Quasi-nano-clustered fuel for enhanced transmutation products separation

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Abstract

The actual separation techniques, relays on used fuel processing, that turns to be a difficult process, because it’s accumulated radioactivity requiring hazardous processes.

An opportunity to ease the separation of the nuclear reactor transmutation products is given by the use of the nano-cluster’s special properties. The internal nano-cluster processes may address a large scale of transmutation products from actinides, where the interest is in fuel breeding to rare isotopes for industry and medicine application, with the condition that the transmuted product to differ from the nano-cluster elements. The new technology described uses the powder technology to create by sinter a new type of fuel element with open porosity, large enough as to allow a washing liquid to pass through it, similar to a frit metallic water filter.

The usage of the washing liquid may further improve the extraction capabilities by fluid-grain interface match. Using a smooth flow the fuel structure might be cleaned up of the transmutation products that may be driven in relatively short time outside the high neutron flux zone, preventing the secondary neutron absorption driving to purity loses. It requires a continuous microchemistry toolbox; outside the reactor in order to separate the transmutation products from the liquid, or another safe liquid transfer system. A novel design of reactor including the two structures will deliver both new fissile fuel and radioisotopes together with high-grade heat for power production. As a consequence the fuel cycle have to be redesigned and enhanced to mitigate the new aspects of storing the separated fission products and minimal nuclear waste.

The novel system is in research phase but it is susceptible to reduce the cost of the transmutation products making them more pure and available.
Introduction

From the total of 443 nuclear reactors operating in the world, more than 80% are using fuels made of ceramic materials mainly uranium, the rest are using metallic, cermet, nitrides, molten salts and mixtures.

The actual use of low-grade mixed fuels is driving to a high isotopic complexity of the nuclear waste and the usage of complex, hazardous used fuel processing as Urex and Purex technologies [1,2] that also exhibit a proliferation hazard. The fuel processing and separation techniques used turn out to be difficult because fuel’s accumulated radioactivity driving to hazardous processes. Many countries have long-term nuclear power strategies based on the future need for the conservation of uranium resources by the deployment of fast reactors and some are developing thorium-based reactors.

During the last half-century the used fuel reprocessing reason has been to recover unused uranium and plutonium in the used fuel elements and thereby close the fuel cycle, gaining some 25% more energy from the original uranium in the process. The reduction of the volume of material to be disposed of as high-level waste to about one-fifth, and the level of radioactivity in the waste form, reprocessing it to a much smaller value so as after about 100 years to fall more rapidly than in used fuel itself was important too.

Fuel recycling is an integral part of these national strategies often the domestic reprocessing capability is based on the prior development of the related expertise for weapons purposes but civilian nuclear fuel reprocessing of, is done commercially on a large scale.

In the last decade interest has grown in recovering all long-lived actinides together to recycle them in fast reactors so that they end up as short-lived fission products. This policy is driven by two factors: i) reducing the long-term radioactivity in high-level wastes; ii) reducing the possibility of plutonium being diverted from civil use, thereby increasing proliferation resistance of the fuel cycle. If used fuel is not reprocessed, then in a century or two the built-in radiological protection will have diminished, allowing the plutonium to be recovered for illicit use but though it is unsuitable for weapons due to the non-fissile isotopes present.

The Purex process is used in all of the world’s reprocessing plants and enables the partitioning, or separation, of the spent fuel into its components: uranium, plutonium, minor actinides (MA) and fission products.

Plutonium can be recycled for consumption as mixed-oxide (MOX) fuel in conventional power or fast reactor fuel [3]. Fissile uranium taken from the fuel can also be recycled as reactor fuel but, because of the still low cost of uranium, this is done in Russia only.

The fission products and the MA are nowadays embedded in molten glass for long-term burial by dispositioning in a geological facility. The chemical form of some isotopes can be altered to decrease their mobility and radiotoxicity in so-called conditioning processes. Reprocessing can reduce the volume and improve the form of these wastes prior to geological storage, but repository is still needed.

Transmutation is aimed at modifying the radioactive isotopes, primarily the long-lived fission products and the MA, by neutron bombardment. This subject requires more research on improved partitioning and the development of subcritical accelerator driven systems. Thus, transmutation offers the potential for the ultimate mitigation of fuel waste but only over the long term.

Up to now, Canadian reactors have exclusively used a once-through fuel cycle in which the fuel is removed from the reactor and stored at the reactors sites pending decisions on a long-term option for its disposition. More advanced fuel cycles are available for CANDU reactors that are primarily aimed at the optimum use of uranium. Several of these fuel cycles have a requirement for reprocessing. A future decision to reprocess CANDU fuel either for reasons of uranium conservation, or more likely to reduce the volume and radio toxicity of the fuel, will encounter almost no purely technical obstacle to domestic reprocessing.
In recent years, the developments of nano-technologies made more obvious potential applications in nuclear power.

The nano-clusters offer modified chemical affinity mechanisms that may increase extraction even when the recoil was not enough to produce implantation into the liquid. In accelerator isotopic production systems, materials are produced by nuclear recoil methods mainly for short-lives ion beams based medical applications.

The current theoretical approaches are based on the property that a nano-cluster has an accelerated rejection capability of point defects due to a shape factor intensification of the crystalline field induced self-stress, similar to the effect of surface tension occurring in liquids. This feature of self-stress interacts with the stress induced by fission generation of point defects, making their influence on the crystal to be reduced while the defect mobility is preferential axial increased as a function of crystal shape and interface faceting [4].

Transmutation

Table 1 shows a wide range of minor actinides and fission products that water-cooled reactors (with light water PWR and with heavy water CANDU) are producing. The long-lived products are increasing the radiotoxicity of the waste storage for very long time and therefore they represent the first candidates for transmutation.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>LWR [%]</th>
<th>CANDU [%]</th>
<th>T_{1/2} [y]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Fresh</td>
<td>Spent</td>
<td>Fresh</td>
</tr>
<tr>
<td>232 U</td>
<td>.1-3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>234 U</td>
<td>.1-3</td>
<td></td>
<td></td>
</tr>
<tr>
<td>235 U</td>
<td>3.30</td>
<td>0.81</td>
<td>0.72</td>
</tr>
<tr>
<td>236 U</td>
<td>4.7</td>
<td>0.51</td>
<td></td>
</tr>
<tr>
<td>238 U</td>
<td>96.70</td>
<td>94.30</td>
<td>99.28</td>
</tr>
<tr>
<td>239 Pu</td>
<td>0.52</td>
<td></td>
<td>0.27</td>
</tr>
<tr>
<td>24(0,1,2) Pu</td>
<td>0.36</td>
<td></td>
<td></td>
</tr>
<tr>
<td>240 Pu</td>
<td>0.28</td>
<td></td>
<td></td>
</tr>
<tr>
<td>241 Pu</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>242 Pu</td>
<td>0.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td>237 Np</td>
<td>0.11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>241 Am</td>
<td>0.0</td>
<td></td>
<td></td>
</tr>
<tr>
<td>243 Am</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>244 Cm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>245 Cm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>247 Cm</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fission products</td>
<td></td>
<td>3.50</td>
<td>0.80</td>
</tr>
<tr>
<td>Fission pr.</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>85 Kr</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>90 Sr</td>
<td>.15</td>
<td></td>
<td></td>
</tr>
<tr>
<td>137 Cs</td>
<td>.08</td>
<td></td>
<td></td>
</tr>
<tr>
<td>126 Sn</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>99 Tc</td>
<td>.05</td>
<td></td>
<td></td>
</tr>
<tr>
<td>135 Cs</td>
<td>.07</td>
<td></td>
<td></td>
</tr>
<tr>
<td>93 Zr</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>107 Pd</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>129 I</td>
<td>.05</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Specific actinides and fission products reactor yield
The fission products are also presenting interesting properties, and the transmutation of the long lives into other stable or short life elements remains an interesting option. Some of the fission products have direct economic uses in nuclear medicine and industry therefore their high purity separation is desired. Some of the minor actinides as $^{244}$Pu and $^{249}$Am have uses in the new space programmes as nuclear radio-batteries.

From the point of view of a nuclear repository, it may be desired as the products to be well separated and stabilised and deposited as to generate the future rare ore deposit for the future applications demand. In Table 1 values show large variation, or if they were very low in ppm range, have not been added in the table.

The radioactivity of LWR generated fission products is the key element in prioritising on the fission products need for transmutation and partitioning. As seen in Figure 1 the used fuel may exhibit after 100 years a radioactivity of about 10 Ci/kg and a radioactive power less than 50 mW/kg.

**Figure 1: LWR 40 MWDay/t FP radioactivity decay**

![Graph showing radioactivity decay over time for various isotopes.]

Figure 2 shows the classification of plutonium after the usage and its source. It is stated that reactor plutonium may not be good for building the explosive devices making them fizzle but still represent a high proliferation threat [5]. Plutonium represents the key for the complete usage of nuclear resources [6]. Plutonium as material is very complex [7], even under the aspect of being benign [8] or malign [9] to human civilisation. The nature of the plutonium and HEU usage depends on human action, smart politics and great science may prevent the malign usages [10]. The actual tendencies to dilute it back to low concentration and immobilisation in the inert matrices [11] is not a good economic solution, but is a means of getting rid of “threat” materials [12, 13].

**Transmutation products**

Table 2 shows some of the radioisotopes of interest that may be produced directly by taking advantage of nuclear recoil kinematics inside nano-cluster materials in order to obtain enhanced extraction.

Depending on the spectral distribution of the transmutation cross-section shown in Figure 3 the compound nucleus gets different recoil energy.

Figure 3 shows that from the many medical interest radioisotopes made now in a nuclear reactor only a few, transmutations that decay by a charged particle changing the chemical element may take advantage of the recoil kinematics and nano-cluster enhanced separation effect.
Figure 2: Plutonium classification

Table 2: Some nuclear reactor produced isotopes

<table>
<thead>
<tr>
<th>Product</th>
<th>Half-life</th>
<th>Production scheme</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Fuel breeding</td>
</tr>
<tr>
<td>$^{235}$Pu</td>
<td>24 100 y</td>
<td>$^{238}$U(n, $\gamma$)$^{239}$U($\beta^-$)$^{239}$Np($\beta^-$)$^{239}$Pu</td>
</tr>
<tr>
<td>$^{235}$U</td>
<td>&gt;1 000 000 y</td>
<td>$^{235}$Th(n, $\gamma$)$^{235}$Th($\beta^-$)$^{235}$Pa($\beta^-$)$^{235}$U</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\alpha$ emitters radioisotopes</td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>87.6 y</td>
<td>$^{237}$Np(n, $\gamma$)$^{238}$Np($\beta^-$)$^{238}$Pu</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>431 y</td>
<td>$^{239}$Pu(n, $\gamma$)$^{240}$Pu(n, $\gamma$)$^{241}$Pu($\beta^-$)$^{241}$Am</td>
</tr>
<tr>
<td>$^{106}$Ru</td>
<td>17 s</td>
<td>$^{100}$Tc(n, $\gamma$)$^{100}$Tc($\beta^-$)$^{100}$Ru</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Medical interest radioisotopes</td>
</tr>
<tr>
<td>$^{64}$Cu</td>
<td>12.7 h</td>
<td>$^{63}$Zn(n, p)$^{64}$Cu</td>
</tr>
<tr>
<td>$^{65}$Cu</td>
<td>2.58 d</td>
<td>$^{64}$Zn(n, p)$^{65}$Cu</td>
</tr>
<tr>
<td>$^{47}$Sc</td>
<td>3.35 d</td>
<td>$^{47}$Ti(n, p)$^{47}$Sc</td>
</tr>
<tr>
<td>$^{33}$P</td>
<td>14.28 d</td>
<td>$^{32}$S(n, p)$^{33}$P</td>
</tr>
<tr>
<td>$^{38}$P</td>
<td>25.3 d</td>
<td>$^{37}$S(n, p)$^{38}$P</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>1.91 y</td>
<td>$^{226}$Ra(n, $\gamma$)$^{227}$Ra($\beta^-$)$^{227}$Ac(n, $\gamma$)$^{228}$Ac($\beta^-$)$^{228}$Th</td>
</tr>
<tr>
<td>$^{227}$Ac</td>
<td>21.77 y</td>
<td>$^{226}$Ra(n, $\gamma$)$^{227}$Ra($\beta^-$)$^{227}$Ac(n, $\gamma$)$^{228}$Ac($\beta^-$)$^{228}$Th</td>
</tr>
</tbody>
</table>

Figure 3: Isotopic production easiness
**Fuel breeding**

Some reactors are required of being capable to produce high purity fissile fuel ($^{239}$U,$^{239}$Pu) to use all the actinide resources [14,15] ($^{233}$Th,$^{239}$U) [2]. Using the transmutation processes shown in Eqs. (1,2):

\[
^{238}U + \frac{1}{2}n + \tilde{v} = ^{239}U + \beta + \tilde{v} = ^{239}Np + \beta + \tilde{v} = ^{239}Pu
\]

(1)

\[
^{237}Th + \frac{1}{2}n + \tilde{v} = ^{237}Th + \beta + \tilde{v} = ^{237}Pa + \beta + \tilde{v} = ^{239}Pu
\]

(2)

These reactions are usually accompanied by other parasitic reactions having as consequence the production of undesired isotopes as is shown in Figure 4.

**Figure 4: Transmutation reaction diagram**

The undesired reactions in the upper-left side of the picture are due to long exposure inside of the nuclear reactor to the neutron flux.

The fast removal from the hot zone of the newly produced fuel, and good isotopic purity minimises the probability of occurrence of the undesired reactions and parasitic concentration in the newly produced fuel as:

\[
^{239}Pu(n,\gamma)^{240}Pu(n,\gamma)^{241}Pu \rightarrow ^{241}Am
\]

(3)

The desired reactions are for $^{239}$Pu and $^{241}$Am when these are produced in a high purity as transmutation products to be used in alpha batteries.

Therefore $^{237}$Np in $^{239}$Pu is a desired transmutation if this is not un-purified by $^{236}$Pu or $^{239}$Pu:

\[
^{237}Np(n,\gamma)^{238}Np \rightarrow ^{238}Pu
\]

(4)

Getting $^{237}$Np it turns more difficult because in ore $^{235}$U has very low concentrations of about 50-70 ppm:

\[
^{235}U(n,\gamma)^{236}U(n,\gamma)^{237}U[&^{238}U(n,2n)^{237}U] \rightarrow ^{237}Np
\]

(5)

The use of $^{235}$U to enrich in $^{236}$U by burn-up is the most common technique followed by $^{237}$Np chemical separation. It is thus possible to use $^{237}$Np to further transmute into $^{239}$Pu by n capture.
**Isotope production by transmutation**

A successful, early $^{238}$Pu production demonstration test was carried out in 1967 in N Reactor [16].

The so-called “heat source grade plutonium” is mainly made of $^{238}$Pu, and all the rest of isotopes being undesired, reducing material performances.

The alpha emitters radioisotopes are the most interesting elements for powering the alpha batteries or RTG elements, that have a set of valuable features for space mission, mainly the simplicity of operation and the robustness. The nuclear waste is the main resource for such radio-elements. The waste fuel reprocessing is a very difficult operation, making the cost of many isotopes prohibitive.

Four main reaction paths are presented in Figure 5. The first path is made starting from $^{236}$U, by $(n,\gamma)$ transmutation via $^{237}$Np up to $^{238}$Pu. Another path of nuclear reactor transmutation is to use $^{240}$Am-$^{241}$Cm cycle. This looks like economic nonsense because $^{241}$Am is suitable to RTG sources and an important part of it will be lost in the process, but drives to a more pure $^{239}$Pu. An accelerator-based process is the $(\alpha,\alpha)$ nuclear reaction channel applied on $^{235}$U using up to 35 MeV alpha beam. The opening of the fission channel represents a concern therefore the beam energy has to be optimised to weight the scheme factor at the desired value. A fourth path is the $^{238}$U$(p,n)$ similar to $(d,2n)$ reaction to $^{238}$Np, with the disadvantage of opening a strong $(p,fission)$ channel. All these nuclear reactions show that $^{238}$Pu is difficult and expensive to be obtained.

**Figure 5: Transmutation strategies**

An important factor of production loss comes from the prolonged exposure to neutron flux as Figure 6 shows.

**Figure 6: $^{237}$Np transmutation in $^{238}$Pu**
Following $^{237}$Np path it is shown that total production efficiency is well below 80% for short neutron flux exposure, and decays with the flux exposure time.

A trade-off between chemical separation costs and production yield or efficiency is made in practical cases aiming to minimise the production cost.

A promising alternative is the use of the properties of nano-clusters in conjunction with the transmutation traction kinematics.

**Nano-cluster sinter fuel**

The energy coming from a neutron scattering or absorption is pushing the compound nucleus outside the lattice and makes it an interstitial atom, generating defects in the nano-structure.

For an epithermal $n$ with the energy in keV region the recoil may produce is small causing a displacement of few nm in urania or thoria. In the case the nano-structure is short enough, 5 nm as in Figure 7, and is washed by a fluid having a density near to water the total range is of about 10-20 nm.

The incident particle colliding with the nucleus in repose may follow three distinct paths as showed in Figure 7 ending either in a scattering and absorption process.

**Figure 7: The kinematics of the nuclear recoil**

The processes ending in scattering the initial particle under the angle $\sigma_1$ or $\sigma_2$ and the initial nucleus under the angle $\theta$ is undesired driving to radiation damage.

The desired process in the transmutation recoil after the absorption nuclear reaction took place. The compound nucleus may be stable or unstable; usually it is unstable because the new structure has to mitigate both the new nuclear configuration and the exceeding of energy transferred from the incident particle.

The neutron activation requires no barrier kinetic energy to induce nuclear transmutation reactions. Thermal and epithermal neutrons have increased cross-section for nuclear absorption but the resultant recoil energy is not enough to dislocate the compound nucleus from the lattice.

The nano-breeding concept would take the above concept one step further by embedding nano-grains of breeding material in a liquid [17]. The relevant scale of interest is even smaller, at least as far as that of the breeding material is concerned.

The scale must be small enough so that epithermal or higher energy neutrons that are used to transform the breeding material would provide a sufficient kick to knock most of the now transformed nuclei out of their nano-clusters and into the surrounding liquid, as shown in Figure 8, which would then be flushed out of the breeding region. Figure 9 analyses the thoria case that is similar to depleted uranium [18].
Figure 8: Recoil range induced by an epithermal neutron in thoria or urania

Figure 9: $^{233}$Th recoiled out of $^{232}$ThO$_2$ in water

For any transformed breeding nuclei not directly ejected from the nano-clusters, the rejection process could be further enhanced using the properties of the nano-clusters and several other effects [19,20] as Figure 10 shows. First, there is the tendency of the atoms of the nano-clusters to arrange themselves in very stable nano-structures [21]. This is further aided by the chemical properties of the transformed nuclei. As a result, “impurity” atoms that are formed as a result of transmutations will tend to be transported out of the interior of the lattice structure to the surface of the nano-clusters, where they can enter the mobile liquid. The energy deposition from the kinetic recoil of the breeding reaction aids the mobility of any trapped transmutation products by displacing them from their lattice positions. The high temperature of the overall system further aids the mobility of the transformed nuclei. Figure 10 shows an example of a depleted urania nano-cluster that produced a transmutation towards $^{239}$Pu via $^{239}$Np and a fission act on $^{238}$U ($^{238}$U can also fission with fast neutrons). The transmutation products and the elastically recoiled products have the stopping ranges of the order of several nm. The fission product (i.e., $^{137}$Cs, $^{89}$Rb) stopping range is several tenths of a micron, enabling them to leave the nano-grains.
Additional benefits brought about by the slow, but continuous removal of the newly bred fuel are:

- The shorter residence time of the breeding product in the reactor core, reducing the presence of secondary breeding products (i.e. an undesired second transformation of the desired transmutated product).
- The ability to, much more easily (and continuously), separate out the newly bred fuel.
- The ability to fully use the breeding material.
- The production of both a richer fuel and much less waste.
- With proper design, the ability to “continuously” replace the consumed breeding material.

In a commercial nuclear reactor inside the irradiation channel the neutron spectrum is thermal with very few epithermal and fast neutrons, therefore the probability of recoil of the neutron absorption generated compound nucleus is very small. In these circumstances very few $^{235}\text{Pa}$ atoms become self-interstitial atoms (SIA) generating cascades of recoils and Frenkel defects. Most of the $^{235}\text{Pa}$ remain in their lattice knot positions behaving similar to doping materials exhibiting chemical incompatibility. Under the cluster shape pressure, similar to that obtained in a liquid droplet as an effect of surface tension, inside the cluster both the holes and impurities are moving towards the border. The pairs of cascades of holes and recoil base material are recombining, by sub-cluster and magic cluster permutations, while the impurity is under the increased diffusion coefficient that pushes it on the cluster’s surface. When it reaches the surface, the lower binding energy allows it to leave the cluster, and recombine in the next material that in our case is a chemical active fluid having free radicals, reacting with $^{235}\text{Pa}$ and forming an oxide. The beta decay occurring in surface bounded Pa helps the cluster to release the new $^{235}\text{U}$ nucleus into liquid. This kind of mechanism under the cluster shape pressure enhanced by internal sub-cluster permutation that repairs the recoil cascade of damage minimises the cluster material loss. This kind of internal mechanism makes possible an enhanced extraction of the newly generated material through the transmutation process simultaneous with an increased stability of the pellet structure. It is observed a tendency of the initial nano-grains to reshape by coalescence of holes in sub-cluster assemblies exchanging atoms for minimising the potential energy towards the magic numbers and shapes. This means that in spite the structure to be stable, it is in continuous transformation stimulated by the radiation field whose thermal spikes act as a pressure wave delivering the stimulation energy for reshaping by cluster fission and cluster fusion processes operating together to establish a global optimal at cluster interface level.
Figure 11 shows the schematic diagram of a nano-bead sinter fuel formed from various dimensions of nano-powders sintered together in a frit like pellet. In the middle, where the finest powders are caught, blocked by smoothly increasing the dimension of the powders up to the coarse layers from the borders. This process stabilises the powders inside making the pellet stable in the liquid flow. The drain liquid that washes the powder has affinity surface forces, behaving like a hydrophilic fluid, opposing a minimal dynamic pressure or surface tensions with the powder.

**Figure 11: The nano-powder pellet schematic diagram**

The nano-flow has to wash the hydrophilic structure and to have dripping level flow rates of about one tube length per month to load a high concentration of grain escaped atoms. That means, the smaller the nano-particle is the more intense the cluster properties are, driving to increased diffusion rate of impurities near boundary interface and increased resilience to radiation damage [22].

The pellet device design started from producing particles by using a plasma torch in a oxygen/noble gas atmosphere, and selecting the powder by hydro-flotation. To stabilise the powder in the cladding tube a metal sintered filter have been used at the ends with tenth of micron porosity as Figure 10 shows.

The smaller particle magnitude particles have to be placed in the middle and stabilised by gradual increasing of the particles magnitude towards the sinter filters at the end. The flow has to wash the hydrophilic structure and to have dripping level flow rates of about one tube length per week to load at a high concentration with the grain escaped atoms.

If the drain fluid makes a complete tube length in several weeks the super grade brand will be obtained.

The contamination may be produced by recoiled material as $^{233}$Th, $^{238}$U, oxygen and cladding material as well by the fission products resulted from the fission of the newly produced fissile material and by residual $^{237}$U in the case of depleted uranium (DU).

Figure 12 shows a simulation for the both cases of breeding starting from pure thoria and DU.

The efficiency of collection depends on particle magnitude and nuclear reaction channel parameters. For $^{239}$Pu the direct recoil extraction rate is about 70% for $^{238}$U, grains of 5 nm diameters and is brought up to 95% by diffusion due to $^{237}$Np incompatibility with uranium dioxide lattice. Figure 12 shows the estimated concentrations of the main products starting from the initial mass concentration in dioxide, to the final expected concentrations, after the extraction. The chart in the left side shows that main contribution in the output of the activation products from water and cladding, followed by the fission products in residual fissile material or newly produced fissile material, and the breeding products that are recoiled in the drain liquid.
Figure 12: The estimated yield

The right side chart is expanding the area of masses from 230-240 to better see the breeding and extraction process with its three phases that initial concentrations, the recoiled products result of scattering and the extracted breeding fissile products. In the simulation was not considered the nano-cluster manifestations that trend to reduce by sub-cluster level recombination the original material lost from the nano-grain, and by accelerated diffusion of SIA [23] due to cluster interface induced stress [24] eliminates with enhanced efficiency the nuclear transmutation products becoming impurities in the cluster of origin.

This type of continuous separation process has the advantage that the transmutation products are in suspension directly in the drain fluid and requires an effective physical separation method, like paramagnetic levitation separation or magnetic gradient diamagnetic-paramagnetic resonant separation. These direct separation methods and online chemistry are mainly in the conceptual stage, but have high chances to perform an effective on-line separation.

If the test is performed in a long tube getting outside the n irradiation area that holds about 1 foot above the reactor upper level the drain flow rate may be adjusted at the desired level as the assembly design in Figure 13 shows. If the drain fluid makes a complete tube length in several weeks the super grade brand will be obtained.

Figure 13: The breeding assembly
The liquid has to be compatible with the transmutation isotopes, stabilising and carrying them outside the area they were generated into an inline chemical separator unit.

The contamination may be produced by recoiled material as $^{232}$Th, $^{238}$U, oxygen and cladding material as well by the fission products resulted from the fission of the newly produced fissile material and by residual $^{239}$U in the case of depleted uranium (DU).

Discussion

The method is relatively new, and it is a mixture of experimental facts with concepts and suppositions in the theoretical modelling and simulation stage. If all the suppositions will be experimentally proved as correct and no other collateral effects not considered in the present evaluation will be further discovered the method has great chances to reduce by more than 50% the need for Ureks, Purex type of processes delivering continuous high quality isotopic products.

The main advantage of this method and device resides in its ability of producing isotopic materials with controllable purity easy to separate from the drain liquid, using the nuclear reactors, with higher yield than the accelerator-based methods and requiring less aggressive chemistry being an easy cheaper alternative for Ureks and Purex processes. These processes are not completely eliminated but their usage is minimised and applied to completely used fuels.

As resulted from the Figure 3 and Table 2 not all the isotopes are suitable to apply this nano-cluster separation method.

In order to apply the cluster separation it is required to have recoil or atomic difference to led to interstitial defects. Most of the (n,$\gamma$) reactions induced in thermal or epithermal spectrum are driving to the same chemical element, little excited at nuclear and atomic level, but without being removed from its position in the lattice. The new isotope will have little isotopic energy levels shift that distorts a little the nano-cluster shape, and field lines but more experiments are needed to learn how various nano-clusters evolve with these isotopic variations inside the structure. Extrapolating the actual knowledge it is possible that some nano-clusters to exhibit separation properties, but many combinations will have very stable configurations with a large isotopic diversity inside the structural elements, and will not be useful for separation purposes.

It also has to be stressed that having a nano-structures washed like a frit filter by a liquid, the nano-cluster's interface has to be chemically and mechanically stable and exhibit affinity with the washing liquid. Any mismatch will make the liquid not flow through, or oppose an important dynamic pressure.

The liquid purity has to be high, avoiding the fractions incompatible with the nano-structure that build-up by separation and accumulation at nano-structure's entry, finally blocking the flow. This is a similar effect to that occurring in membrane filtration units.

It is also not enough to have a nano-cluster separation element that to exhibit formation recoil and elemental transmutation during the process. It is also required to have the manufacturing capability of putting it in the right chemical combination suitable for the desired nano-beads dimensions without that associated chemicals to trap the nuclear process and to reduce the yield of produce difficult to eliminate chemical contamination.

Conclusions

Nano-particle-based fuels are opening new effective ways for nuclear material breeding believed as being more efficient than the actual nuclear fuel processing procedures if all the necessary technologic aspects will be well defined. The actual developments rely on few experiments, more simulations and theoretical evaluations, being a subject under research and innovation.
Both the theoretic and experimental data are driving to the same conclusion that the study of the nano-particles, nano-clusters and nano-structures is far from being over, being susceptible of generating novel interesting applications.

The usage of nano-clustered sinter fuels improves the process of production of $^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{235}\text{U}$ and some of the medical interest isotopes, but in some cases the production cost still remains high, due to the nuclear reaction channels the production path that have to be used. The two-stage independent $^{239}\text{Pu}$ production brings the advantage of higher purity materials and the specific production ratio control with potential of lowering the production cost.

The development of new accelerator-based systems for fusion energy and high $n$ yield spallation targets applied to $^{244}\text{Np}$ and most of the medical isotopes production may be a valuable alternative for the production of these isotopes.

Nano-technology improved nuclear reactor based methods if developed may prove the most proficient and cost effective ways for $^{238}\text{Pu}$, $^{239}\text{Pu}$, $^{235}\text{U}$ mass production.

The nano-grain based fuel immersed in a drain fluid presents real breeding advantages compared to the actual breeding products extraction procedure reducing their scale of utilisation.

The nano-cluster properties active in nano-grains exhibit very useful properties for transmutation products continuous extraction as dimension-shape dependent grain interface induced stress that provides selective diffusion enhancement and sub-cluster movements that enhance the self-repairing and self-purification effects driving to enhanced transmutation products extraction.

The present developments are in early stages of development being supported mainly by theoretical evaluations, computer simulations and a few collateral experiments; many aspects remain to be clarified.

The nano-flow simulations are more difficult in these structures due to the large number of variables spanning over a wide range of values, high gradients, plenty of transitory phenomena, that requires more development efforts.

References


