Design of a Spherical Fuel Element for a Gas Cooled Fast Reactor

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A study is done to develop a fuel cycle for a Gas Cooled Fast Reactor (GCFR). The design goals are: highly efficient use of (depleted) uranium, application of Pu recycled from LWR discharge as fissile material, high temperature output, and simplicity of design. The design focuses on spherical TRISO-like fuel elements, a homogeneous core at startup, providing for easy fuel fabrication, and self-breeding capability with a flat k_{eff} with burnup. Nitride fuel ($^{15}N > 99\%$) has been selected because of it's favourable thermal conductivity, high Heavy Metal density and compatability with PUREX reprocessing. Two core concepts have been studied: one with coated particles embedded inside fuel pebbles, and one with coated particles cooled directly by helium. The result is that a flat k_{eff} can be achieved for a long period of time, using coated particles cooled directly, with a homogeneous core at startup, with a closed fuel cycle and a simple refueling and reprocessing scheme.

1. Introduction of the Gas Cooled Fast Reactor

The Gas Cooled Fast Reactor (GCFR) is a nuclear reactor with a fast neutron spectrum and gas cooling. In the late sixties and early seventies, several research programmes for GCFR concepts were initiated[2]. The designs featured both classical fuel pins and coated particles, and used CO_2, He, or N_2O_4 as a coolant. The main problems then were related to the structural materials and the inability to provide adequate cooling in the case of a LOCA. The GCFR was abandonned in favour of the liquid metal cooled fast reactor. Recently, the Generation IV International Forum has included the GCFR as one of the six reference reactor concepts for the future, focusing on the advantages of a gaseous coolant: helium is transparent, neutronically and chemically inert, cannot boil and enables operation at high temperatures.

One of the characteristics of FR systems is the high core power density for economical reasons. This is related to the large fissile inventory required to obtain a critical system, and the desire to have a short doubling time. The fuel costs can be reduced by using a mix of depleted U and LWR discharge Pu. Then the core power density can be lower whilst maintaining an economical system. The design presently under investigation focuses on high temperature operation, He coolant, and a simple design, including a fully homogeneous core at startup, for which only 1 type of fuel element needs to be produced. The core should have a self-breeding capability, and a flat k_{eff} during burnup. The fuel cycle should be closed, i.e. only fission products should leave the cycle after reprocessing.

2. Coated particles for a Gas Cooled Fast Reactor

We envisage the use of coated particle (CP) fuel for the GCFR. We have prepared a design of a coated particle based on the TRISO CP as used in thermal HTR applications. The TRISO CP consists of a spherical fuel kernel (typical diam. 500 μm), surrounded by a low density graphite buffer

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[2] See for instance: *GCFR fuel assemblies*, J. Chermanne, Gas Breeder Reactor Association, 1972, and *Design study of a He gas-cooled Fast Breeder Reactor*, Mochizuki et al., Kawasaki Heavy Industries, 1972.

layer (porosity 50% or more), an inner layer of pyrolytic carbon (IPyC), a sealing layer of SiC, and an outer layer of pyrolytic carbon (OPyC). The buffer layer provides voidage to store the gaseous fission products that are released from the fuel, prevents mechanical interaction between kernel and sealing layer, and protects the sealing layer from damage by recoiling fission fragments. The SiC sealing layer acts as cladding, retaining all fission products within the coated particle. The IPyC and OPyC layers contract under irradiation, thereby partly releaving the stresses in the SiC layer caused by the pressure within the CP[3]. To reduce the amount of moderating material in the GCFR core, the pyrolytic carbon layers are removed from the particle.

The SiC cladding layer fails when the pressure inside the CP is too large. The maximum allowable pressure within the CP can be caluated as follows: assume a sphere of radius R surrounded by a shell of thickness δ. If the pressure inside the shell is higher than outside, the tangential stress in the shell can be expressed as a function of the pressure difference ΔP acting on the shell. If $\delta << R$ (thin shell approximation), the tangential stress in the shell is given by:

$$\sigma_{xx} = \sigma_{yy} = \frac{R}{2\delta} \cdot \Delta P .$$

Note that σ_{xx} is a function of the ratio R/δ. The maximum allowable ΔP is fixed by the choice of material (σ_{max}) and the geometry (R/δ) of the particle.

The fission process leads to swelling of the fuel and the generation of gaseous fission products. If we assume an ideal gas model for the gaseous FP in the buffer, the pressure in the buffer can be written as a function of burnup as:

$$P_{buf} = \frac{FIMA \cdot n_0 \cdot z \cdot k \cdot T_{buf}}{V_{buf}} ,$$

in which FIMA[4] stands for <u>F</u>issions per <u>I</u>nitial <u>M</u>etal <u>A</u>tom, n_0 is the number of heavy metal atoms in the fuel kernel at Start of Life (SOL), z is the number of gas atoms released into the buffer per fissioned metal atom, k is Boltzmann's constant, ε is the porosity of the buffer layer, and T_{buf}, V_{buf} are the temperature and free volume of the buffer. Note that V_{buf} is a function of burnup. The pressure in the buffer layer must not exceed the limits of the sealing layer.

Let the number of gaseous FP produced per fission be z^*. A fraction of these atoms will be released into the buffer layer. Swelling of the fuel is caused by the increase in the number of atoms in the fuel, and by the generation of fission gas bubbles . If the fraction of gaseous FP released from the fuel is high, swelling is reduced and vice versa. In fuel pins swelling can be reduced by using a high porosity fuel that is able to release a large fraction of the gaseous FP to a fission gas plenum. CPs on the other hand are a closed system, so there is no possibility to reduce the pressure increase with burnup.

For a coated particle consisting of a fuel kernel, a graphite buffer layer and one SiC sealing layer, the pressure in the buffer was calculated as a function of the kernel radius and burnup. The outer radius of the CP is fixed at 450 μm, the thickness of the cladding layer is fixed at 75 μm, while the kernel radius varies between 180 and 350 μm with the buffer thickness changing accordingly. Other parameters are given in Table 1, and the result is given in Fig. 1. The plane in the figure indicates the

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[3] *Considerations pertaining to the achievement of high burn-ups in HTR fuel*, D.G. Martin, Nuclear Engineering and Design, 2002.
[4] Throughout this paper we will use FIMA as a measure of burnup. 1% FIMA (meaning that 1% of initial heavy metal has been fissioned) corresponds roughly to a burnup of 9500 MWd/t.

maximum allowable buffer pressure. A coated particle can be used for any combination of R_k and FIMA on the surface under the plane. For small R_k 100% FIMA can be accomodated, but for increasing R_k and corresponding decreasing buffer thickness, the maximum attainable FIMA decreases rapidly. A small kernel leads to a low fuel fraction in the core and hence the fissile loading needs to be increased. For coated particle fuel in a GCFR there will always be a trade-off between the fissile fraction and the maximum attainable burnup.

Table 1: Material properties used to calculate the buffer pressure in a TRISO coated particle

Porosity (ε) of the buffer	60%
Nr. of gas atoms per fission / release [%]	0.263[a] / 55[b]
Nr. of volatile atoms per fission / release [%]	0.278[a] / 55[b]
Nr. of gas atoms in buffer per fission	0.3
Fuel swelling ΔV/V per percent FIMA [%]	1.5[c]
σ_{max} SiC cladding layer [MPa]	500
Max. allowed pressure in buffer [MPa]	182

[a] Calculated with ORIGEN-S and JEF-2.2 based library[5]. Noble gases: He, Kr, Xe. Volatiles: I, Cs, Rb, Te.
[b] In Bailly et.al.[6] values are given for fission gas release from FR MOx fuel pins. Fission gas release in FR MOx fuel is high (>80%) because of the steep thermal gradient and the presence or formation of a central void in the pellets. The thermal gradient is small for CPs, so the release of fission gas is estimated at 55%.
[c] Bailly et.al.[4] state that the swelling of nitride fuel is always lower than oxide, with the standard figure for FR MOx being 0.7%. Petti[7] states that swelling of oxide fuel in HTRs is considerably higher than in PWRs. Given the low estimated fission gas release the swelling is estimated at 1.5%.
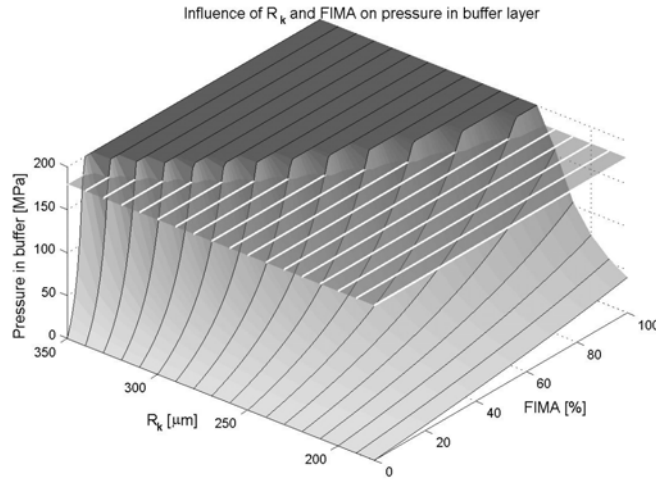

Fig. 1: Maximum allowable FIMA in a coated particle as a function of R_k. The plane corresponds to P_{buf} = 180 MPa. A coated particle can be used until the pressure in the buffer reaches this maximum allowable value. CPs with a large kernel and corresponding small buffer can reach only low burnup.

3.   Calculation scheme

A short overview of how the simulations of this paper were performed: A 172-group cross section library based on JEF-2.2 is used. Calculations are 1-D, with an axial buckling.

[5] *Generation and Validation of ORIGEN-S Libraries for Depletion and Transmutation Calculations Based on JEF2.2 and EAF3 Basic Data,* J.E. Hoogenboom and J.L. Kloosterman, Nuclear Engineering and Design, 1997.
[6] *The nuclear fuel of pressurized water reactors and fast reactors*, ed. H. Bailly, CEA, France, 1999
[7] *Coated Particle Fuel Behaviour Under Irradiation*, D. Petti, INEEL, USA, 2002

SCALE[8] CSAS (BONAMI - NITAWL - XSDRNPM) is used to generate the cell-weighted cross-sections to calculate the flux pattern and power profile. Fuel depletion in each zone of the reactor is calculated with COUPLE - ORIGEN-S. The Pu used for all calculations presented in this paper has been recycled twice in LWRs. The isotopic vector[9] at SOL is given in table 2.

Table 2: The isotopic vector of the Pu used in the simulations presented in this paper

Isotope	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Fraction at SOL	1 %	62 %	24 %	8 %	5 %

4. A pebble bed for the GCFR

The materials in the reactor can be classified as Coolant, Fuel and Moderator (CP cladding layers and structural materials). For a pebble bed system with spherical CPs packed into spherical fuel elements (pebbles), the following volume fractions apply:

$$F_c = (1 - \beta_p)$$

$$F_f = \beta_p \beta_T \left(\frac{R_i}{R_o} \right)^3 \left(\frac{R_k}{R_c} \right)^3$$

$$F_m = 1 - F_f - F_c$$

β_p is the packing fractions of the pebbles in the core, β_T is the packing fraction of TRISO CPs within the fuel zone of the pebbles. For a Random Close Packing (RCP), β is 0.63. All other symbols are indicated in Fig. 2. The volume fractions can only change by choosing different geometries for the pebble and the CP.
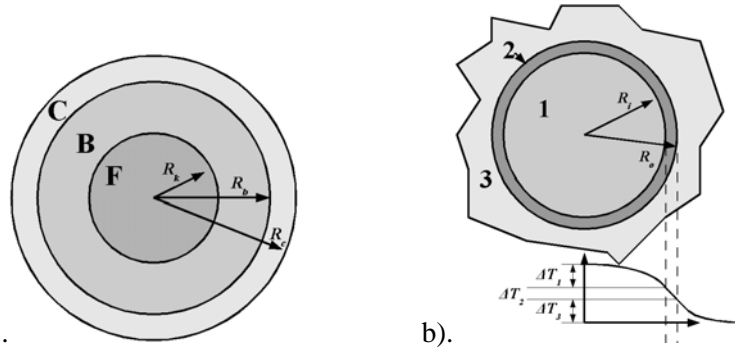

a).   b).

Fig. 2a.: A schematic of a coated particle. **C = C**ladding layer, B = **B**uffer layer, F = **F**uel kernel, R_c = radius of cladding, R_b = radius of buffer, R_k = radius of kernel.

Fig. 2b.: schematic of a fuel pebble. **1**: fuel zone, coated particles in a matrix material, **2**: unfueled shell, **3**: coolant, R_i = radius of fuel zone, R_o = outer radius of pebble. Underneath the pebble the temperature profile inside the pebble is indicated.

Heat is generated in the CPs. The fuel pebble consists of a fuel zone with CPs, and an unfueled surrounding shell. Assuming a homogeneous fuel zone , and denoting the total power produced per pebble as q_p, the temperature gradients ΔT_1, ΔT_2 and ΔT_3 (as indicated in Fig. 2b.) are[10]:

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[8] *SCALE-4.2 Modular code system for performing standardized computer analyses for licensing evaluations*, Oak Ridge National Laboratory, Tennessee, US, 1994

[9] *Plutonium (1st and 2nd generation) cell burnup benchmark specification*, H.J. Ruetten, J.C. Kuijper, Document HTR-N1-02/06-s-3.1.1, European Commission, 2003

[10] *Hochtemperaturreaktortechnik*, K. Kugeler, R. Schulten, Springer-Verlag, 1989

$$\Delta T_1 = \frac{q_p}{8 \pi R_i \lambda_1} \, ,$$

$$\Delta T_2 = \frac{q_p}{4 \pi \lambda_2} \left(\frac{1}{R_i} - \frac{1}{R_o} \right),$$

$$\Delta T_3 = \frac{q_p}{4 \pi R_o^2 h_c} \, .$$

λ_i is the thermal conductivity of zone i. The temperature difference between coolant and the centre of a fuel pebble equals:

$$\Delta T = \sum_{i=1}^{3} \Delta T_i$$

As expected the difference between the centerline temperature and the temperature of the coolant is proportional to the power produced per pebble, and inversely proportional to the thermal conductivity of the fuel zone. For the GCFR, 63% of the fuel zone of the pebble is made up of TRISO particles. The centerline temperature in the fuel pebble will be very high if a coolant temperature of 1000 K or more is desired. The matrix material should have a melting point, high thermal conductivity and low neutron absorption. In thermal HTRs graphite is used, but graphite (C) is unacceptable in the fast spectrum because it is a good moderator. Therefore, we chose Zr-metal for the matrix, because of its low neutron absorption cross section, high melting temperature and low moderating power.

Simulations were performed for a cylindrical pebble bed GCFR, in which $\beta_p = \beta_T = 0.63$. Pebble geometry: $R_o = 3$ cm, $R_i = 2.75$ cm, core geometry $R_{core} = 250$ cm, $h_{core} = 500$ cm. The core is surrounded by a 1 meter thick reflector of stainless steel. The average power density in the core was chosen as 300 MW/m^3. This power density was chosen to be comparable to the core power density of a 'conventional' breeder reactor. Coated particle geometry as in Section 2, with R_k variable between 180 and 350 μm. The core is homogeneous at SOL. The result is that there is a trade-off between kernel size and fissile fraction to obtain $k_{eff} > 1$ at SOL. The dependence of k_{eff} on kernel size and Pu content (Pu / (Pu + U) atomic denstity) is given in Fig. 3.

The variation of k_{eff} with burnup was calculated for 3 configurations: $R_k = 350$ μm, Pu = 18% and 30%, and $R_k = 300$ μm, Pu = 24%. For $R_k = 350$ μm, a Pu content of 18% gives $k_{eff} > 1$ for a fresh core. To get roughly the same k_{eff} with $R_k = 300$ μm, 24% Pu is needed. The decrease of k_{eff} with burnup is less steep with lower Pu content. To reach the design targets of a flat k_{eff} the Pu content has to be around 13%. A CP with $R_k \leq 350$ μm, Pu = 13%, gives $k_{eff} < 1$ at SOL.
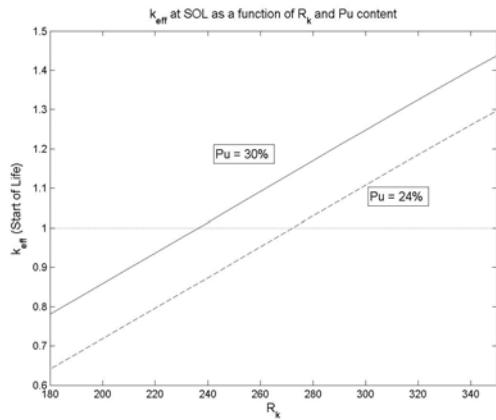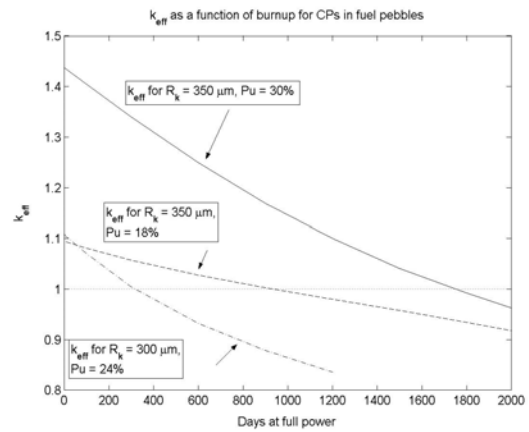
Fig. 3a.: The influence of the kernel radius on k_{eff} at SOL for a homogeneous pebble bed GCFR. As expected, a higher Pu content leads to higher k_{eff}.

Fig. 3b.: The variation of k_{eff} as a function of burnup in a homogeneous core. High initial Pu content deteriorates breeding, a low Pu content leads to a flatter curve for k_{eff}. However, to get k_{eff} roughly constant, the Pu content should be well below 24% for a fresh, homogeneous core.

The main issue concerning the pebble bed GCFR core is the fraction of fuel in the core, which is very low. For the geometry used in this simulation, the fuel fraction F_f varies between 14% and 2% of V_{core} (compare: > 35% for LMFBR). The large amount of moderator material in the core (39% to 61%) leads to a softening of the spectrum and large parasitic neutron capture.

We discarded the pebble bed core, because it does not meet the design goals we set for the GCFR.

5. Coated Particles cooled directly by helium

To reduce the fraction of moderator material, the coated particles in this core design are cooled directly by helium. In this concept, the volume fraction of coolant is the same as in the pebble bed concept, but the fraction of moderator material is reduced. With R_k between 180 and 350 µm the fraction of fuel is between 4% and 29.6%. This layout also eliminates problems concerning the temperatures inside the fuel elements, reduces the temperature gradient within the fuel element and increases the heat exchanging surface per unit fuel material. A simulation was done for a reactor of R_{core} = 150 cm, h_{core} = 275 cm, with a stainless steel reflector with a thickness of 1 m.. Note the difference with the previous reactor geometry. CPs have a geometry as given in Section 2, with R_k varying between 180 to 350 µm, and Pu = 12% to 30%. The result is illustrated in Fig. 4. Again there is a trade-off between kernel radius and Pu content, but the fissile fraction required to obtain $k_{eff} > 1$ at SOL with a homogeneous core is reduced drastically.

A design with R_k = 310 µm, Pu = 13% was selected for a burnup study. The buffer pressure model illustrated in Fig. 1 indicates that R_k = 310 µm corresponds to a maximum FIMA value of roughly 25%, so we set the burnup target at 20% FIMA. The core is homogeneous at SOL: all coated particles in the core are identical, with the same fuel composition. This results in a large power peaking in the centre of the core. The fuel in the centre of the core will burn up faster than the fuel in the outer zones. To model this, the core is divided into 5 concentric zones of equal volume, numbered 1 to 5 (center to edge). At SOL, all CPs are identical. In Fig. 5 the results are illustrated.
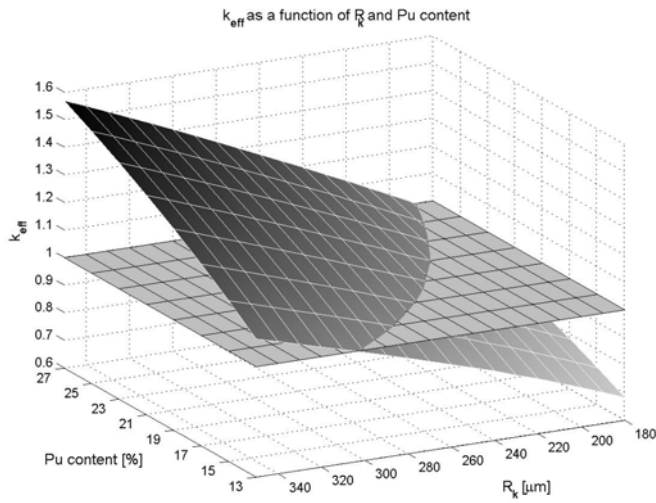
Fig. 4: k_{eff} as a function of R_k and Pu content at SOL. All combinations of R_k and Pu content that result in $k_{eff} > 0$ are usable. If the core is homogeneous and k_{eff} is required to be constant with burnup, the Pu content must be chosen as low as possible. This means that only CPs with a large kernel ($R_k > 300$ µm) can be used.

The result is that when zone 1 (centre of the reactor) reaches the target burnup of 20% FIMA, the outer zone has reached only 7% FIMA. Recycling all fuel when zone 1 reaches the target burnup is inefficient, because 80% of the fuel in the core is still usable. Therefore, a refueling scheme is introduced in which each zone is refueled when the burnup reaches 20% FIMA. From Fig. 5b the refueling intervals ([days]) were chosen as: Zone 1: 1100, Zone 2: 1400, Zone 3: 1800, Zone 4: 2200, Zone 5: 2800. The refueling scheme is illustrated in table 3: the first and second batch of fuel are identical for each zone. The material discharged from a zone after irradiation is reprocessed while a new batch of fuel is being irradiated in that zone. All U and Pu are recycled from the discharged material, and depleted U (U-238 = 99.8%) is admixed to the material to get the same HM density as at SOL. This material is then inserted into the core as new fuel when the present batch of fuel is discharged.
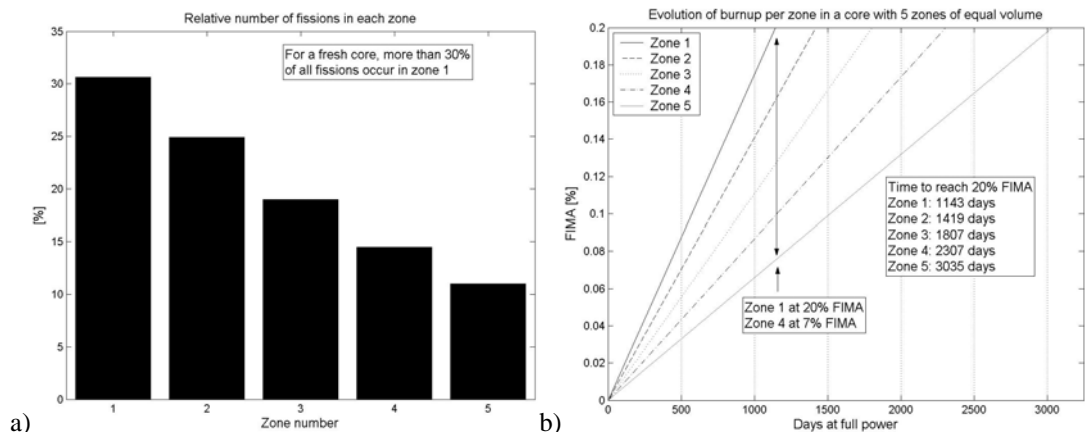

Fig. 5a.: The power distribution at SOL. At SOL, 30% of all fissions occur in zone 1, and some 11% in zone 5. As a result, fuel burnup is 3 times faster in zone 1 than in zone 5.
Fig 5b.: Time to reach 20% FIMA in each zone.

Table 3: The refueling and reprocessing scheme for the GCFR

Material in a certain zone of the reactor	Cycle 1: fresh fuel, used until FIMA = 20%	Cycle 2: fresh fuel, used until FIMA = 20%	Cycle 3: Fuel based on reprocessed HM from cycle 1.	Cycle 4: Fuel based on reprocessed HM from cycle 2
Material of a certain zone, outside of the reactor		Reprocessing of material discharged at end of cycle 1	Reprocessing of material discharged at end of cycle 2	Reprocessing of material discharged at end of cycle 3.

The calculated k_{eff} for a GCFR with the proposed refueling scheme is given in Fig. 6 for a period of 7250 days at full power. k_{eff} does not drop below 1 and shows an increasing trend. In the figure refueling of each zone is indicated by the numbers in the boxes. Sometimes, more than 1 zone is refueled at one time.

The increasing k_{eff} with burnup means that there is no requirement to have a large extra reactivity at SOL to accomodate high burnup. The requirement was to have a flat k_{eff}. This can probably be achieved by using a lower density of HM in the later cycles. The core can reach high burnup without the need of burnable poison or control rods, increasing safety and decreasing fuel costs.
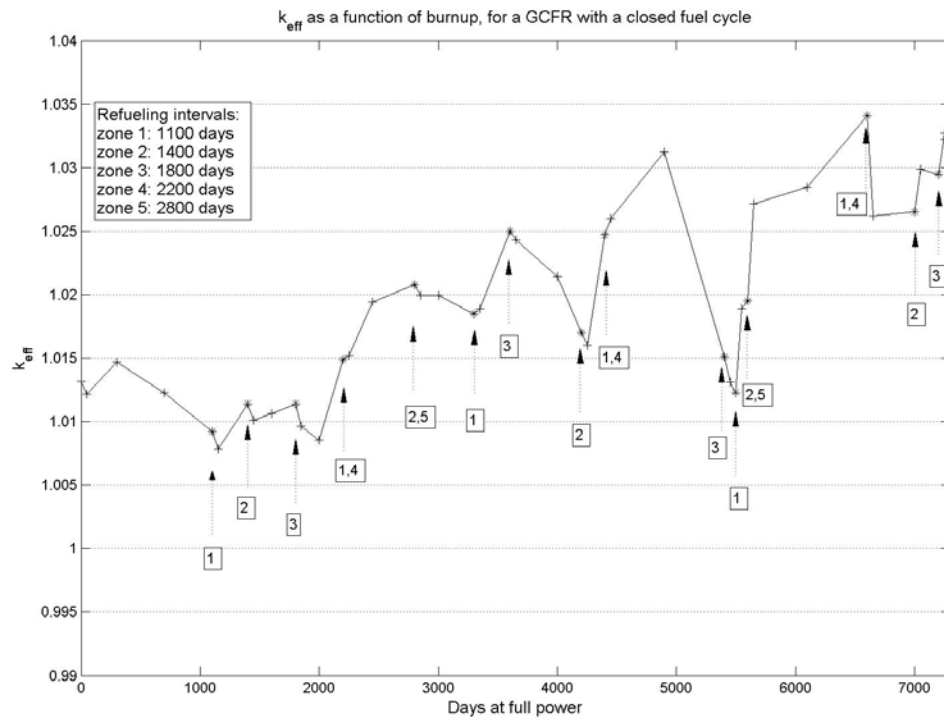

Fig. 6: Calculated values of k_{eff} for a GCFR with 5 zones, and a refueling scheme as discussed above. k_{eff} calculations are indicated with a '+', refueling events with '*'. The numbers in the boxes indicate which zone is/are refueled. The step size is variable, but at least 0 and 50 days after refueling k_{eff} is calculated to get the correct densities of long- and short-lived fission products.

In Fig. 7 the evolution of the 4 most abundant Pu-isotopes (Pu-239 to Pu-242) is illustrated. Pu is bred during burnup, but the increase in Pu is almost entirely Pu-240, so the fissile mass (Pu-239 and Pu-241) does not increase. The behaviour is similar both for a single zone (zone 1, illustrated in Fig. 7a) and the entire core (illustrated in Fig. 7b). The change in isotopic vector of Pu is given in table 4.

Table 4: isotopic vector of Pu at SOL and after 7200 days

	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242
Start of cycle 1	1 %	62 %	24 %	8 %	5 %
After 7200 days	0.3 %	54.8 %	34.4 %	6.2 %	4.3 %

Note that only U-238 is added to the fuel cycle after the reactor is switched on: all fissile material required is bred by the core itself. The only time when Pu is added to the system is at SOL. U-238 is a rest product of enrichment for LWR plants, and the Pu used at SOL is material recycled from MOx fuel for LWRs. Hence the fuel material is cheap, which is offset by the fact that the production of CPs is probably quite expensive. The breeding gain, especially breeding of fissile material, can be improved by using shorter refueling intervals.
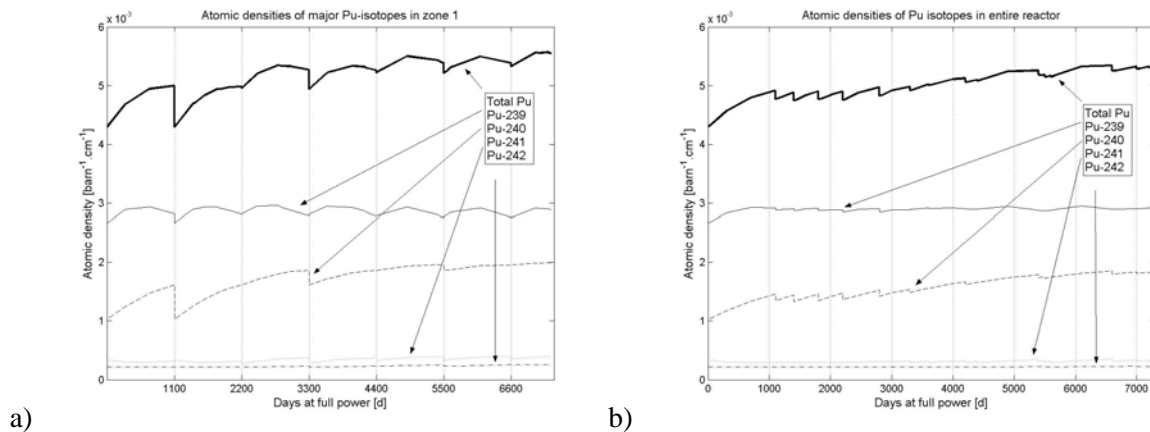
a)   b)

Fig. 7a.: The evolution of the atomic density of 4 Pu isotopes (Pu-239 to Pu-242) in the central zone.
Fig. 7b.: The evolution of the atomic density of 4 Pu isotopes (Pu-239 to Pu-242) in the reactor as a whole. During irradiation, the concentration of Pu-239 is roughly constant, while Pu-240 shows a steady increase. The thick line is the total concentration of all Pu isotopes.

6. Conclusions and future research.

A design is made for a GCFR core with the following objectives: coated particle fuel, a homogeneous core at SOL, and a constant k_{eff} with burnup.

A pebble bed core, with coated particles embedded in large fuel spheres, is discarded because a flat k_{eff} cannot be reached, and the thermal behaviour is doubtful.

A core with coated particles cooled directly by helium has a better performance. This concept has a homogeneous core at SOL with 5 zones of equal volume, and with a simple refueling and reprocessing scheme, k_{eff} is roughly constant over a long period of time. Only U-238 is added to the core during it's lifetime. No Pu is taken out of the fuel cycle, giving a nearly closed fuel cycle (Minor Actinides are not recycled). This concept uses only inexpensive fuel materials: the Pu at SOL is recycled from LWR cores, and for the U-238 the tails of the enrichment process can be used. In the near future the core concept will be extended to a system with continuous refueling.