Eur. Phys. J. Plus (2019) **134**: 601
DOI 10.1140/epjp/i2019-13014-4

**THE EUROPEAN PHYSICAL JOURNAL PLUS**

Regular Article

# Extension of the "42-0" approach to ternary fuels and application to a thorium-fed, minor-actinides-burner ADS[⋆]

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Received: 27 May 2019 / Revised: 8 August 2019

*In memory of Prof. Carlo Artioli, for his insatiable hunger*
*for achieving a rational understanding of the surrounding world,*
*from which the original "42-0" concept was generated.*

**Abstract.** When dealing with an accelerator-driven system (ADS), fuel cycle performances are a main element to be considered in the design. The higher cost of these systems, relative to critical fast reactors, makes indeed their justification sustainable only in a perspective of serving fuel and waste management strategies. Disposing of a logical scheme, and an analytical frame, for driving the definition of the core configuration so as to target by design the desired performances, might therefore be a cornerstone for fully exploiting the potential of an ADS. In this work, such a logical scheme, originally developed for the case of an ADS fueled with a binary mixture of plutonium and minor actinides, to identify the optimal design of a proliferation resistant core for the transmutation of the minor actinides, is extended to the more general case of ternary fuels —encompassing almost all cases of practical interest. The rationale for the approach is presented and justified, and analytical considerations added for quantitative estimates to be possible. Finally, a practical example of application is presented to a specific case, with rather general driving criteria to show the potential of the presented approach.

## 1 Introduction

This paper takes the move from the original "42-0" approach [1], conceived by Prof. Artioli to rationalize the design of an accelerator-driven system (ADS) so as to target by design *a priori* defined fuel cycle performances.

Specifically, the name recalls the burnup figures (in $\mathrm{kg/TWh_{th}}$) for minor actinides (MAs) and plutonium, respectively, in the optimal operational point identified for the European Facility for Industrial Transmutation (EFIT) [2], an uranium-free (thus plutonium-sustained) sub-critical ADS meant for burning legacy MAs recovered by spent light water reactor (LWR) fuel.

The core of that original work was to figure out which are the relevant quantities under time evolution, provided that there is the trivial —but previously unrecognized— constraint that the algebraic sum of all the contributions must yield a fixed amount. Generally speaking, indeed, whatever the composition of the fuel, the net removal of actinides due to fission can be accounted at a fixed rate, only depending from the average energy that can be recovered per fission event. For example, in the EFIT case, the net burnup normalization was to $\approx 42\,\mathrm{kg/TWh_{th}}$ (directly resulting from the $\approx 210\,\mathrm{MeV/fission}$ of plutonium). The "apparent" removal of actinides, instead, is due to the combined effect of fission —which, as mentioned, determines a net loss— and transmutation, which instead has net zero losses in the fuel system overall. This can be visualized in the scheme of table 1 for a binary fuel.

Under this premise, the specific operational point "42-0" identified for EFIT was indeed the one providing the highest MAs burning rate while not burning nor breeding plutonium.

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[⋆] Focus Point on "Advances in the Physics and Thermohydraulics of Nuclear Reactors" edited by J. Ongena.

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**Table 1.** Removal rates for the species of a binary fuel.

| Removal rates | Species A | Species B | Total |
|---|---|---|---|
| Fission [kg/TWh$_{\mathrm{th}}$] | $-x$ | $-y$ | $-42$ |
| Transmutation [kg/TWh$_{\mathrm{th}}$] | $+l$ | $-l$ | 0 |
| Total [kg/TWh$_{\mathrm{th}}$] | $-x+l$ | $-y-l$ | $-42$ |

**Table 2.** Removal rates for the species of a ternary fuel.

| Removal rates | Species A | Species B | Species C | Total |
|---|---|---|---|---|
| Fission [kg/TWh$_{\mathrm{th}}$] | $-x$ | $-y$ | $-z$ | $-42$ |
| Transmutation [kg/TWh$_{\mathrm{th}}$] | $\pm l$ | $\pm m$ | $\pm n$ | 0 |
| Total [kg/TWh$_{\mathrm{th}}$] | $-x \pm l$ | $-y \pm m$ | $-z \pm n$ | $-42$ |

In fact:

- any lower MAs burning rate (*e.g.*, 34 kg/TWh$_{\mathrm{th}}$) would imply the complement to 42 to be imputed to plutonium, whose burning in an ADS —more expensive than any other critical reactor of the same type— is not economic;
- any higher MAs burning rate (*e.g.*, 65 kg/TWh$_{\mathrm{th}}$) would just masquerade as fission the amount of losses exceeding 42, the difference being instead removed by transmutation into plutonium, which is in contrast with the non-proliferation aim implied by the choice of an uranium-free fuel.

Starting from this consideration, defining beforehand the target fuel cycle performances sought for EFIT, a comprehensive approach was elaborated detailing how the design of the core would had to be set so to ensure fulfilling the target. Indeed, despite the actual evolution of the isotopic composition of the fuel is ruled by the Bateman's equations [3], when focusing on plutonium and MAs as whole, the effective removal channels can be identified in fission (net loss) and mutual transmutation (no net loss, conversion of one species into the other). The associated removal rates are functions of the respective atom densities and effective cross-sections, the latter depending on the neutron spectrum in the fuel. In any reactor design, although the spectrum is mostly set by the materials used (*e.g.*, the fuel type: oxide, metallic, nitride, etc.; or the coolant: water, sodium, lead, etc.), it is possible to pursue a fine tuning by adjusting the volume fractions in the core, with a two-fold effect on the removal rates:

- a direct one, through the change of the effective removal cross-sections;
- an indirect one, through the change of all cross-sections together, requiring an adjustment of the atom densities of the two species to bring the reactor to the required criticality (or sub-criticality, in the specific case of an ADS) level.

The key point, therefore, is identifying the atom fractions in the fuel, so that the combined effect of fission losses on both species and net transmutation among the latter can result in the target MAs removal.

The chance for this work to extend further the "42-0" approach was suggested by the objective of a collaborative research project promoted by the International Atomic Energy Agency (IAEA) to investigate the viability of a low enriched uranium (LEU, *i.e.*, with $^{235}$U enrichment below 20%) ADS, and its capabilities to serve as a demonstrator for a thorium-uranium cycle or as a MAs burner. The main question we tried to address is whether these two apparently alternative objectives could have been achieved at the same time, and how to enlarge the scope of the "42-0" approach to such an use.

## 2 Rationalization of the problem

A first extension of the "42-0" approach to ternary fuels was performed dealing with critical reactors in the so called "adiabatic[1] reactor" model [4]. As pointed out in the previous section, the very first and general idea of the "42-0" approach is how the system should handle out its 42. In the case of the adiabatic reactor, once more the attention was brought on developing the algebra of 42 to identify suitable design decisions implementing *a priori* fixed fuel cycle burnup figures. In fact, also in a ternary fuel the depletion rate of $\approx 42$ kg/TW$_{\mathrm{th}}$ holds, but with all three species concurring to it. In parallel, mutual transmutations occur, again contributing to an overall net zero balance, as shown in table 2.

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[1] Adiabatic here is used to indicate that the reactor, in its whole fuel cycle, has no net exchange with the outside environment of any relevant materials, which are instead preserved into the system.

In the specific case investigated in [4], a fast reactor was conceived to be self-sustaining in plutonium (species B) and capable to burn its own produced MAs (species C), *i.e.*, a reactor where the removal and production rates of both plutonium and MAs are exactly balanced, maintaining their net amount stable in a fuel "equilibrium". According to the symbols in table 2, it means $-y \pm m = 0$ and $-z \pm n = 0$. In that work, also the impact of real operation on the equilibrium was considered, including the effects of fuel cooling after irradiation, of reprocessing and of refabrication.

When dealing with nuclear fuels in a cycle perspective, clear roles are to be appointed to all the species involved. Notably, the key roles that can always be anticipated in fuel for a fast-spectrum reactor (be it critical or sub-critical) are:

- the fissile one, which is the main contributor to sustaining the fission chain reaction; and
- the fertile one, which is the main responsible for the formation of new fissile in the fuel.

Alongside with these, if sustainability considerations are taken into account for the fast-spectrum reactor to also serve fuel management policies, a waste can be identified, which is the component of the fuel meant for removal.

Analyzing the cases already investigated in the past, it can be easily seen that, for the adiabatic reactor, plutonium is the fissile, MAs are the waste and uranium is the fertile; in the binary fuel of the EFIT case, instead, while plutonium maintains its clear role of fissile, MAs act both as waste and as fertile. In this latter case, while the role of waste for MAs is implicit in the objective sought for EFIT, their fertile role comes from their transmutation into plutonium, allowing the regeneration of the latter which otherwise would have been lost by fission.

The first fundamental consideration to achieve a rational understanding of the problem is the role of reintegration during the out-of-pile stages of the fuel cycle. During irradiation, the fuel undergoing fission is depleted at the well understood rate of $\approx 42\,\mathrm{kg/TWh_{th}}$. Once achieved the target burn-up, it is then unloaded from the core, cooled down and reprocessed to separate the fission products. During the successive refabrication phase, it has to be reintegrated by an amount exactly compensating the net losses (which, in a more realistic view, also include losses during the reprocessing phase). The composition of the feed is in fact what determines the system to continue operating with the desired performances.

In the adiabatic case, within the reactor the production of MAs from plutonium is exactly compensated by MAs removal due to their fissioning and to their transmutation back into plutonium; at the same time, the losses of plutonium by fission are exactly compensated by breeding from uranium. Overall, all depletions are appointed to the sole uranium, so that —for the next loading to repeat the same operation— the feed has to be made of uranium only. Should instead the feed comprise only MAs, the reactor would progressively shift to the same operating cycle of EFIT: the excess of loaded MAs (with respect to the amount removed) would be only partially compensated by their increased fission, while higher transmutation to plutonium would appear, substituting its breeding from uranium (lesser and lesser effective as the total amount of uranium in the fuel tends to disappear, being not fed anymore).

Coming to the specific case proposed for study in the IAEA's collaborative research project, roles are to be defined for all the species predicted in the fuel by analyzing the purpose and desired goals meant for the system.

First of all, the context of a LEU ADS suggested to not pursue any breeding of plutonium, as it is not already loaded into the core, the fissile role being initially played by $^{235}$U. Then, as anticipated at the end of the previous Section, the will to prove that the two objectives proposed in the project are not necessarily alternative, but can be simultaneously achieved, brought to the decision of having the system operating in the thorium-uranium cycle while burning MAs. It is worth anticipating that the loading of MAs (required to burn them) inevitably implies the formation of plutonium, which is in contradiction with the first consideration. To conciliate this, the formation of plutonium is to be considered only as a by-product not to be exploited in a fuel cycle perspective.

From these considerations, the roles of the fuel species, at equilibrium, appear evident as follows:

- thorium is anticipated to be the fertile;
- $^{233}$U is accordingly the reference fissile;
- MAs are explicitly required to be the waste, along with plutonium according to the will to avoid proliferation issues, so that collectively all transuranics (TRU) are waste.

As seen, no specific role is assigned to LEU since, for the desired operation, the fuel is requested to evolve into a system made of the above species only. In the proposed investigation, LEU is therefore only used for the initial startup of the reactor.

Now that the three species have been identified in the fuel, it is possible to extent the "42-0" approach to the case of such ternary fuel. Before doing so, it is useful to recall all possible transmutation trajectories that can happen in the fuel:

- thorium can transmute into uranium (specifically, to $^{233}$U);
- uranium (specifically, $^{238}$U) can transmute into plutonium, while some minor isotopes can transmute into thorium;
- plutonium can transmute backwords to uranium, but mostly forward into MAs;
- MAs can transmute into plutonium.

| **Scenario** | 1 | (intermediate) | 2 |
|---|---|---|---|
| **Start-up cycle** | LEU + Th | LEU + Th + MAs | LEU + MAs |
| **Feed** | Th | *x*%Th + (100-*x*)%MAs | MAs |
| **Equilibrium cycle** | Th-U | Mixed | TRU (Pu-MAs) |

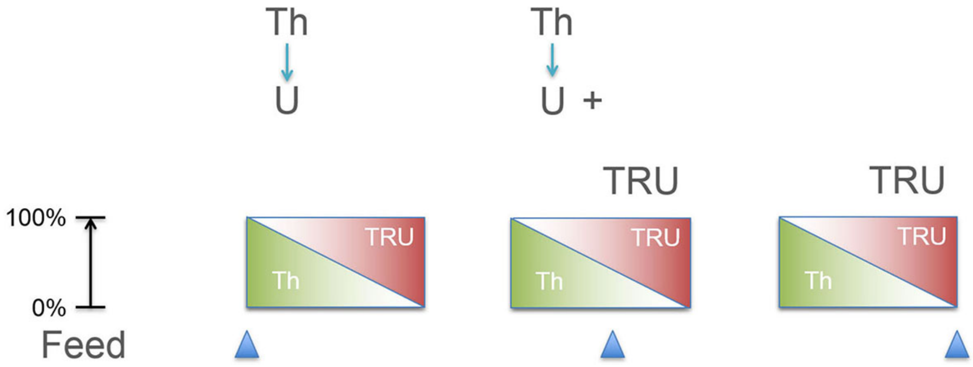

**Fig. 1.** Graphical representation of the connection between feed and equilibrium chains; scenario 1 is due to a pure thorium feed, scenario 2 follows a pure MAs feed and the intermediate scenario derives from a mixed thorium-MAs feed.

Considering now that the back-transmutations of plutonium into uranium and of uranium into thorium are minor phenomena, the whole picture can be represented by two chains that contribute, in general terms, to the equilibrium core: the transmutation of thorium into $^{233}$U and the mutual transmutation between plutonium and MAs.

If therefore one neglects the initial loading of LEU (which, being not fed anymore, is destined to disappear from the system), it appears evident how these two chains are fairly independent. It is also of no relevance to discriminate the mutual transmutations of plutonium and MAs, as both are accounted for together as waste. Using the nomenclature of table 2, and assuming thorium, uranium and TRU as species A, B and C, respectively, it results:

$$\begin{aligned} l &= -m, \\ n &= 0, \end{aligned} \tag{1}$$

the disconnection between these two chains being reflected by the value 0 for the transmutation balance of TRU, $n$.

The coexistence of these two chains can be practically ruled by setting the composition of the feed vector as depicted in fig. 1. Keeping in mind the normalizer for removal, thus for reintegration, as $\approx 42\,\mathrm{kg/TWh_{th}}$, the two extreme cases —a feed made only of thorium, leading to a pure thorium-uranium cycle (scenario 1), or a feed made only by TRU (and more specifically, MAs, aiming to a proliferation-resistant fuel cycle), leading to a pure plutonium-MAs cycle (scenario 2)— can be attained by feeding the core at reprocessing with only thorium or MAs, respectively. Any intermediate feed composition will make the two chains to coexist, and with relative importances which are proportional to the fractions of thorium and MAs in the feed (intermediate scenario).

### 2.1 Visual representation of ternary fuels

A useful tool for the visualization of ternary fuels comes from triangular plots. As is shown in fig. 2, the sides of the triangle are associated with axes, each indicating the fractional content of the respective species in the fuel, so that any point in the graph can unequivocally represent a fuel composition. In the case being discussed in this work, the selected species are indeed thorium, uranium and TRU.

Assuming as reference the fuel composition at core loading, the two extreme cases discussed above can be visualized on the plot with two points, lying on the Th-axis when no TRU (MAs) are loaded as feed (orange star in fig. 3), or on the U-axis when no Th is fed (green star in fig. 3). Any intermediate composition will bring the reactor to operate, at equilibrium, with a fuel composition lying on the red line of fig. 3 which connects the two extreme points.

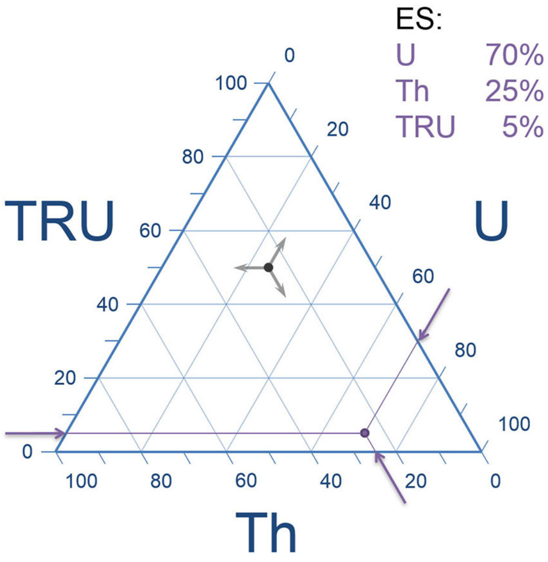

**Fig. 2.** Use of triangular plot to unequivocally identify fuel compositions in %.

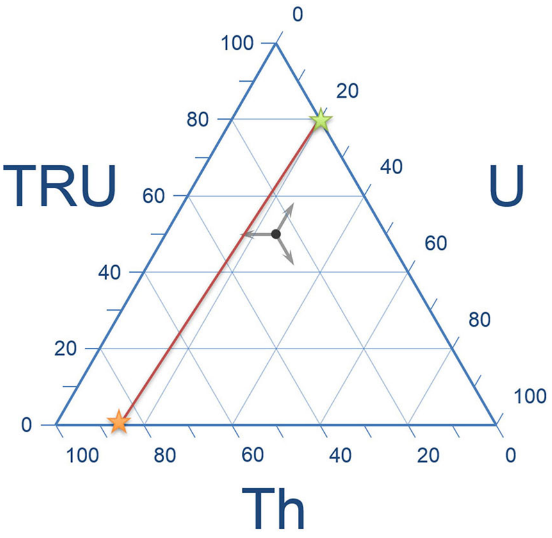

**Fig. 3.** Visualization on a triangular plot of the equilibrium compositions of a LEU ADS fuel with feed made by any combination of Th and TRU.

## 2.2 Analytical formulation of the problem

Borrowing the same approach presented in [4], the qualitative considerations above for the fuel equilibrium can be formally cast in the analytical problem of eq. (2)

$$\vec{N}(t_O + t_D + t_R + t_F) = \boldsymbol{R}\,\boldsymbol{A}_D\,\boldsymbol{A}_O\vec{N}(0) + \vec{F}, \tag{2}$$

where $\vec{N}(t)$ is the vector of isotopic concentrations in the fuel at time $t$, $\boldsymbol{A}$ and $\boldsymbol{R}$ are the transformation and reprocessing operators, respectively, $\vec{F}$ is the vector of isotopic composition of the feed, and the $_O$, $_D$, $_R$ and $_F$ subscripts indicate operation, decay, reprocessing and fabrication phases, respectively.

The elements of the transformation operators $\boldsymbol{A}_D$ and $\boldsymbol{A}_O$ are defined from the Bateman's equations as

$$A_{D,ij} = \begin{cases} -\lambda_i & i = j, \\ \lambda_{j\to i} & i \neq j, \end{cases} \tag{3a}$$

$$A_{O,ij} = \begin{cases} -\sigma_i f \phi & i = j, \\ \sigma_{j\to i} f \phi & i \neq j, \end{cases} \tag{3b}$$

where $\lambda_i$ is the decay constant of isotope $i$, $\lambda_{j\to i}$ is the decay constant of isotope $j$ into isotope $i$, $\sigma_i$ is the removal cross-section of isotope $i$, $\sigma_{j\to i}$ is the transmutation cross-section of isotope $j$ into isotope $i$, $f$ is the availability factor of the reactor and $\phi$ is the neutron flux. The reprocessing operator is a diagonal matrix with elements equaling the reprocessing recovery efficiencies for each isotope.

From the system of eqs. (2) the net balances per species of interest can be summarized as

$$\varDelta_{TRU} = -z := \sum_{i\,\mathrm{in}\,TRU} F_i - \sum_{i\,\mathrm{in}\,TRU} \sigma_i f \phi, \tag{4a}$$

$$\varDelta_{Th} = -x + l := \sum_{i\,\mathrm{in}\,Th} F_i - \sum_{i\,\mathrm{in}\,Th} \sigma_i f \phi. \tag{4b}$$

## 3 Application to a MAs burner ADS operating in the thorium-uranium cycle

Once perimetered the domain of the problem, with all principles understood, the target performance sought for the system are to be defined to fix the equilibrium core. In the specific case, as previously mentioned, the sought ADS is required to be capable of burning MAs while self-sustaining in $^{233}$U. As seen before, a multitude of different performances can be attained, all targeting —to different extents— both objectives. Several criteria might be used to select the most appropriate configuration, *e.g.*, a well-defined MAs transmutation rate. In this case, however, as additional criterion it is chosen to fix the power rating for the reactor to $400\,\mathrm{MW_{th}}$. The target power rating, in fact, corresponds to a target core size (under the assumption of an almost fixed power density, being this dictated by technological constraints): therefore, for the core to achieve the required sub-criticality, a specific fissile content is required, maintaining which imposes a precise fertile content in the feed, hence its counterpart of waste and, in turn, the MAs burning performances.

The power size is chosen as for EFIT, so that this study can be applied as an investigation of alternative operations for that system. From EFIT it is also borrowed the same sub-criticality target, as well as information on the fuel reactivity, to be used as reference to identify the fissile content at the equilibrium for the new configuration here investigated. Since, as explained in [4] and as visible from eq. (2), the equilibrium concentration for a given reactor (*i.e.*, a given spectrum) solely depends on the feed composition, establishing requirements for the former implies a selection of the latter. Therefore, by solving eq. (2) for the feed $\vec{F}$ with the target fissile content at equilibrium, the results in table 3 are retrieved, where it can be seen that the thorium fraction has to be fixed at 60%, the remaining portion being TRU. With this feed and by solving eq. (2) for $\vec{N}(t)$, at equilibrium the composition of the fuel results of 62.5% thorium, 14.7% uranium (out of which 7.2% is $^{233}$U) and 22.8% TRU (about half of which is plutonium). Given the fraction of TRU in the feed, the MAs burning rate at equilibrium ($\varDelta_{TRU}$ in sect. 2.2) is anticipated to be $\approx -16.8\,\mathrm{kg/TWh_{th}}$, *i.e.*, 40% of the normalizing $42\,\mathrm{kg/TWh_{th}}$.

The last element of the design approach concerns the start-up core. Depending on the resulting equilibrium configuration, the corresponding fuel might be unavailable —or impracticable, or unfeasible. In the present case, to manufacture the equilibrium fuel for the initial start-up, $^{233}$U should be available, which may not be the case; also, plutonium should be included, conflicting with the general aim of a proliferation resistant system. Therefore, it is necessary to foresee the system transitioning from a first core to the equilibrium one. For the selection of the start-up core, several approaches can be followed, depending on the aimed objective. As an example, the minimization of the current range required to the accelerator, to sustain the system all along evolution to equilibrium requires the reactivities of the start-up and equilibrium fuels —and possibly also that of all transition compositions— to be similar. Whatever the driving consideration, and the corresponding composition of the start-up fuel, if the successive loadings are prepared with the equilibrium feed, the composition will unequivocally evolve towards the same final point, as shown in fig. 4.

The consideration of using a fixed feed, so to guarantee the equilibrium core meets desired performances, suggested to adopt as driving criterion for the selection of the start-up fuel to have constant MAs transmutation rates ($-16.8\,\mathrm{kg/TWh_{th}}$) while relying on LEU, according to the original mission of the IAEA's research project. The visualization of the fuel composition trajectory during evolution from the start-up core to the equilibrium one in the final case of this study is shown in fig. 5.

**Table 3.** Feed composition for the MAs burner ADS in the Th-U cycle.

| Component | Isotope | $N_i$ [at.%] |
|---|---|---|
| Th | Th232 | 60.000 |
| TRU | Np237 | 1.582 |
| | Np238 | 0.000 |
| | Np239 | 0.000 |
| | Am241 | 30.242 |
| | Am242m | 0.101 |
| | Am243 | 6.377 |
| | Cm242 | 0.000 |
| | Cm243 | 0.026 |
| | Cm244 | 1.187 |
| | Cm245 | 0.449 |
| | Cm246 | 0.035 |
| | Cm247 | 0.001 |
| | Cm248 | 0.000 |

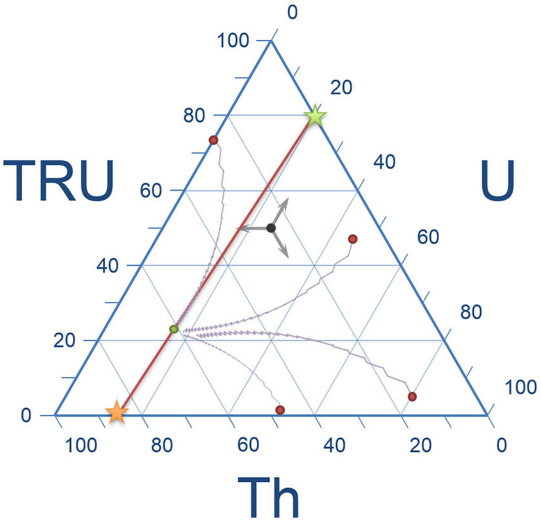

**Fig. 4.** Visualization on a triangular plot of the chosen equilibrium composition (green dot on the red line), and of possible evolutions of the start-up fuel towards the final one (violet paths, each relative to a different start-up fuel composition).

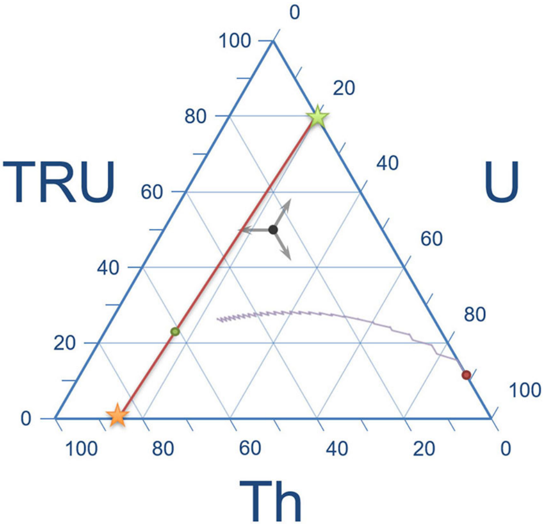

**Fig. 5.** Visualization on a triangular plot of the anticipated trajectory describing the evolution of the selected start-up fuel into the desired equilibrium one.

## 4 Conclusions

In this paper, the original "42-0" approach for the design of an ADS fed with a binary plutonium-MAs fuel so to target *a priori* defined fuel cycle performances, has been analyzed for its extension to ternary fuels. The general considerations have also been related to the analytical problem allowing the quantitative assessment of transmutation rates anticipated for the equilibrium fuel.

The approach, already appreciated in its early formulations, although partial in scope as they were developed for very specific cases (nominally, a binary-fueled ADS and an "adiabatic" critical reactor), confirmed its flexibility and accuracy also when applied to the extended case of a LEU-fueled ADS, for which it was possible to devise operation in the thorium-uranium cycle while providing MAs burning capabilities.

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