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D23.21 - Report on Establishment of a small generic Test Facility (pebble mill)

Authors: Max Schreier, Martin Lustfeld, Wolfgang Lippmann, Antonio Hurtado (all TUD)

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Max Schreier, Martin Lustfeld, Wolfgang Lippmann, Antonio Hurtado (all TUDresden)		
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Summary

Graphite dust generation due to the abrasion of graphite core material is a significant safety issue in Pebble Bed High Temperature Reactors. Objective of this task is to establish a small-scale experimental facility producing dust particles under conditions as close as possible to those existing in a reactor pebble bed.

The test facility has been designed with the aim of covering most possible abrasion scenarios: Pure grinding of graphite fuel pebbles on graphite reflector walls, a combination of grinding and rolling of pebbles on each other and rotation of pebbles onto each other around a single point of contact. The maximum applicable load has been chosen equal to the maximum possible load occurring in a reactor core. Experiments may be performed under ambient air or shielding gas conditions. The relative velocity at the point of interaction can be varied in a wide range. Two different kinds of fuel pebbles are available for abrasion experiments: A-3 and MLRF-1 blind pebbles. Therefore, it can be determined how the parameters type of contact, graphite grade, relative velocity and load affect particle characteristics such as quantity, size distribution and shape.

Approval

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All technical participants	ARCHER	
Steering Committee members	ARCHER	

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1 Introduction

Superior safety standards are a crucial characteristic of the High Temperature Reactor (HTR) concept. Most importantly, the occurrence of a meltdown of the reactor core (triggering a major release of radioactive fission products) can be excluded if designed accordingly [1]. However, in some accident scenarios radioactive contamination of the environment cannot be ruled out entirely, such as in the case of a depressurized loss of forced cooling (DLOFC).

In a Pebble Bed High Temperature Reactor, the reactor core consists of a large number of spherical graphite fuel elements with a diameter of 60 mm. In each pebble, nuclear fuel is dispersed in several thousand so-called TRISO-coated particles with a diameter of 1 mm, yielding about 7 g of nuclear fuel per pebble [2]. The pebbles are loaded into the reactor core as a packed bed with conical floor. During operation, used pebbles can be removed at the bottom of the cone to be replaced with fresh ones loaded to the top of the bed. Due to this overall movement in the pebble bed, relative movements between pebbles and reflector as well as between pebbles among each other are unavoidable. This leads to slow, but continuous abrasion of the graphite fuel pebbles and thereby to generation of nano- and micro graphite dust particles which are dispersed in the helium coolant and can be transported throughout the reactor primary circuit. Additional potential sources of dust production in a HTR core are chemical reactions between impurities in the coolant and the surfaces of the graphite structures and pebbles [3].

Adsorbing fission products such as Ag-110m, I-131 or Cs-137 present in the primary circuit, these particles can become radioactively contaminated and deposit on the surface of primary circuit components during normal operation. During a DLOFC, these particles could be remobilized due to the pressure gradient in the primary circuit [4]. The possibility of leaking graphite particles crossing the reactor system boundaries and causing an unacceptable contamination of the environment needs to be ruled out for successful licensing of such a reactor type. In order to design accurate barriers (such as filter systems), a precise knowledge of shape and size distribution of generated graphite particles is indispensable. Additionally, particle characterization is highly relevant for particle transport simulations in order to identify places of preferential particle deposition in the primary circuit and to estimate the fraction of remobilized particles.

Graphite dust particles generated in the core of previously operated HTRs such as prototype reactor AVR Jülich [5], [6] and demonstrator THTR [7] had been collected and thoroughly investigated with respect to their shape, size and radioactivity in prior work. However, basic characteristics such as size distribution of particles found in each reactor turned out to be completely different from each other making it impossible to define a prototype graphite dust based on these results. Furthermore, it is likely that parameters such as graphite grade, type of relative movement (point or line contact, rolling or grinding) and load on the contact surface highly affect the characteristics of generated particles. Therefore, generic experiments on graphite particle generation need to be carried out which provide a better understanding of abrasion mechanisms and particle characteristics as basis for particle transport simulations required for licensing purposes.

Experimental investigations with similar objectives have been conducted since the 1960s [8], [9]. Recently, investigations have been performed particularly in China within projects related to construction and operation of HTR-10 and HTR-PM [10-12]. However, no unambiguous information on the mentioned particle characteristics could be obtained. Therefore, the aim of this task is to gather sufficient data for completing the necessary data base by using a small-scale generic test facility under realistic abrasion conditions and varying parameters. The design of this test facility, called pebble mill, is based on the authors' experience according to the parameters relevant for particle generation. Consideration of high operation temperatures as a further parameter is out of the scope of this task and should be objective of future projects.

2 Design of the test facility

Objective of the small-scale test facility presented in this work is to produce graphite dust particles by abrasion of graphite core structural material under conditions as close as possible to a pebble bed core. Main goal during the course of construction has been to establish a realistic relative movement between pebbles and reflector walls similar to that of pebbles transported through the reactor pebble bed.

Several parameters affect both the amount of generated particles and particles characteristics such as shape and size distribution. Of major influence are the surrounding atmosphere (particularly oxygen fraction), the shape of the graphite structures under investigation, contact load, graphite grade, relative velocity and operating temperature. Investigations within this task using this test facility will focus on the variation of the following parameters:

- atmosphere (shielding gas with varying fraction of oxygen)
- contact type of the interacting surfaces (e. g. pebble-pebble contact)
- load on the contact surface
- relative velocity in the center of the contact surface
- graphite grade (investigation of 2 different kinds of blind pebbles)



Figure 1: Model of the assembled pebble mill without acrylic vessel

All experiments will be conducted at ambient temperatures. The investigation of the influence of a varying temperature is out of the scope of this task and needs to be considered in future projects. Contrary to experimental investigations conducted in [10-13] on the basis of generic graphite samples, in the experiments planned within this study typical graphite pebbles with 60 mm diameter will be used. Blind pebbles of type A-3 and MLRF have been supplied by FZ Jülich and blind pebbles of type MLRF have been supplied by SGL Carbon. The maximum attainable range of experimental parameter variation of the pebble mill is given in table 1.

Table 1: Range el parameter variation	Table 1. Range of parameter variation of experimente with the test rability		
Parameter	Variation range		
Atmosphere	Mixture of shielding gas (He, N ₂) and Oxygen		
Interacting surfaces	Pebble-Pebble, Pebble-Pebble-Pebble, Pebble-Wall		
Contact load	10 – 800 N		
Rotational velocity	10 – 100 min ⁻¹		
Graphite grades	A 3, MLRF blind pebbles / NBG-17 graphite disc		

In figure 1 the small-scale test facility "pebble mill" is shown without surrounding acrylic vessel. Main components are the actuator, the rotating disc with bearing and the load application device allowing for a precise variation of the normal force at the contact point of the grinding components.

2.1 Contact configurations of interacting surfaces

In order to accurately simulate the pebble movement within a reactor core, three types of interaction between different components have been identified and implemented in the pebble mill presented in the following paragraph. An overview is given in figure 2.



Figure 2: Different contact configurations applicable in the pebble mill

2.1.1 Contact type 1: Pebble – Pebble – Pebble contact ("ppp")

A pebble-pebble-pebble-contact is the default case in which a specific pebble is in contact with several other pebbles in the reactor core, where the surface of contact is not normal to the gravitational force. This has been accomplished by placing a free full pebble between 3 fixed half pebbles (on the top) and 3 rotating half pebbles (on the bottom). This leads to a complex relative movement between these pebbles consisting of a rolling and a grinding component as occurring in a pebble bed reactor core.

2.1.2 Contact type 2: Pebble – Wall contact ("pw")

In the second case, the contact between a moving pebble and the flat reflector wall is investigated. A graphite disc of NBG-17 graphite supplied by SGL Carbon is used for this purpose. In this case, the disc is coupled to the actuator, rotating against the fixed half pebbles on the top.

2.1.3 Contact type 3: Pebble – Pebble contact ("pp")

Based on this configuration, the rotation of a pebble on top of another pebble at a single contact point will be investigated. Several previous investigations have been concerned with this extreme scenario where the contact surface between pebbles is perpendicular to the gravitational force.

Comparison of particles generated in all three cases will give a clear indication on to which extent the configuration affects particle characteristics. Further configurations are applicable with the pebble mill such as a contact between metallic components (fuel handling system) and graphite pebbles if the investigations of graphite components among each other yield satisfactory results.

2.2 Load on contact surface and rotational velocity

2.2.1 Conservative estimation of the load acting on a pebble in the pebble bed

In order to specify an appropriate range for the normal force applicable with the pebble mill at the point of contact between the grinding components, the forces acting within a real pebble bed need to be estimated. The following conservative assumptions yield an upper limit for the normal force on a horizontal layer of pebbles within the pebble bed:

- The normal force due to gravity is not balanced by the horizontal reflector walls.
- Each horizontal layer is charged with the entire load of all pebbles above this layer, i.e. the normal force acting on one pebble in a certain layer in the core is the gravitational force of all pebbles in the pebble bed above this layer divided by the number of pebbles in this layer.
- The void fraction of the pebble bed is ε =0.377 [14].

The heavy metal content of each pebble is about 7 g [2], [15].

The total mass of graphite pebbles loaded in the pebble bed can be calculated considering the void fraction ϵ through:

$$m_{core} = V_{core} \cdot \rho_{graphit} \cdot (1 - \epsilon) \tag{1}$$

The graphite density has been determined based on gravitational measurement of a sample of 100 graphite blind pebbles yielding an average mass of 197 g per pebble. Taking into account an additional 7 g of heavy metal content per nuclear fuel pebble [15], a pebble density of 1799 kg/m³ can be assumed. Given the void fraction ε an average pebble bed density of 1121 kg/m³ is derived, slightly less than the value of 1184 kg/m³ found in [10]. However, due to lack of further information in [10], the origin of the difference between these values could not be revealed.

Based on the total pebble mass in the reactor core and the cross section of the pebble bed, the load with respect to the area can be determined for the bottom pebble row as:

$$\widehat{m}_{core} = \frac{m_{core}}{A_{core}} \tag{2}$$

In order to determine the load on a single pebble in a specific horizontal layer, the number of pebbles in this layer needs to be estimated. The following calculation is based on a simplified methodology after Groemer [16]. The schematic diagram in figure 3 is intended to improve the understanding of the following equations.

Since the pebbles are small in relation to the core diameter, the distance between pebble rows is approximately

$$\nu = \frac{\sqrt{3}}{2} \cdot d_{pebble} \tag{3}$$

In a linear pebble row, the number of pebbles, n_{row} , is simply the length of the row, s, divided by the diameter of the pebble, $n_{row} = s/d$. The number x of rows in a square area of length s and width s accounts for:

$$x = \frac{s-d}{\nu} + 1 \tag{4}$$

Based on the combination of both equations (3) and (4), the number of pebbles per area can be estimated as:

$$N = \frac{x \cdot (2 \cdot y - 1) + 1}{\nu} \tag{5}$$

where y is the number of pebbles per horizontal layer. Accordingly, the upper limit for the normal force onto a pebble within the bottom layer (assuming the reflector walls do not contribute to balancing the gravitational force) can be determined as:

$$F_{pebble} = \frac{\widehat{m} \cdot g}{N} \tag{6}$$



Figure 3: schematic diagram of the arrangement of the pebbles in a horizontal array

In the case of the HTR-PM with a core diameter of 3 m, this simplified calculation yields about 1980 pebbles per horizontal layer in the pebble bed. Considering the very long core with a height of 11 m, the resulting load per pebble of 44.3 kg can be assumed to be a very conservative estimate for the upper limit, meaning that the highest value for the contact force in a real pebble bed will be lower than 430 N. The most important design parameters of the HTR-PM are given in table 2. Additionally, the upper limit for the load and contact force for a specific pebble as a function of its vertical position in the pebble bed is given in figure 4.

Table 2: Design and calculated parameters of the HTR-PM [15]

Design parameters		
$H_{core} = 11 \text{ m}$	$D_{core} = 3 m$	$ ho_{graphit} = 1799 \text{ kg/m}^3$
$d_{pebble} = 0.06 m$	ε = 0.377 [14]	
Calculated parameters		





Figure 4: Upper limit for the load on pebbles as function of position in the core (conservative assumption: gravitational force is not balanced by vertical reflector walls

2.2.2 Calculation considering the friction between pebbles and reflector walls balancing a fraction of the gravitational force

A more precise value for the load acting on a pebble in the pebble bed can be derived if using the relation of Janssen [17] which takes into account that for non-ideal pebbles with a friction coefficient $\mu > 0$ a significant fraction of the load can be balanced by the side reflector:

$$p_{\nu}(z) = D_{core} \cdot \frac{\rho_{core} \cdot g - a}{4\mu_{wall} \cdot k} \cdot \left[1 - exp\left(\frac{-4\mu_{wall} \cdot k \cdot z}{D_{core}}\right)\right]$$
(7)

where p_v is the vertical pressure in the pebble bed, D_{core} is the diameter of the reactor core; a is the pressure drop of helium coolant; z is the depth of the pebble bed, μ_{wall} is the friction coefficient at the reflector wall and the pebbles; ρ_{core} is the density of the pebble bed. The factor k is determined as follows:

$$k = \frac{\sqrt{1 + \mu_{pebble}^2 - \mu_{pebble}}}{\sqrt{1 + \mu_{pebble}^2} + \mu_{pebble}}$$
(8)

Based on equation (7), the average force acting on each pebble can be calculated as a function of its position in the reactor core:

$$F_{\nu}(z) \approx \frac{\pi \cdot d_{pebble}^2 \cdot \int_0^{\tilde{z}} p_{\nu}(z) dz}{4 \cdot \int_0^{\tilde{z}} dz}$$
(9)

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$$= D_{core} \cdot \frac{\pi \cdot d_{pebble} \cdot (\rho_{core} \cdot g - a)}{16\mu_{wall} \cdot k} - D_{core}^{2} \cdot \frac{\pi \cdot d_{pebble}^{2}(\rho_{core} \cdot g - a)}{64 \cdot z \cdot \mu_{wall}^{2} \cdot k^{2}} \\ \cdot \left[1 - exp\left(\frac{-4\mu_{wall} \cdot k \cdot z}{D_{core}}\right)\right]$$

Calculation results based on different friction coefficients and the assumption $\mu_{wall} = \mu_{pebble}$ are shown in figure 5. The case of $\mu=0$ reflects the conservative case where the fraction of the horizontal force component balanced by the reflector wall is zero. Therefore, equations (6) and (9) yield the same values in the case of $\mu=0$. However, for $\mu\neq0$, a significant fraction of the horizontal force is transmitted to the reflector wall, increasing with increasing friction coefficient. At $\mu=0.4$, a value typical for the graphite grade used for these investigations [18], the normal force acting on a single pebble is less than half the conservative value. Above $\mu=0.4$ however, no further significant decrease can be observed.



Figure 5: Calculation of the force acting on a pebble taking into account friction between pebbles and reflector walls according to [13]

In order to investigate the influence of a variation of the contact force on amount and characteristics of generated particles, the design of the pebble mill allows for an incremental variation of the total load between 1 and 60 kg. Optionally, experiments with up to 90 kg can be performed if necessary. The guide shown in figure 6 is designed to allow for a smooth guidance of the load ruling out any tilting.



Figure 6: Model of pebble abrasion mechanism (p-p-p-configuration) with incrementally adjustable load (steel plates)

The relative movement of the pebbles among each other and between the pebbles and the wall can be varied by controlling the rotational speed of the actuator by a frequency converter. The applicable rotational speed is limited by the gearing to a range between 10 and 100 min⁻¹. Compared to other work [11], [12], this yields a relatively slow relative velocity implying a low particle yield. However, it has to be taken into account that the actual relative velocities occurring between pebbles in a reactor pebble bed are significantly lower, even compared to the low velocities chosen for this study. Therefore, an important result of the experiments will be the observation to which extent the absolute value of the relative velocity affects particle characteristics. In the case of high interdependence, a lower velocity should be chosen in future experiments in order to reproduce the particles generated in a reactor as accurately as possible. In the case of no obvious relation between relative velocity and particle characteristics, on the other hand, the velocity could even be increased to obtain a higher dust particle yield.

2.3 Design details

In order to fix the original pebbles on a support in the pebble mill, they are cut into half pebbles with 8 holes on the flat surface as shown in Figure 7.



Figure 7: Manufactured half pebbles to be used in the pebble mill

Owing to the 8 holes, 8 different positions for each half pebble on the support can be chosen, allowing for 8 different contact points in the ppp-contact configuration and therefore a very efficient usage of the available pebbles. The original surface of one pebble can be used in 8 subsequent experiments. In figure 8, the horizontal fixation mechanism of a half pebble onto the support is

illustrated. While the cover plate fixes each pebble horizontally preventing it to become loose, the pins of the support sticking into the holes in the half pebble prevent it from rotating.



Figure 8: Horizontal fixation mechanism of pebbles in the pebble mill

In order to investigate the impact of different atmospheres, i. e. of the oxygen content of the surrounding gas, the pebble mill is surrounded by an acrylic gas-tight vessel. This vessel is fixed onto the floor of the pebble mill and can be flooded with different atmospheres through respective valves. Basic experiments will be conducted in helium atmosphere. The test facility with surrounding vessel is shown in figure 9. Helium flow through the cylinder is made possible by small holes in the bottom part of the cylinder as shown in detail in figure 6.



Figure 9: Completely assembled pebble mill (model)

The vessel can be easily installed and removed and allows for visual observation of ongoing experiments. Due to very different results in previous experiments, the size classification of

generated particles cannot be estimated at this point. Results of the investigations in [5-7] and [12] indicate that mainly nano particles will be generated whose transport characteristics are not governed by gravity. In order to collect these particles, the interacting pebbles are therefore surrounded by an acrylic cylinder which will be charged electrically, attracting the particles. The electrical charge is expected as a result of the friction between the rotating cylinder (fixed on the bottom plate) and the helium shielding gas. After each experiment, particles can be gathered by discharging the cylinder and dipping it into ethanol. After the ethanol is evaporated, photomicrographs of these collected particles can be taken. With the help of an image processing program the photomicrographs will be analyzed and the shape, size and size distribution of the collected particles can be specified.

2.4 Choice of the electric motor

For the choice of a suitable electric motor, the following parameters need to be known:

- friction coefficient between graphite components,
- length of the lever arm between rotation center of the disc and point of contact of the grinding components,
- maximum rotational speed,
- maximum normal force due to the applied load.

As mentioned, these parameters need to be chosen with the aim of representing reactor conditions in the pebble bed as closely as possible. Values for the friction coefficient are estimated based on literature values. Experimental investigations in [10] have yielded dynamic friction coefficients among graphite components under ambient and shielding gas atmosphere in the range between 0.34 and 0.52 with a mode of μ =0.4. Friction coefficients for different graphite grades have been determined in other work and are documented in table 3.

Material	Dynamic friction coefficient	Reference	
ATJ against ATJ	0.38	[9]	
H-205 against H39	0.40	[9]	
PGX against PGX	0.41	[9]	
MHLM against MHLM	0.38	[9]	
AXF-50Q against AXF-50Q	0.52	[19]	
Polycrystal against polycrystal	0.38	[20]	
Lanzhou graphite against itself	0.34	[21]	
Shanghai Sangao graphite against itself	0.43	[21]	
IG-11 against IG-11	0.40	[21]	

Table 3: Measured friction coefficient of different graphite grade	icient of different graphite grades
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Based on data given in table 3, a value of μ =0.5 has been assumed for the friction coefficient, subsequently multiplied by a safety factor of F_S=1.5. Thus, the required motor power can be calculated by:

$$P_M = M_t \cdot \omega = 2\pi \cdot n \cdot M_t \tag{10}$$

with
$$M_t = F_N \cdot \mu \cdot l \cdot F_S = m_N \cdot g \cdot \mu \cdot l \cdot F_S$$
 (11)

Given a maximum rotational speed of n=100 min⁻¹, a load of $F_N=900$ N, a safety factor of $F_S=1.5$ and a lever arm of l=30 mm, a minimum motor power of 210 W is required. In order to maintain sufficient reserves, a motor with an electric power of 250 W has been chosen. The low rotational speed of the pebble mill is obtained using a separate worm gear and the frequency is controlled using a frequency converter.

3 **Experimental Investigations**

In order to estimate the amount of particles generated by the pebble mill through abrasion, the wear rate K_{ad} needs to be estimated first. Several experimental studies have been carried out to determine this value for different graphite grades and under different conditions. A summary of these studies can be found in [13] (figure 10). For the pebble mill used in this study, these values cannot be consulted without further ado since these values significantly depend on operating conditions such as atmosphere, temperature and, above all, graphite grade. For different conditions, values for K_{ad} found in the literature vary by several orders of magnitude. Even for experiments at room temperature severe differences in Kad for different graphite grades are observed (Table 4).



Figure 10: Wear rates of different graphite grades as a function of temperature [13]

Table 4: wear	rates in helium and room temperature	
Material	Notes	Wear coe

Material	Notes	Wear coefficient	Reference
		[x 10 ⁻⁶ g/(N⋅m)]	
MHLM	25 °C, helium	1.50	[9]
MHLM	400 °C and 800 °C, helium	0.15	[9]
KG-11	31 N; air, room temp., line contact	1.02	[12]
IG-11	30 N; helium; room temp., average	0.11	[11]
IG-11	30 N; helium; room temp.; first 30 min	0.37	[11]
IG-11	30 N; helium, 400 °C, average	0.53	[11]

For the performance estimation of the pebble mill only the known wear rates determined at room temperature can be considered since K_{ad} is highly dependent on the operating temperature [11]. Therefore, a range between 0.1 and 1.5×10^{-6} g/(Nm) can be considered as a first estimate. The rate of generated dust particles can be estimated using the following relation.

$$\dot{m}_{wear} = K_{ab} \cdot F_N \cdot L_{contact} \cdot n \tag{12}$$

with
$$L_{contact} = 2 \cdot \pi \cdot \frac{d_{pebble}}{4}$$
 (13)

with F_N being the normal force at the point of contact, L_{contact} the distance covered by the grinding components per one complete rotation and n is the rotational speed. In the case of ppp-contact, the horizontal distance between point of contact and pebble is equal to d_{pebble}/4 (figure 11).



Figure 11: Pebble configuration

The estimated rate of generated dust according to equation (12) is shown in figure 12 per point of contact as a function of the contact force at a rotational velocity of 100 min⁻¹.



Figure 12: Range of expected dust production rate attainable with the pebble mill

Due to the high spreading in literature data, only a range of values for \dot{m}_{wear} can be estimated as shown in figure 12. The values in the order of mg/min emphasize however, that most likely only a very small total amount of graphite dust can be generated with this facility. This will be sufficient for the characterization of generated particles (which is the objective of this work), but not for the supply of large-scale experiments requiring large amounts of generic graphite dust. Therefore, it is suggested that based on the characterization of particles (objective of subsequent tasks), a commercially available graphite dust grade will be identified which of the characteristics should be as close as possible to those generated in the pebble mill. For graphite dust transport simulations, e. g. with CFD codes, the characterization results can be readily implemented.

4 Summary

Objectives of task 2.3.2, "Graphite Dust Production and Deposition Behaviour", are:

- Establishing a small generic test facility (pebble mill) for generation of graphite dust particles through abrasion of moved pebble fuel elements.
- Characterizing generated particles as a function of abrasion conditions in order to identify a commercial graphite dust grade appropriate for large-scale graphite dust transport experiments and as input for dust transport simulations, e. g. using CFD codes.
- Using the identified commercial graphite dust in a small He-test loop to investigate the deposition behaviour at flow obstacles.

This deliverable is concerned with the first objective, forming the basis for future work within this task. Main objective of the design of the pebble mill has been to establish very flexible operation procedures in order to vary as many parameters as possible (atmosphere, type of contact, load at the point of contact and graphite grade). The rate of dust particles to be generated is estimated to be sufficient for characterization but not for supplying large-scale experiments. Characterization of generated particles is objective of the next deliverable due within this task.

5 Annexes

Annex 1 – Document approval by beneficiaries' internal QA

Annex 2 – Literature

Annex 3 - Nomenclature

5.1 Annex 1: Document approval by beneficiaries' internal QA

#	Name of beneficiary	Approved by	Function	Date
1	TU Dresden	Wolfgang Lippmann	Task Responsible	30/09/12

5.2 <u>Annex 2: Literature</u>

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symbol	unit	account
а	Ра	pressure drop of helium coolant in the reactor core
d	mm	diameter
D	m	diameter
F	Ν	force
Н	m	height
К	mg / m	wear rate
I	mm	crank of a lever
m	kg	mass
în	kg / m²	distributed load
ṁ	mg / min	mass flow
Μ	Nm	torque
n	1 / min	rotation speed
Ν		number of pebbles per unit area
р	Ра	pressure
Р	kW	power
S	m	length
V	m ³	volume
z	m	vertical position in the pebble bed
3		void fraction
μ		friction coefficient
v	mm	distance between two circles in the densest sphere packing
ρ	kg / m³	density

5.3 Annex 3: Nomenclature