

Development of a stress analysis code for TRISO particles in HTRs

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Abstract

The PUMA project is a Specific Targeted Research Project of the European Union EURATOM 6th Framework Program. Complementary with other initiatives, the PUMA project is mainly aimed at providing additional key elements for the utilisation and transmutation of plutonium and minor actinides in high temperature gas-cooled reactor designs. Within PUMA, new coated particle designs for Pu/MA incineration will be explored. For this purpose, Delft University of Technology (TU-Delft) participates in the optimisation of the fuel design of HTRs fuelled with plutonium and/or minor actinides by calculating the failure fraction of TRISO particles during irradiation in a given reactor core design. This is achieved by coupling a neutronics, thermal-hydraulics and fuel depletion code system, which delivers the fuel temperature, fast neutron flux and power density profiles, to an in-house developed stress analysis code. The latter is being further developed to provide a reliable and realistic failure fraction. Application to a PBMR-400 design fuelled with 1st generation Plutonium and with a target burn-up of 700 MWd/kgHM shows promising results in terms of Pu-burning capabilities. The TRISO particle failure fraction is also calculated and compared to U-based fuel particles. It is shown that the Pu-based fuel particles need a better design and this can be achieved by use of the present stress analysis code.

1 Introduction and objectives of the PUMA project

The sustainability of the nuclear fuel cycle and the reduction of plutonium (Pu) and Minor Actinides (MA) stockpiles are key issues in the definition of the future nuclear energy mix in Europe. The High Temperature gas-cooled Reactor (HTR) can incinerate both Pu and MA due to its unique and unsurpassed safety features.

Apart from the inherent safety features offered by this reactor type, the nature of the coated particle fuel offers a number of attractive features. In particular, it can withstand burn-ups far beyond that in either LWR or fast reactor systems, as demonstrated in former tests (Alberstein, 1994).

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Within PUMA, new coated particle designs for Pu/MA incineration will be explored. For this purpose, helium and swelling behaviour models for coated Pu and MA particle fuels together with fuel performance models, appropriate to HTR fuels, need to be further developed. Finally, the impact of such an utilisation of the HTR on economics and the entire fuel cycle will be assessed.

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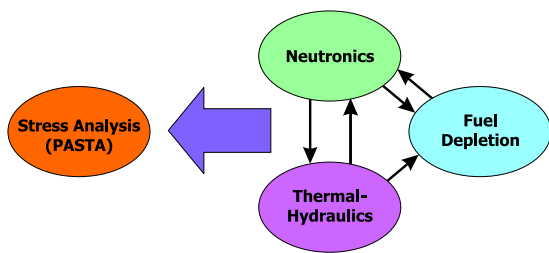


Fig. 1: Fuel performance model overview.

The project consortium consists of 15 organisations from 8 European countries and 1 from the USA. The paper presents the activities and achievements of Delft University of Technology (TU-Delft) in the PUMA project.

2 Optimisation of Pu/MA loaded V/HTR fuel design

TU-Delft participates in the optimisation of the fuel design of HTRs fuelled with plutonium and/or minor actinides. To this end, the failure fraction of TRISO coated fuel particles is calculated during irradiation by coupling a neutronics, fuel depletion and thermal-hydraulics code system (Boer et al., 2008a), which will deliver the fuel temperature, burn-up and power density profiles, to an in-house developed stress analysis code called PASTA (PARTicle STress Analysis) (Boer et al., 2008b). The calculation scheme is depicted in Fig. 1. In the following, all calculations are limited to the case of the pebble-bed reactor core, although calculations for other core geometries are also possible.

The PASTA code describes the mechanical behavior of TRISO particles during irradiation and aims at calculating the failure probability of coated particle fuel. PASTA is a one-dimensional analytical and multi-layer model that takes into account the visco-elastic behavior of the coating layers and the surrounding graphite during irradiation. The main source of stress in all layers is due to the pressure build-up from the gaseous fission products in the buffer layer. Moreover, the Pyrocarbon layers exhibit irradiation-induced dimensional changes and creep

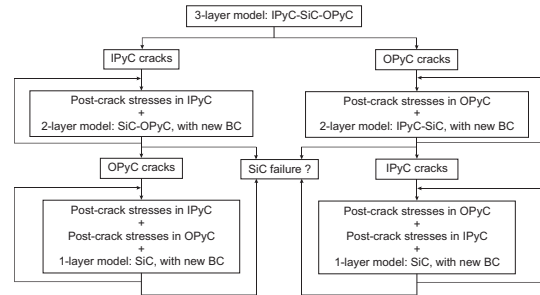


Fig. 2: Description of the crack-induced failure model.

(in the radial and tangential directions). Finally, the model allows thermal expansion of all layers.

Once all equilibrium profiles have been calculated for normal operating conditions in a given reactor core, the particle failure fraction during the lifetime of the pebbles can be calculated for several design parameters of the TRISO particles and fuel pebbles. For instance, the effect of varying the fuel kernel size and the thicknesses of the various coating layers surrounding the fuel kernel, the actinide density in the fuel kernel, the concentration of TRISO fuel particles in a pebble, and the pebble flow velocity in the reactor core can be investigated.

A first and necessary step in this challenging task consists in improving the PASTA code for an even more reliable and realistic modelling of failure mechanisms (including the effect of PyC cracking and a more accurate model of fission gas production and release) and in including specific mechanisms to the use of plutonium and minor actinides in the fuel kernel, such as the contribution of helium in the buffer pressure.

2.1 Effects of PyC cracking on SiC layer behavior

The SiC layer provides retention of all fission products under normal operating conditions. It acts as a pressure vessel and is modeled as such in many codes (see for instance in (Sawa et al., 1996)). When the stress in the SiC layer is tensile and high enough, the layer is said to fail due to over-pressure. Recently, Wang and Ballinger (2004) proposed a model in which PyC layers can fail first (due to over-pressure), creating a radial crack. This results in an increase in the local stress at the PyC/SiC interface and produces

Table 1: Distribution parameters.

Property	PuO _{1.7}		UO ₂	
	Mean value	Standard deviation	Mean value	Standard deviation
Kernel diameter (μm)	200 ^a	10	500 ^b	10
Buffer thickness (μm)	90 ^a	5	95 ^b	5
iPyC thickness (μm)	40 ^a	5	40 ^b	5
SiC thickness (μm)	35 ^a	5	35 ^b	5
iPyC thickness (μm)	40 ^a	5	40 ^b	5
iPyC characteristic strength (MPa.m ^{3/β})	24 ^c	9.5 ^c	24 ^c	9.5 ^c
SiC characteristic strength (MPa.m ^{3/β})	9.64 ^c	6.0 ^c	9.64 ^c	6.0 ^c
oPyC characteristic strength (MPa.m ^{3/β})	24 ^c	9.5 ^c	24 ^c	9.5 ^c

^aTaken from Alberstein (1994)

^bUsual values for U-fuel TRISO particles (Boer et al., 2008b)

^cTaken from Wang et al. (2004)

a local crack tip stress intensity K_I^{SiC} .

If the value of K_I^{SiC} exceeds the fracture toughness (K_{IC}) of the SiC layer then the layer fails even when the far field stress in the SiC is compressive. The model was called the "crack-induced failure model" by its authors. For instance, the calculation of the mode I stress intensity factor in the SiC layer due to a cracked iPyC layer reads:

$$K_I^{SiC} = K_I^{iPyC}(t) \sqrt{\frac{d}{a_{iPyC}}} + \bar{\sigma}_{SiC}(t) \sqrt{\pi d} \quad (1)$$

In Eq. 1, d is the SiC grain diameter, a_{iPyC} is the crack length in the iPyC layer, $\bar{\sigma}_{SiC}(t)$ is average circumferential stress in the SiC layer, and $K_I^{iPyC}(t)$ is the Mode I stress intensity factor in the radially cracked iPyC layer.

The crack-induced failure model proposed by Wang and Ballinger (2004) has been implemented in the PASTA code and is used to assess the effect of a PyC layer failure on the TRISO particle failure fraction during irradiation. By considering that both iPyC and oPyC layers can fail, many failure scenarios must be taken into account, as shown in Fig. 2.

2.2 Stable fission gases and Helium release in Pu/MA HTR fuels

The release-to-birth ratio of stable (long-lived) fission gases from the kernel is calculated by a numerical method, which solves the diffusion equation

from an equivalent sphere of radius a :

$$\frac{\partial C}{\partial t} = \frac{D}{r^2} \left[\frac{\partial}{\partial r} \left(r^2 \frac{\partial C}{\partial r} \right) \right] + \beta \quad (2)$$

where C is the fission gas concentration ($atoms.m^{-3}$), D the atom diffusion coefficient ($m^2.s^{-1}$), β the stable fission gas production rate ($atoms.m^{-3}.s^{-1}$). Although the numerical method allows non-zero boundary conditions and is very accurate, the computation time can be an issue (see next section). An alternative solution is based on an analytical method provided by Kidson (1980), which is especially adapted to non-constant conditions (multiple cycles):

$$F^k(t) = 1 - \sum_{n=1}^{\infty} \left[\frac{6a^2}{n^4 \pi^4} \times \sum_{s=1}^k \frac{\beta^s}{D^s} (1 - \exp(-\phi^s)) \right. \\ \left. \times \exp(-\Delta\Phi^{ks}) \right] \times \left[\sum_{s=1}^k (\beta t)^s \right]^{-1} \quad (3)$$

where $F^k(t)$ is the release-to-birth ratio of fission gases at cycle k , $\phi = n^2 \pi^2 (Dt/a^2)$ and $\Delta\Phi^{ks} = \sum_{r=s}^k \phi^r$. In the latter equations, the superscripts k , s and r are not an exponent but refer to the cycle number. The last summation in Eq. 3 is simply the total gas atom production during all k cycles. In practice, n is chosen to provide both fast calculations and good accuracy.

A very cheap option in computation time consists of an approximate solution that was deduced from the well-known Booth solution in constant irradiation conditions and extended in the PASTA code to cyclic

irradiation history for the cumulative release f :

$$f = 4 \left(\frac{Dt}{\pi a^2} \right)^{1/2} - \frac{3}{2} \left(\frac{Dt}{a^2} \right) \quad (4)$$

In Eq. 4, the (Dt) terms contain the temperature dependence of the irradiation history and are simply accumulated from one cycle to another. Eqs. 3 and 4 are used in a sensitivity analysis in order to determine the effect of the buffer pressure on the final failure fraction.

The calculation of the buffer pressure also includes the contribution of helium. As a first step, it is assumed that 100% helium is released at the considered kernel temperature. This assumption is conservative although the production of helium in Pu-based fuel is still one order of magnitude less than stable xenon and krypton, as calculated with the present code system. In the future, the helium behaviour will be improved by integrating an empirical model for helium swelling and release in Pu/MA HTR fuels that will be delivered by other partners in the PUMA project.

2.3 Sampling of particle geometry and material properties

By use of the analytical stress analysis code (Boer et al., 2008b), the stress field in all coating layers can be calculated as a function of fast neutron fluence. This allows the failure probability to be calculated, for instance with the widely used Weibull distribution function (Sawa et al., 1996):

$$P = 1 - \exp \left(- \ln 2 \left(\frac{\sigma_t}{\sigma_{med}} \right)^m \right) \quad (5)$$

σ_t and σ_{med} are the maximum tangential (tensile) stress and median strength in the SiC layer, respectively. m is the Weibull modulus.

However, Eq. 5 gives the failure probability of the mean or nominal TRISO particle, which is different from the average failure fraction of all particles that are being irradiated in the core. The two methods for the calculation of failure probability/fraction differ because of the statistical variations of the TRISO geometry and material properties that occur during the fabrication process. In order to take this into account, Monte Carlo (MC) sampling has been deployed.

In such techniques, the size of the sample plays a major role. In order to calculate a failure fraction as low as 10^{-6} with good accuracy, the modeler should make use of a sample of 10^8 particles with Direct Monte

Carlo method. Advanced MC techniques aim at increasing the accuracy for the same sample size, or equivalently at decreasing the sample size for a given accuracy (here 10^{-6}). For instance, the sample size can be reduced to 10^6 particles in Conditional Monte Carlo, and to 10^5 particles in Importance Sampling MC (Michel, 2007). However, as highlighted in section 2.1 and shown in Fig. 2, the crack-induced failure model introduces many possibilities for the SiC layer to fail. As a consequence, only the Direct MC method is suitable here to calculate the failure fraction of TRISO particles.

Once the sampling method is selected, the next step consists in choosing the most realistic distributions for the particle geometry and mechanical property. For this purpose, the truncated gaussian distribution is chosen for sampling the kernel diameter and all coating layer thicknesses, which are calculated as follows:

$$x = \mu + \sigma \sqrt{2} \operatorname{erf}^{-1} \left[\operatorname{erf} \left(\frac{a - \mu}{\sigma \sqrt{2}} \right) + \left(\operatorname{erf} \left(\frac{b - \mu}{\sigma \sqrt{2}} \right) - \operatorname{erf} \left(\frac{a - \mu}{\sigma \sqrt{2}} \right) \right) \times R \right] \quad (6)$$

where x is the value of the parameter that follows the truncated gaussian distribution, a and b are the lower and upper bound of the possible values for x , respectively. μ and σ are the mean value and standard deviation, respectively, erf is the error function, and R is a random number.

The two-parameter Weibull distribution is preferentially used to sample the value of the characteristic strength of the coating layers, which is given by:

$$x = \mu \exp \left(\frac{\ln(-\ln R)}{\sigma} \right) \quad (7)$$

The variables in Eq. 7 have the same meaning as in Eq. 6.

Tab. 1 shows the values used in the truncated gaussian and Weibull distributions when the MC technique is applied. The values of the standard deviation for the kernel diameter and layer thicknesses are not referenced because they mainly depend on the fabrication process. The paper does not aim at validating a fuel performance code, but rather at analysing sensitive parameters and mechanisms.

3 Application to a PBMR-400 core design

Based on past Pu-burning experiments at Peach-Bottom (Alberstein, 1994), in which $\text{PuO}_{1.7}$ fuel kernels of 200 μm diameter were irradiated up to very

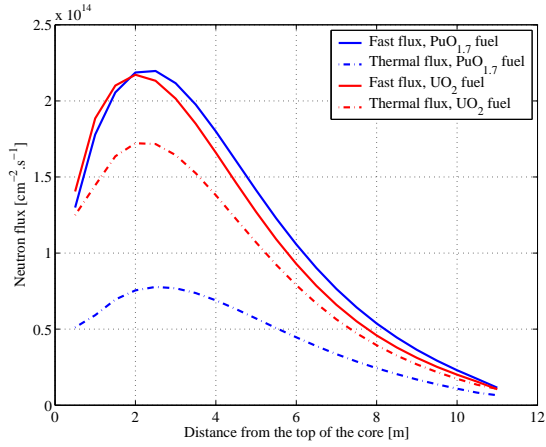


Fig. 3: Fast and thermal neutron fluxes in the core for both UO_2 and $\text{PuO}_{1.7}$ fuels.

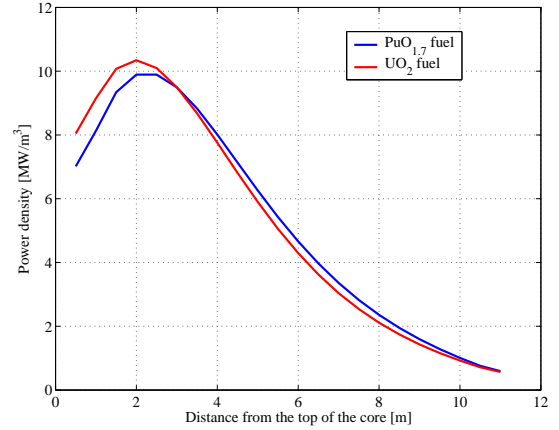


Fig. 4: Power density for both UO_2 and $\text{PuO}_{1.7}$ fuels.

high burn-ups, the starting fuel for all analyses in the PUMA project was chosen to be $\text{PuO}_{1.7}$ fuel kernels containing first generation Plutonium (i.e. from recycled LWR UO_2 fuel). The isotopic composition vector is given in Tab. 2. In the case of a pebble-bed type HTR, preliminary results were obtained with the use of (U-free) Pu-containing coated particle fuel in a contemporary design of the PBMR-400 pebbled-bed HTR operating in continuous multi-pass refuelling mode (de Haas et al., 2006), where it was assumed that each fresh fuel pebble contains 2 g first generation Pu in coated particles with a kernel diameter of 240 μm .

Calculations performed so far by use of the PANTHERMIX code on a Pu-loaded HTR-MODULE (Kuijper et al., 2004) show that a discharge burn-up of about 740 MWd/kgHM can be reached with 67% fissile Pu (1st generation, see Tab. 2), where k_{eff} is still slightly above 1 in the equilibrium (fuel mixture) state. In the present paper, the calculation of equilibrium state has been performed with a target burn-up of 700 MWd/kgHM, which should correspond to a value achievable without any particular optimisation of the core.

3.1 Calculation of equilibrium state

The code system developed at TU-Delft (Boer et al., 2008a) and shown in Fig. 1 enables to calculate the equilibrium state of the reactor core for a given target discharge burn-up. The results shown be-

low allow to make sure that the pebble-bed reactor is suitable for the transmutation of Plutonium and minor actinides during normal operating conditions. For this purpose, the equilibrium state has been calculated as well for UO_2 fuel, although for a much lower discharge burn-up (96 MWd/kgHM).

Fig. 3 shows the fast and thermal flux profiles within the core. It is shown that both fluxes with $\text{PuO}_{1.7}$ fuels are comparable to those with UO_2 , in terms of order of magnitude.

Despite the fact that the thermal flux for $\text{PuO}_{1.7}$ is lower than for UO_2 , the fissile content in the latter fuel is higher, and this results in very comparable power density profiles as shown in Fig. 4.

Although not shown here for sake of brevity, the maximum fuel temperature profiles deduced from the power density profiles are also almost the same for both fuels.

3.2 Burning capabilities

The next step consists in analysing the density profile of Plutonium isotopes during irradiation. This is shown in Fig. 5. In particular, both Plutonium fissile isotopes Pu^{239} and Pu^{241} are greatly burnt.

Tab. 2 shows the Pu-vector for fresh and discharged fuel. From this, it is calculated that the fissile Pu-fraction is reduced from 66.98 to 2.245 wt.%. Moreover, the discharged amount of Pu reduces to 22.98 wt.% of the original amount, while the total amount of actinides (U up to Cm) is reduced to 28.37 wt.%, as shown in Tab. 3.

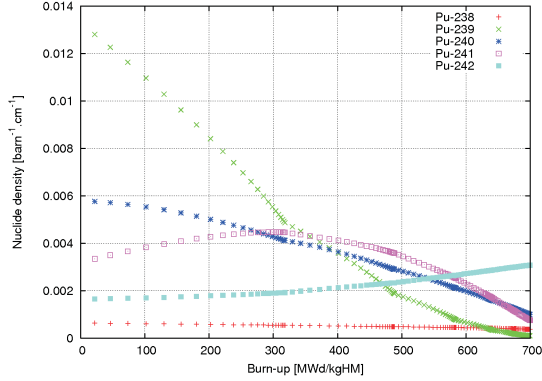


Fig. 5: Density of Plutonium isotopes as a function of burn-up.

Table 2: Vectors for fresh and discharged fuel (wt.% of initial heavy metal mass).

Burn-up (MWd/kg)	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
0	2.59	53.85	23.66	13.13	6.78
700	1.228	0.206	5.494	2.039	14.010

Table 3: Pu-burning capabilities (wt.% of initial heavy metal mass).

Burn-up (MWd/kg)	U	Np	Pu	Am	Cm
0	0.0	0.0	1.0	0.0	0.0
700	0.037	0.00079	22.98	3.447	1.901

At the target discharge burn-up of 700 MWd/kgHM, the burning rates of fresh Plutonium and minor actinides that are produced during irradiation are very promising for the use of the pebble-bed type HTR for transmutation purposes.

3.3 Stress analysis

From the above equilibrium state results, various profiles such as fast neutron fluence, burn-up, pebble-surface temperature and inventory of stable fission gases are used as inputs for the stress analysis code. When running the PASTA code, the fission gas release and the pressure in the buffer are calculated first. Fig. 6 shows typical fission gas release and pressure build-up profiles for the various models that are available in the PASTA code. In the legend, "numerical solution" refers to the solution of Eq. 2 by use of the finite differences method (Boer et al., 2008b) and is considered here as the reference, "analytical solution" with 5 or 10 iterations refers to Eq. 3 with $n = 5$ or 10, respectively, and "approx. solution" to Eq. 4.

Fig. 6 shows that the fission gas release as calculated with the analytical method with $n = 10$ converges quickly to the numerical solution. With only 5 iterations, the error is relatively important at low fluence. However, this results in very small discrepancy in the pressure profile, due to a low production of gaseous fission products at beginning of irradiation. Inversely, the approximate solution for the fission gas release is in good agreement with the numerical solution at low fluence but rapidly deviates from it. This results in a lower buffer pressure as irradiation proceeds. In section 4, the sensitivity of both approximate and analytical solutions is investigated.

Once the buffer pressure is calculated, the calculation of the stress field in all coating layer is performed. Fig. 7 shows typical tangential stresses in the iPyC, SiC and oPyC layers. When the PyC layers are not allowed to crack, the SiC layer remains under compression for a very wide range of buffer pressure profiles. However, in case that any PyC layer fails, there is an important change in the stress of the SiC layer, which can become tensile. The effect of this change is also investigated in section 4.

4 Discussion

By use of the stress analysis code, the calculation of failure fraction of TRISO particles during irradiation is investigated. For this purpose, it has been chosen to study the role of the sample size, fission gas release model and crack-induced failure model on the failure fraction. For a better analysis, all sensitivity calculations have been performed for both the fuel of interest here, i.e. $\text{PuO}_{1.7}$, and for UO_2 as a reference. The results are shown in Tab. 4.

In the second column of Tab. 4, 10^x refers to the number of sampled particles, "crack model" and "NO crack" whether the crack-induced failure model is employed or not, and FGR to the fission gas release model used in the given case: 100% means that all stable gaseous fission products are instantaneously released into the buffer, "analy. 5" means that Eq. 3 is used with $n = 5$, and "approx" when Eq. 4 is used. The results shown in Tab. 4 enable to draw some conclusions:

- The impact of PyC-cracking on the behaviour of the SiC layer, hence on the failure fraction can be analysed from cases 3, 5, and 7 with respect to cases 4, 6 and 8. When the crack-induced failure model is employed, this results in a failure fraction at least

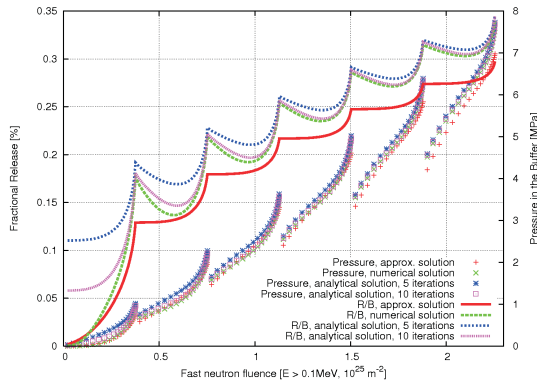


Fig. 6: Typical Fission Gas Release and pressure profiles during irradiation.

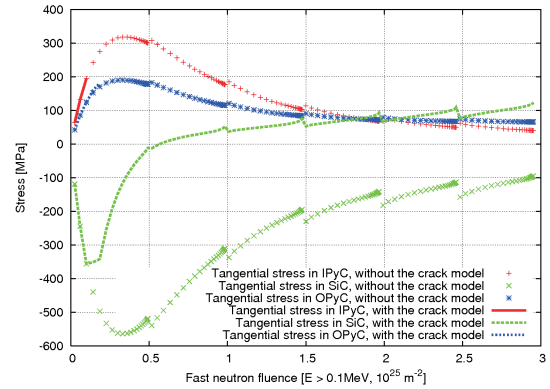


Fig. 7: Typical mechanical behaviour of a TRISO particle during irradiation, with and without the crack-induced failure model.

two orders of magnitudes higher than without the crack model. This shows clearly that attention must be put as well as on the PyC layers when optimising the design of TRISO particles. However, it should be mentioned that no crack propagation through the SiC occurred in any of the TRISO particles which were predicted to fail. The failure of the SiC layer only occurred due to the change in the stress field as shown in Fig. 7, which eventually becomes tensile. Further validation of the crack model as proposed by Wang and Ballinger (2004) is therefore necessary.

- The pressure build-up in the buffer is one of the main sources of stress induced in the coating layers. Therefore, both calculations of production in the kernel and release into the buffer of gaseous fission products are very important. With or without the crack model, a comparison between cases 3, 5 and 7 (or 4, 6 and 8) shows that the failure fraction is very sensitive to FGR. As shown in Fig. 6, the approximate solution given by Eq. 4 provides an underestimated buffer pressure, hence a lower failure fraction, with respect to the analytical solution provided by Kidson (1980). Moreover, assuming 100% gas release always leads to a higher and unrealistic failure fraction (compared to maximum 35% for some sampled particles), and this model should not be used in fuel performance codes.
- As already emphasized in section 2.3, the size of the sample is very important to obtain an accurate result. By comparing cases 1 to 3 and 2 to 7, con-

vergence in the failure fraction is almost achieved for $\text{PuO}_{1.7}$ fuel, but not for UO_2 fuel. The failure fraction of TRISO particles has therefore to be calculated from a bigger particles sample.

- Due to the very high discharge burn-up and a smaller buffer layer, it is shown that the Pu-based fuel particle has a much higher failure fraction than the U-based fuel particle. This implies to propose a new design in case that Pu-based fuels are employed in HTRs.

Table 4: Failure fractions for both $\text{PuO}_{1.7}$ and UO_2 for different configurations of sample size, FGR and stress models.

Case	Description	$\text{PuO}_{1.7}$	UO_2
1	10^6 particles, FGR=approx, crack model	2.3101×10^{-2}	3.1×10^{-5}
2	10^6 particles, FGR=analy, 5, crack model	4.6164×10^{-2}	5.7×10^{-5}
3	10^5 particles, FGR=approx, crack model	2.313×10^{-2}	4.0×10^{-5}
4	10^5 particles, FGR=approx, NO crack	3.0×10^{-5}	0
5	10^5 particles, FGR=100%, crack model	21.481×10^{-2}	7.10×10^{-4}
6	10^5 particles, FGR=100%, NO crack	7.5×10^{-4}	0
7	10^5 particles, FGR=analy, 5, crack model	4.647×10^{-2}	4.0×10^{-5}
8	10^5 particles, FGR=analy, 5, NO crack	3.00×10^{-4}	0

5 Conclusions

The aim of the present work is to develop a tool for the performance of TRISO particles fuelled with plutonium and minor actinide oxide in the framework of the PUMA project. For this purpose, a code system for the calculation of the equilibrium state of the

reactor core has been developed at TU-Delft, in addition to the stress analysis code for the calculation of failure fractions. In this way, particular attention is put to provide realistic modelling of particles during irradiation: the behaviour of all coating layers is taken into account through the integration of a crack-induced failure model in the PASTA code, the fission gas release is calculated accurately with special effort in the computation time, the contribution of helium is taken into account, and statistical deviation of the particle geometry and mechanical properties that occurs during the fabrication process is applied.

The results presented in the paper show that this tool is very efficient, flexible and appropriate for the design of TRISO particles with innovative fuel compositions. Moreover, the calculations performed so far show that the Pu-based fuel particles should have a new design.

Further development of the stress analysis code will include a detailed description of helium behaviour in Pu/MA-based fuels. Moreover, a parallel version of the code will be proposed in order to perform much more calculations in a reasonable time. Finally, the present code will be applied to fuel kernels that contain minor actinides taken from recycled PWR spent fuel after five years of cooling. This is an option that has to be considered in the analysis of the possible PWR-HTR fuel cycles.

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