Plutonium recycling in LWRs

Jan Willem Storm van Leeuwen
independent consultant

member of the Nuclear Consulting Group

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storm@ceedata.nl

Note

In this document the references are coded by Q-numbers (e.g. Q6). Each reference has a unique number in this coding system, which is consistently used throughout all publications by the author. In the list at the back of the document the references are sorted by Q-number. The resulting sequence is not necessarily the same order in which the references appear in the text.

MOX fuel in LWRs

The plutonium recovered from spent fuel, usually labeled reactor-grade plutonium Pu_{rg} could be used to fabricate MOX (Mixed OXide) fuel elements consisting of natural or depleted uranium oxide and plutonium oxide instead of a higher content of uranium-235, to be used in light-water reactors (LWRs). At present about 30 nuclear power plants, mostly in Europe, are using MOX fuel elements, not more than about 30% of the core. Special reactor designs are required to replace all enriched uranium fuel in the core fully by MOX fuel. Reprocessing of commercial nuclear fuel to make MOX is done in the United Kingdom and France, and to a lesser extent in Russia, India and Japan [wiki-*mox* 2017] Q741.
Plutonium from reprocessed fuel is usually fabricated into MOX as soon as possible to avoid problems with the decay of short-lived isotopes, in particular Pu-241 that decays (half-life 14.1 years) to americium-241, a strong gamma emitter. Within 5 years typical reactor-grade plutonium would contain too much Am-241 to safely handle [WNA-*mox* 2016] Q246.

How much natural uranium could be displaced by using MOX fuel instead of enriched uranium in nuclear power stations?
According to [WNA-*mox* 2016] the current commercial MOX fuel, equivalent with enriched uranium at 4.2% U-235, has an average plutonium content of 9.5%, containing 65% fissile plutonium (Pu-239 + Pu-241).
This study assumes that depleted uranium is used for the MOX fuel, because reprocessed uranium poses problems, see next section. The fissile content of depleted uranium (about 0.3% U-235) is ignored for convenience.
Based on the WNA figures 1 Mg reactor-grade plutonium would correspond with 10.5 Mg MOX, of which 9.5 Mg depleted uranium. To produce an equivalent amount of enriched uranium (4.2% U-235) about 100 Mg natural uranium would be needed, assumed a feed/product ratio of 9.5 and 0.3% U-235 tails assay of the enrichment. So 1 Mg Pu_{rg} could save 100 Mg U_{nat}.
According to this study the feed/product ratio should be 9.85 if the process losses of the upstream processes are taken into account, see report m19 *Advanced reference reactor and EPR*.
[WNA-*mox* 2016] states that the present global inventory of reactor-grade plutonium available for commercial reactors is 320 Mg and that this amount could save 60000 Mg natural uranium.
According to the estimate of this study 320 Mg Pu_{rg} would be equivalent to 33000 U_{nat}. This is about one half of the current world annual consumption of U_{nat}. It is unclear how the World Nuclear Asdsociation derived its estimate.
If all spent fuel of the global nuclear fleet would be reprocessed some 60 Mg/year of Pu_{rg} would become available for MOX fabrication. This could save about 6200 Mg/ year of U_{nat} , about 10% of the current annual consumption.

During burning of MOX the ratio of fissile (odd numbered) isotopes to non-fissile (even numbered) drops from around 65% to 20% depending on burnup [wiki-*mox* 2017] Q741. For that reason used MOX fuel is not reprocessed.

The MOX option has a negative energy balance, alike other nuclear concepts that depend on reprocessing of spent fuel. It takes more energy to reprocess spent fuel, including a proportional part of the energy requirements of the dismantling of the reprocessing plant, and to fabricate MOX fuel than can be generated from that fuel. Moreover, the use of MOX fuel poses serious proliferation and terroristic hazards, as will be discussed in one of the following sections.

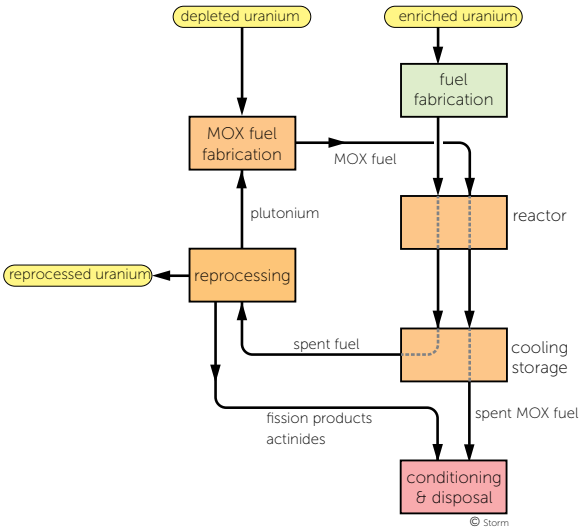

Figure 1

Outline of plutonium recycling in LWRs. The reactor starts up loaded with conventional enriched uranium fuel. The spent fuel is reprocessed, after a cooling period. The recovered plutonium is used to enrich depleted uranium. The resulting MOX fuel is placed into the reactor. Spent MOX fuel is not reprocessed. The reprocessed uranium is seldom reused, because it has a troublesome isotopic composition and is contaminated by fission products.

Reprocessed uranium

Recycled uranium, from reprocessed spent fuel, also called reprocessed uranium U(rep), has a different composition than natural uranium and contains a number of nuclides which degrade its properties as nuclear fuel [Forsey & Dickson 1987] Q239 such as:

- uranium-232,decaying to thallium-208 with high alpha and gamma activity
- uranium-234, a strong alpha emitter
- uranium-236, a strong neutron absorber; fuel with this isotope needs a higher enrichment assay or more fissionable plutonium to compensate for it,
- traces of fission products, like ruthenium-106 and technetium-99, which increase the gamma activity of the uranium
- traces of trans-uranium elements, e.g. neptunium and plutonium.

Above problems worsen each time the fuel passes the fuel cycle. The gamma activity of recycled uranium increases with the number of cycles, because of a growing content of gamma-emitting decay products.

Recycled uranium needs addition of more fissile plutonium than depleted or natural uranium, because of the neutron absorbing properties of U-232, U-234 and U-236. These even-numbered isotopes of uranium are not fissile.
Enrichment of recycled uranium by gasdiffusion or ultracentrifuge is questionable, because the uranium is enriched more effectively in the two lightest isotopes U-232 and U-234 than in U-235 and is enriched also in U-236, to lesser extent, making the enriched product strongly radioactive. The reenriched uranium would contain the largest part of the unwanted uranium isotopes and some other unwanted nuclides mentioned above as well. Besides, the enrichment plant would be contaminated with highly radioactive compounds, hampering its operation.
For above reasons reprocessed uranium has not been used in power reactors. In addition the fabrication of fuel elements containing reprocessed uranium is difficult and has to be done by remotely controlled equipment due to its high radioactivity. Utilisation of reprocessed uranium has a negative energy balance, due to high energy consumption of fuel fabrication and the high energy investments of reprocessing of spent fuel including dismantling of the reprocessing plant at the end of its operational lifetime.

The World Nuclear Association [WNA-*mox* 2016] Q246 cites a figure of 45 Gg reprocessed uranium being available for reuse, which would be equivalent to 50 Gg (1 Gg = 1000 Mg) of natural uranium. The displacement figure of 50 Gg is inconsistent with the publication [Foresey & Dickson 1987] Q239. Reprocessed uranium contains slightly more U-235 than natural uranium (around 0.8% vs 0.7%) indeed, but it needs a higher fissile content than equivalent conventional nuclear fuel, as pointed out above.
Evidently this highly radioactive material poses health risks when released into the public domain, by accidents, terroristic actions or otherwise.

Plutonium

Plutonium is generated from uranium-238 (non-fissile) by neutron irradiation in nuclear reactors. The isotopic composition of the plutonium varies with the irradiation time in the reactor. At first the fissile plutonium-239 is formed and from this isotope heavier isotopes are formed by subsequent neutron captures: Pu-240 (non-fissile), Pu-241 (fissile) and Pu-242 (non-fissile). In nuclear fuel at low burnup little Pu-239 is transformed into heavier isotopes. The higher burnup of the fuel, the longer the stay time in de reactor and the more non-fissile heavy plutonium isotopes are generated.

Weapons-grade plutonium contains typically 93.6% Pu-239 [O'Connor 2003] Q599 is produced by neutron irradiation of uranium in special military reactors. The nuclear fuel from these reactors has a very low burnup (about 100 MW(th).days/Mg) before reprocessing, so only small amounts of the higher isotopes of plutonium (e.g. the non-fissile isotope Pu-240) are formed. Higher isotopes of plutonium and trans-plutonium elements make plutonium more radioactive and less suitable for production of nuclear weapons.

Reactor-grade plutonium originates from spent fuel from civil power reactors and contains typically less than 65% fissile plutonium isotopes (Pu-239 + Pu-241). In commercial reactors the fuel elements stay far longer and get a higher burnup (33000-46000 MW(th).days/Mg) than in military reactors. Due to longer stay times in the reactor, more of the heavier plutonium isotopes are formed: Pu-240, Pu-241 and Pu-242, but also Pu-238. The even isotopes are not fissile in LWR's and in bombs. Moreover, trans-plutonium elements, e.g. Am-241, Am-243, Cm-244, are formed from plutonium isotopes by neutron capture. According to [WNA-*pu* 2016] Q247:

> The term 'fissionable' applies to isotopes that can be made to undergo fission. If a fissionable isotope only requires neutrons with low kinetic energy to undergo fission, then it is said to 'fissile'. Thus, all fissile isotopes are fissionable. Pu-240 is fissionable, as it undergoes fission in a fast neutron reactor - but it is not a fissile isotope.

Contrary to statements of the nuclear industry [WNA 2012b] Q541 reactor-grade plutonium is suitable for nuclear explosives, according to [Barnaby 2005a] Q339 and [Barnaby 2005b] Q340, [Glaser 2005] Q593, [Schneider 2007] Q590.
Plutonium has a much lower critical mass than uranium. The bare-sphere critical mass of weapons-grade plutonium is 11.5 kg (diameter 10.5 cm) and of reactor-grade plutonium 14.6 kg (diameter 11.5 cm). With a neutron reflector of 15 cm the figures are: 3.71 kg (7.20 cm), respectively 4.58 kg (7.72 cm), according to [Glaser 2005] Q593.

Pu-238 is a strong alpha emitter. By beta decay, plutonium-241 is transformed into americium-241; Am-241 is a strong gamma emitter, greatly increasing the gamma activity of the plutonium. Within a few years storage time, the concentration of Am-241 builds up to a level the plutonium cannot be handled safely anymore. With a content of Am-241 higher than 1% it has to be purified again [Hulst & Mostert 1979] Q242, a costly process. For recycled plutonium from LWR with MOX fuel, the repurifying limit due to Am-241 may be reached about one year after reprocessing. Americium-241 decays to neptunium-237, a fissile nuclide.
Most plutonium and trans-plutonium isotopes emit neutrons, as some of their nuclei spontaneously fission (the other nuclei decay by alpha or beta emission). The presence of all these nuclides makes reactor-grade plutonium a hazardous substance, with troublesome properties as reactor fuel. The problems with increasing gamma, alpha and neutron radiation aggravate with each recycling of the plutonium. The same holds true for the burnup of the fuel from which the plutonium is extracted: the higher the burnup, the longer its stay time in the reactor and consequently the less the isotopic quality of the plutonium.

Conclusion

The contribution of today's Pu inventory to the global nuclear energy supply is neglible. For the future an energy balance should be made: reprocessing of spent fuel is an energy-intensive process, see report m20 *Reprocessing of spent nuclear fuel*. Decommissioning and dismantling of a reprocessing plant might require very large energy investments see report m04 *Decommissioning and dismantling*. In addition MOX fuel fabrication may consume much more energy than conventional fuel.
A thermodynamic analysis indicates that the energy balance with reprocessing might be negative.

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