‘THE NUCLEAR FUEL CYCLE IN 2040: THE CHALLENGES AND SOLUTIONS TO ACHIEVE SUSTAINABLE NUCLEAR GENERATION IN EUROPE’
[Thomas DIRKS, INSTN Paris-Saclay/ TU München, 2017]

1. Introduction

Many European nations have been making low carbon electricity through nuclear generation, beginning with the UK in 1956. [1] The European Union currently generates more than one-quarter of all its electricity- and more than half of its low carbon electricity- from nuclear sources. [2] However, the current process for collecting, preparing, and disposing nuclear fuel uses uranium resources poorly, has risks of nuclear proliferation, and does not address the issue of nuclear waste satisfactorily.

Figure 1: From the very beginning at Calderhall [3], to the far future with nuclear fusion [4]-
How should the transition be like?

None of these problems are without solutions. The frontend fuel preparation can be updated to use advanced enrichment technologies to take advantages of depleted and reprocessed uranium stockpiles, peacefully use plutonium, including that left over from nuclear weapons programs, and cleanly extract uranium from seawater without the need for mines. Advanced reactors can more safely and efficiently use uranium and plutonium resources. Reprocessing technology can be used to significantly reduce the amount and toxicity of nuclear waste. With continued efforts to advance technology and overcome negative perception of nuclear power, a sustainable nuclear fuel cycle is certainly achievable by 2040!
2.1 Frontend: Re-enrichment of tails

There are large stockpiles of reprocessed and depleted uranium, so there is little need to find alternative uranium resources in the near future. Years ago, people enriched uranium with wasteful and energy intensive technologies. Today, high performance centrifuges dominate the enrichment market; however, a deterministic method of enrichment using a laser will probably conquer the market. This paper does not evaluate the technology, which is mostly kept tightly under wraps, but it does evaluate its impact should it fulfil its promise.

**Laser-enrichment**

There are three generations of enrichment technology. One of the first has been gaseous diffusion, which was widely used for reactor fuels. More recently, gas centrifuges have become the leading technology, since their process is less energy intensive, more flexible, and has a lower footprint. [5]

Potential of further depletion

After many years of using enriched uranium, the stockpile of depleted uranium has grown. The total amount of depleted uranium (DU) is about 1.5 million tonnes. [6] Assuming that the DU stockpiles have an average concentration of 0.3% of $^{235}\text{U}$ and laser enrichment forms tails of 0.05% of $^{235}\text{U}$, the equivalent mass of natural uranium $m(U_{\text{nat}})$ that can be formed is:

$$m(U_{\text{nat}}) = 0.3\% - 0.05\% = 0.72\% - 0.05\% = 1.5 \cdot 10^6 \text{ t} = 559,701 \text{ t}$$

Considering that the annual natural uranium demand is about 66,000 t, the re-enrichment with low tail technology can feed the entire world demand several years.

Potential of reprocessed uranium

Uranium from spent uranium oxide fuels (UOX) in light water reactors (LWRs) still contains up to 1% of $^{235}\text{U}$. Today, its enrichment is less attractive, since it contains $^{236}\text{U}$, which is a great neutron absorber. To use reprocessed uranium in today’s enrichment facilities, it has to be more enriched than natural uranium. [7] If laser enrichment can selectively separate $^{235}\text{U}$, reprocessed uranium will be much more attractive than it is today.

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**Figure 2: Visualizing the laser beams [23]**

Laser enrichment is meant to be one of the third generation processes. Whereas the first and second generation’s processes use differences in the kinetics of $^{235}\text{U}$ and $^{238}\text{U}$ to separate the two isotopes, laser enrichment takes advantage of fine differences in the two isotopes’ quantum structures, allowing extraction of specific isotopes of uranium or plutonium [5].

For instance, an international project called *Global Laser Enrichment* is developing a molecular laser enrichment process called *SILEX*; which stands for “Separation of Isotopes by Laser Excitation”. The technology is in its third generation and has been licensed to be used by the *United States Enrichment Corporation*, *GE Hitachi*, and *Cameco Corporation*, the world’s largest uranium producer. If it is successful in scaling to industrial levels, the technology will offer a number of advantages, including (1) the highest ever enrichment efficiency, (2) a smaller footprint than gaseous diffusion or centrifuge plants, and (3) the lowest anticipated capital cost of any enrichment technology. [5]
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2.2 Frontend: Uranium from seawater

Power from nuclear fission has been criticized as being an unsustainable carbon-free energy source due to limited uranium resources. Although conventionally mining uranium and using an open fuel cycle in LWRs does limit the sustainability of nuclear power, closed fuel cycles substantially reduce uranium demand and sea water extraction opens a virtually limitless supply of uranium.

Limits of conventional mining
Current international uranium reserves are defined as uranium sources proven to be economically extractable at the current market price. At the current demand of 66,000 tonnes per year, the current reserves will last for 90 years if the cost is not allowed to increase more than 50%. [8] These 90 years are based on the assumption that no new ore deposits will be found in the future. Improvements in uranium leaching or the potential of seawater extraction is not taken into account, because breakthroughs and improvements are not predictable. Therefore, these estimations regarding the reserves are inaccurate.

Advantages of seawater extraction
Conventional mining has significant drawbacks. Workers in mines have to resist high radiation doses. Leaching uranium from ores requires polluting chemicals. Mines are often far from civilisation, so logistics consume a lot of work, time and energy. Seawater extraction does not have any of these drawbacks. Almost every coast in the world can be used for extraction. Regions with strong ocean currents might be favourable, as they constantly bring in new uranium. Furthermore, it has the potential of lowering the activity and toxicity of the sea, having a net positive effect on the environment.

Figure 3: Illustration of uranium seawater farming [26]

A huge amount, a big challenge
In seawater, uranium appears as uranyl (UO$_2^{2+}$) dissolved with a concentration of 3 mg/m$^3$. For the oceanic volume, the total mass of dissolved uranium is 450 billion tonnes. [9] Today, the most reasonable approach to get uranium from seawater is chemical extraction, which has to fulfil the following criteria towards uranyl:

- high affinity while adsorption
- low affinity while rinsing
- selectivity

Additionally, the extractant should not be strongly affected by seawater conditions, including:

- high NaCl concentration
- flora and fauna that could coat the extractant’s surface

Economic competitiveness sets also some criteria that should be fulfilled:

- low investment costs
- simple dilution method on an industrial scale
- reusable for several extraction cycles

Should these development objectives be met, uranium seawater extraction has a real chance to become the big player on the uranium market.

At the current demand, the amount of uranium from seawater could feed our LWRs for nearly 7 million years. Even when the whole energy demand was based on nuclear power, the reserves would last the foreseeable future. [9]
2.3 Frontend: Weapon-grade plutonium in the fuel cycle

Since people first experienced the disastrous effects of nuclear weapons, there has been a continuous desire to decrease the amount of weapon-grade fissile material on the world. Highly enriched uranium has already been limited for many years. The elimination of weapons-grade plutonium is more challenging, but doable. Once weapons-grade plutonium has been integrated into the fuel cycle, the gain is very attractive from at least two points of view.

**Plutonium characteristics as fuel**

Instead of using UOX fuel, plutonium as PuO₂ can be mixed with UO₂ to form mixed oxide (MOX) fuel, a nuclear fuel with similar properties. Plutonium coming from industrial reprocessing of uranium fuels consists mainly of five isotopes. One defines the quality of plutonium $Q$ as the ratio of the mass of fissile isotopes to the total mass of plutonium.

\[
Q = \left( \frac{m(^{239}\text{Pu}) + m(^{241}\text{Pu})}{m(\text{Pu})} \right) \cdot 100 \quad (1)
\]

Plutonium is categorized by its isotopic composition. Mainly, there is reactor-grade and weapon-grade plutonium. Reactor-grade, usually coming from spent uranium fuel in civil reactors, has a significant amount of $^{240}\text{Pu}$. Weapon-grade plutonium was produced by very brief irradiation of fuels in special reactors. It has much lower $^{240}\text{Pu}$ content and therefore a very high quality. [11][12]

Reactor-grade plutonium is very unattractive for a fission bomb, because (1) $^{238}\text{Pu}$, $^{241}\text{Pu}$ and its decay product $^{241}\text{Am}$ are gamma emitters and a strong heat source which hinders handling and fabricating of a bomb [11]:

(2) Plutonium isotopes with even mass number, like $^{240}\text{Pu}$, additionally have a relatively large spontaneous fission rate which makes weapons less reliable and therefore very unattractive, especially for easy construable gun type designs. [11]

The isotopic composition of plutonium depends on the fuel and its burn-up. In thermal spectrum LWRs, the quality decreases as the burnup increases, because more non-fissile isotopes of plutonium are produced. Today’s LWRs have a much larger burnup than earlier reactor designs. Using MOX fuels in LWRs, the quality of plutonium decreases even further. [12]

In Table 1 and Figure 4, the isotopic compositions of plutonium from different origins and burnups are shown.

<table>
<thead>
<tr>
<th>Table 1: Isotopic composition of Plutonium in % [10]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}\text{Pu}$</td>
</tr>
<tr>
<td>Weapon-grade Pu</td>
</tr>
<tr>
<td>Magnox Pu</td>
</tr>
<tr>
<td>LWR Pu</td>
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<tr>
<td>Second gen. Pu</td>
</tr>
</tbody>
</table>

![Figure 4: Isotopic composition of plutonium from different origins (compare Table 1)]
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The issue of low-quality plutonium
The lower the quality of the plutonium, the more PuO₂ needs to be mixed into the UO₂ matrix to achieve an equivalent reactivity. High plutonium contents in today’s LWR’s fuel induce safety concerns for the following reasons:

- **Enhanced minor actinide production**
The abundance of low quality plutonium shifts the balance towards heavier isotopes. Therefore, fewer neutrons are needed to form minor actinides, which are the major concern of the spent nuclear fuel. [13][14]

- **Positive void effects**
Plutonium is more likely to fission than capture neutrons in the fast spectrum. Therefore, the harder spectrum present after a loss of coolant accident (LOCA) could actually increase the reactivity when the plutonium content in the fuel is above a certain level. [13]

- **Hot spots and cladding failure**
The more PuO₂ is mixed with UO₂, the more the fuel tends to conglomerate, resulting in hot spots and increased local fission gases, risking fuel failure. [15]

- **Increasing helium production**
Plutonium and the significant amount of minor actinides in MOX fuels have a much higher helium production than UOX fuels. This is due to their higher decay activity and their enhanced ternary fission reactions which have often helium as a product. [10]

Storing Pu for Gen IV: a risky strategy
The nuclear reactors of the Gen IV, having fast neutron spectra are the most efficient way to destroy low-grade plutonium and minor actinides. However, it is not smart to accumulate hundreds of tonnes of plutonium for a potential future technology, since (1) it is highly toxic and its storage is a security and safety issue, (2) its fissile isotope $^{241}\text{Pu}$ decays by a half-life of about 14 years and forms even more radiotoxic $^{241}\text{Am}$ [13]:

$$^{241}\text{Pu} \xrightarrow{\beta^-} ^{241}\text{Am} \quad (2)$$

Some people expect Gen IV reactors to come around by 2040, but large scale reactors that can use current and future plutonium stockpiles may be many years away.

Weapon-grade plutonium in the fuel cycle
In 2000, Russia and the USA signed an agreement committing each country to dispose 34 metric tonnes of plutonium coming from dismantled weapons. The USA has one MOX fabrication facility under construction at the Savannah River Site in South Carolina. They plan to fabricate MOX fuel for LWRs using 5% pure weapons-grade plutonium. [16]
The civilian use of weapon-grade plutonium can be worrying. A great security effort is needed for fabrication and transportation until the fuel is set in a reactor. Furthermore, weapon-grade plutonium as fuel for LWRs has a lower conversion rate than those MOX fuels from conventional reactor-grade plutonium, meaning that the fuel reactivity decreases faster. [13]

Current situation in Europe
Countries strongly dependent on foreign uranium like France are preparing themselves to be able to use more plutonium in their nuclear fuel cycle. The EPR project, actually under construction in Flamanville, will be able to operate with 100% MOX fuel load. [17]
In France, annually around 75% of spent UOX fuel is reprocessed. Spent MOX fuel’s plutonium quality is too low and therefore it is not considered to be reprocessed for the use in LWRs. [13][18]
In the UK, chemical instabilities of metallic fuels in their Magnox reactors forced them to reprocess. Without fabricating a large number of MOX fuels, the UK has accumulated more than 100 tonnes of plutonium from spent uranium fuels by 2014, without a reliable perspective for its use. [12]
Whatever the future energy strategy in Europe will look like, many tonnes of plutonium already exist. Its storage is expensive and its hazard potential is enormous. Reliable future plans for its destruction are highly desirable!
Fortunately, there are several possibilities to limit the increase of plutonium which we store today.

**Solution: mixing low and high quality Pu**

One way to limit the increase of plutonium stockpiles, or even decrease the amount we have today, is multi-reprocessing of plutonium by mixing high and low quality plutonium. The major benefits are:

- Limitation of unattractive second generation plutonium accumulation
- More efficient use of resources, saving uranium resources
- Higher conversion rate in the fuel, compared to pure weapon-grade MOX fuels
- No weapon-grade plutonium entering the civilian fuel cycle

Impact on the fuel cycle

Much information regarding amount and quality of plutonium is strictly classified. Without using detailed information on weapon-grade plutonium stockpiles, the question is:

“How much mass of second generation plutonium can be transformed into fresh MOX plutonium quality (Q=65) by mixing one tonne weapon-grade plutonium (Q=95)?”

To answer this, consider comparing the plutonium qualities of reprocessed MOX fuel and weapons-grade plutonium, $Q_{\text{MOX}}$ and $Q_{\text{WPN}}$, with the quality of reprocessed UOX fuel, $Q_{\text{UOX}}$

$$Q_{\text{WPN}} \cdot m_{\text{WPN}} + Q_{\text{MOX}} \cdot m_{\text{MOX}} = Q_{\text{UOX}}(m_{\text{WPN}} + m_{\text{MOX}})$$

Using the given values from Table 1 and assuming that fresh MOX plutonium has a quality of 65%, one achieves:

$$1t \cdot 95 + 58 \cdot m_{\text{MOX}} = 65 \cdot [m_{\text{MOX}} + 1t]$$

$$\Rightarrow \quad m_{\text{MOX}} = 4.29\ t$$

Finally, one tonne of weapon-grade plutonium can convert 4.29 tonnes of second generation plutonium into 5.29 tonnes of useful reactor-grade plutonium. Considering that Russia and the USA want to dispose 68 tonnes of weapon-grade plutonium, [16] 291 tonnes of second generation plutonium could be converted into 359 tonnes of reusable reactor-grade plutonium. This would save more than 67,000 tonnes of natural uranium reserves. [14]

The UK could employ a similar strategy in the multi-reprocessing of the plutonium from their Magnox reactors. Mixing it with low-grade plutonium from France, plutonium can be effectively reused.

**Figure 5: Atoms for peace stamp [24]**

Second generation plutonium coming from civilian reprocessing facilities could be brought to a military facility to be mixed with weapons-grade plutonium to achieve a reactor-grade plutonium mixture. Then the new plutonium composition could be sent to a MOX fabrication facility. This would prevent weapons-grade material from ever entering civilian facilities.
3. Generation IV reactors: Redefining nuclear power

Across Europe, the dominant reactor technology is the LWR. Despite their advantages, they have a number of disadvantages, including inefficient utilization of uranium, production of toxic minor actinides, and risks associated with safety and nuclear proliferation in the fuel cycle. Taking these into consideration, the Generation IV Forum (GIF) set out to redefine nuclear power in the twenty-first century with reactor concepts that (1) minimize waste, (2) resist nuclear proliferation, (3) excel in safety and reliability, and (4) are more economically competitive than existing technologies. [19]

With these goals in mind, GIF proposed six reactor concepts, but instead of discussing every concept, the paper faces two different scenarios, having a wide commercialization of (1) Very High Temperature Reactors, or (2) Sodium-cooled Fast Reactors.

### Very High Temperature Reactors (VHTR)

VHTRs are thermal spectrum reactors that use graphite as a moderator and helium as a coolant. The safety advantages are twofold: first, a meltdown is impossible since graphite only sublimes at 4000°C, and second, accidents from operator error are prevented by low excess reactivity at full power (especially for pebble bed designs) and a strong Doppler-effect, which simplify operation. High temperature operation not only increase electrical generation efficiency, but also opens the possibility of providing heat for industrial processes, making metals, glass and chemicals, including synthetic, carbon neutral fuels for vehicles.

VHTR fuel can reach very high burnup, allowing an open fuel cycle without the need for reprocessing. Furthermore, if a VHTR core is operated at low power density, a $^{232}\text{Th}/^{235}\text{U}$ open fuel cycle can be used, producing negligible amount of minor actinides while exploiting a more abundant element for nuclear power, thorium.

The combination of safety, efficiency, application to industry, and a simple, flexible fuel cycle make VHTR one of the most promising future reactor concepts, and particularly attractive to countries just beginning nuclear power generation, like Poland. [20]

### Sodium-cooled Fast Reactors (SFR)

SFRs are fast spectrum reactors that use molten sodium metal as a coolant. Sodium is one of the best coolants for fast reactors. It does not strongly moderate neutrons, and it can successfully cool the decay heat even when the reactor is without power. Operation temperatures of about 500°C increase the thermodynamic efficiency. The biggest advantages of SFRs comes from their fast neutron spectrum, which enables them to (1) breed depleted uranium into fissile fuel, (2) use and convert low grade plutonium as fuel, (3) transmute actinide waste from other reactors, and (4) vary output based on demand, due to a very low neutron poison effect.

These abilities minimize today’s concerns about nuclear power. SFRs virtually eliminate the need for uranium mining, and when combined with reprocessing allow existing nuclear waste to be used as fuel, producing much shorter lived radiotoxic waste. Nuclear power with SFRs would be much less environmentally impactful and unlimited for the foreseeable future. Their ability to transmute, and to follow the load make them very attractive to countries with large existing nuclear systems, like France. [13] [19]

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Figure 6: Historic trailblazers for Gen IV reactors- (left) The AVR in Jülich, Germany [29], and (right) the Superphenix in Creys-Malville, France [28]
Backend: Waste clustering

Since commercial nuclear power generation began, no satisfying solution for the high-level waste has been decided. Public surveys have shown that the acceptance for nuclear power depends strongly on finding a reasonable solution for the nuclear waste. At first, reprocessing has to be improved while focussing on the development of future transmutation devices. In order to do this, economic improvements, simplifications and more separation possibilities are goals that have to be tackled as soon as possible.

Perspectives

A method that allows separating every different element in the nuclear high level waste at low cost would scale up the benefit of nuclear reactors by orders magnitude. The following waste groups that need to be separated are ranked by decreasing importance:

1. Minor actinides
   These elements are highly radiotoxic and have long half-lives. Removing only the actinides would decrease the necessary storage time by thousands of years. [15]

2. Long-lived fission products
   Although less radiotoxic than actinides, removing and transmuting these elements would also reduce necessary storage time. [15]

3. Rare earth metals
   Even after only a few years of storage, spent fuel contains a number of stable isotopes of extremely valuable metals, including for instance ruthenium, rhodium and cerium. Extracting these could reduce concerns over the rarity of these materials as well as be a profitable venture.

In this chapter, the focus is on all the elements that are vitrified in the PUREX process. It is difficult to separate single elements from others with similar chemical properties, but it is much less difficult to split all elements with similar characteristics to form a cluster.

Minor actinide cluster

For instance, the separation of 3+-charged actinides from a number of lanthanides does not find economic justification. Whereas the separation of one cluster containing lanthanides and minor actinides would be much cheaper. Even though clustering does not find economic justification yet, but it would be a relatively low volume storage that could be processed when technical development will have been improved. Right now, there are many research programs going on that may bring the great improvement in the future. Chemical separation, fuel fabrication and transmutation reactors are the major challenges to implement for the destruction of minor actinides. Vitrification is irreversible, but reprocessing is modifiable. Therefore, we should start to trust in future development and put non-vitrified minor actinide-waster-clusters in intermediate storage. Maybe pyroprocessing and 14 MeV neutrons coming from nuclear fusion will once make transmutation to an easy business.

Heat cluster: separation of Cs and Sr

Caesium and Strontium are abundant fission products. In the first 120 years, they dominate the heat contribution in high level waste which complicates its handling and geologic disposal. Fortunately, their most active and toxic isotopes, $^{137}$Cs and $^{90}$Sr, have half-lives of about 30 years, and so they decay rapidly. [15] In aqueous solution, caesium and strontium prefer oxidation states 1+ and 2+, and therefore they differ in separation characteristics from actinides and lanthanides. This allows relatively easy separation from the high level waste which has also been considered in the American UREX process. [15]
The benefit of forming a Cs/Sr-cluster is to simplify handling of the remaining waste and to create a cluster of strong heating with “short term danger”.

Despite good scientific reasoning behind interim oceanic storage, it is unlikely that the current public perception of nuclear waste will allow such storage. The challenge is to educate the public on nuclear science and enable political discussion on the topic.

**Extraction of rare earth elements**

Since some decades, rare earth elements are ones of the most desired metals on earth. They find various approaches, especially in modern electronic devices. Instead of doing environmental impacting mining, one can consider extracting these elements from the high-level nuclear waste where some of them occur in relatively high abundance and without having long-term radiotoxicity. [15] [22]

Realistically, as long as no strategy for economical minor actinide isolation has been developed, there is no justification for considering selective rare earth element extraction. The difficulties in separating the minor actinides prove that a beneficial extraction of single rare earth elements is definitely utopia for the nuclear fuel cycle in 2040.

**The economic issue**

Literature shows that much more separation procedures are possible than are actually used. Economic benefit remains the single largest obstacle. For example, using the PUREX process to separate americium and curium is considered to be too expensive, so they are left in the fission product mixture for vitrification. [15]

Finally, the ultimate goal of complete waste separation has to be set in the far future. In the near future, simple improvements to existing processes should be implemented while focusing on making the backend of the nuclear fuel cycle cleaner and cheaper than it is today. As a first step, waste-clustering can be a cost effective strategy to follow.
5. Conclusion

In 2016, nuclear power has proven to be a robust, scalable, and cost effective low-carbon energy source widely believed to be an essential part of the climate change solution. Although the current fuel cycle leaves much to be desired, there are many feasible solutions to these problems.

At the frontend, re-enrichment of depleted tails and reprocessed uranium is expected to be a viable alternative source of uranium which could provide fuel at low costs and low environmental impact. With more development, especially in the area of mitigation cost, extraction of uranium from seawater could provide a virtually infinite source of uranium. Lastly, through intelligent reuse of plutonium both spent nuclear fuel and obsolete weapons programs, the stockpiles of both can be reduced while providing safe, abundant, carbon free energy to people across Europe.

Newer Generation IV reactors build on the benefits of an enhanced frontend by improving the safety, efficiency, and economics of nuclear generation. Some countries are currently focusing on developing VHTRs, which offer much higher efficiencies paired with unparalleled safety advantages. France, especially, continues to pursue SFR technology, which, in addition to high thermodynamic efficiency, offers the ability to transmute the long-lived isotopes present in the nuclear waste. Even when the cost and benefits of the two reactors are very different, both are enormous improvements over the current technology and the substantially improve of the nuclear fuel cycle.

Likely the most important improvements are at the backend of the fuel cycle. Simply burying the plutonium and minor actinides is not only wasteful, but it necessitates hundreds of millennia of storage, which is a major reason for public being against nuclear power. The best strategy for the next decades is to simplify and enhance the current liquid-liquid separation methods to overcome economic objections to reprocessing. Following this, further strategies can be developed to handle individual waste clusters.

Although there are many solutions available to improve the nuclear fuel cycle, the single largest obstacle to overcome is neither scientific nor engineering related- it is public opinion. Although nuclear power is the only low-carbon technology that can generate baseload power on a large scale, a segment of the public has vocally opposed its use. Nowhere is this clearer than in Germany, which has formally outlawed nuclear power generation, much to the dismay of many scientists, environmentalists, and the nuclear supporters. Much of this opposition comes from irrational fear which stems from an ignorance to understand. Unlike coal, wind or solar energy, nuclear power is abstract and very difficult for a layperson to understand. By educating the public about nuclear science and the promising solutions it offers, public support can be won, and a sustainable nuclear fuel cycle achieved by 2040.
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