The proposed separation methods rely on physical properties mainly, such as density, used on separation centrifuges, and magnetic susceptibility, which groups the materials into ferromagnetic, para- or diamagnetic, which may be used as separating forces.

Cluster and atomic segregation properties active in liquid-solid interfaces may be used in Czochralski-like drain liquid purification that to produce a pure solid and fission products concentrate that to be re-centrifuged and further separated. Figure 12 shows the Fission product distribution as 2 sphere caps overlapped on the
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Only the paramagnetic material exhibits gradient field selectivity, being the upper mass, half of each probability density distribution. Using density and magnetic properties one may partition the fission products in about 4-6 categories (mass in centrifuge, magnetic (brown, red, green) in magnetic separator and densities, that may be applied to all selections), and only after obtaining a concentrate in each category, more selective separation processes might be applied, which may rely on other physical and chemical methods. The difficulty is related to the very low concentration of the fission product in the drain fluid, for an operation of 100 MW-Day produces 100 g of fission products, distributed in more than 100 liters of drain liquid, and the law of masses, makes extraction very difficult at very low concentrations, where only liquid-solid interface nano-cluster affinity, may work efficiently, even better than ion extraction resins, that are also selective to the type of ion.

There are very few gases among fission products, as Kr, Xe, while Ar and Rn have very low probability of occurrence, but even transitory passage through gas phase as Cs, this instantaneous lower density or large atomic radius, may be an useful advantage.

Presence of O, C, N or F may be a radiolisys byproduct, in the fuel and its coating. Po, At and Rn may appear as transmutation products from LBE, or Mg, Ca, Al, Sc may occur in NaK systems. Fuel transmutation products, that have a recoil range in nm are retained inside the micro-bead, and are further used in breed and burn process.

Because by eliminating the fission products the nuclear reactor exhibits a surplus of neutrons compared with the actual neutron balance, that may be used for near-perfect burning in breed and burn processes, or to be used in distinct, neutron absorption processes, as some fission products rectification, actinide burner or separate column breeding or isotope production [48]. For the moment, these functions are avoided in nuclear electric power reactors, because trend to complicate the operation due to variable reactivities that may occur in liquid processes.

Conclusions

The use of engineered micro-nano-beaded structure drew a new type of “dispersion” fuel, where a liquid is used instead of an inert matrix, driving to a nuclear reactor structure that resembles a MSR (Molten Salt Reactor) but the fuel is steady and only the fission products are circulating in a drain fluid that is by orders of magnitude less chemically aggressive.

The project is in it’s infancy, TRL=3 (Technology Readiness Level) with some collateral experiments pushing it towards TRL4, but not yet there, where many aspects remained to be clarified yet, to make it operational.

The enhancements in the direct extraction fuel which are susceptible to lead to a transmutation process are challenging, but at hand in these micro-nano structures.

The theoretical simulations based on SRIM and MCNP are near their limit, many aspects are yet to be clarified, and codes to be improved.

New separation and stabilization on-line units with direct extraction of the fission products and transmutation products are proposed, where micro and nano flow simulations are more difficult to be performed using the actual hydraulic codes, due to high gradients, and plenty of transitory phenomena.

Drain liquids make this class of nuclear reactors resemble the living beings, for example trees, or plants that transport via drain fluids and capillarity various substances.

As shown in Figure 1, nano-particles based fuels may treat nuclear material breeding, and nano-particles and nano-clusters in complex nano-structures is susceptible of generating novel interesting applications.

Potential benefic effects are applied on nuclear reactor neutron balance, fuel damage mitigation and reduction, potential proliferation robustness increase finally improving the entire...
nuclear power systems, procedures and the fuel cycle.

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