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Justification of Tungsten Particles Production Estimate

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1. SUMMARY

This document will justify the assumption of the particle amount, produced by the tungsten and the tungsten cassette, used in the Target System design and the Hazard Analysis.

In the TDR it is assumed that "The tungsten target is prone to ablation and dust formation". The document states a reasonable upper limit ablation rate of 10 µm/y. If the ablation rate is assumed to be equal on the entire tungsten surface the TDR design will produce 3-4 kg of dust per year.

The results from a number of EDD's (Engineering Development and Demonstration) and calculations, performed during the Concept Design of the Target System, question the TDR assumption. Taking the EDD's and the calculations into account a more realistic, but still conservative, assumption of dust formation is suggested to be 10 g/y.

Based on the EDD's and the calculation's presented in this document, a dust formation rate of 10 g/y will be used during the Final Design of the Target System.

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2. TDR ASSUMPTION

TDR chapter 10.4.1:

> "The tungsten target is prone to ablation and dust formation. An ablation rate of 10 µm /y of all surfaces is a reasonable upper limit for target deterioration [648,649]. This amounts to 0.07% of the target mass becoming dust per year, or roughly 3 kg/y to 4 kg/y."

TDR Reference 648 "Filtering System for the potential tungsten Dust" [1] describes the possibility to filter tungsten particles out from a Helium flow. The number of the dust production rate is not explained in this document.

TDR Reference 649 "Dust borne activities in gas-cooled spallation sources, experience from gas cooled reactors and from fusion devices" [2]. This document shortly mention the dust production:

> "Dust production in gas-cooled reactors is different from spallation sources: Mostly friction is responsible for dust formation, although to some extent decomposition of oil (lubricant in blowers) and chemical reactions (ingressing air into an He-circuit) are known to be sources for dust. Hopefully these sources can be excluded in a He-cooled W-target system. It should however be noted that air or steam impurities in such a He-cooled target system definitively would induce formation of tungsten oxide dusts. Neutron embrittlement may also facilitate dust formation, but there are no data available on that matter. Concerning friction we have to bear in mind that in pure He-systems friction is usually very high. For that in gas-cooled reactors a very small amount of oxidizing impurities was added to the Helium. By that measure the vanishing of self-protecting oxide layers on metal surfaces (SS) was avoided and their friction behaviour was improved. It is not clear whether this is required for tungsten-based systems, too, because chemical destruction of oxide layers is mainly a high temperature problem (> 500°C). On the other hand, destruction of oxide layers by friction/erosion will occur, too and in this case sufficient oxidizing media have to be present in order to heal the wounded metal surfaces. However, addition of oxidizing gases will lead in any case to a corrosion of W.
>
> Dust production in He-cooled target systems will probably be more similar to the mechanisms of dust formation in fusion devices: Here the alpha-particles bumping on the surface are probably the main reason for the formation of tungsten dust. Quantitative knowledge on W-dust formation in fusion devices is still limited: In safety analyses performed for Iter the dust production rate was taken as parameter (total dust amount: 0.1-100 kg). Perhaps such a procedure is adequate for ESSS, too, although the assumption of an eroded layer by the beam of about some 10 µm seems to be not unreasonable."

The assumed eroded layer of 10 µm mentioned above is without any time dimensions.

Some source of knowledge, mentioned in the references, discuss the dust formation in pebble bed reactors. Focus in these documents is the abrasion both between pebbles within the core, and between pebbles and the fuel handling system. However, the tungsten bricks in the target vessel are structurally secured in the target cassettes and abrasion between tungsten bricks is not relevant in the Target Wheel.

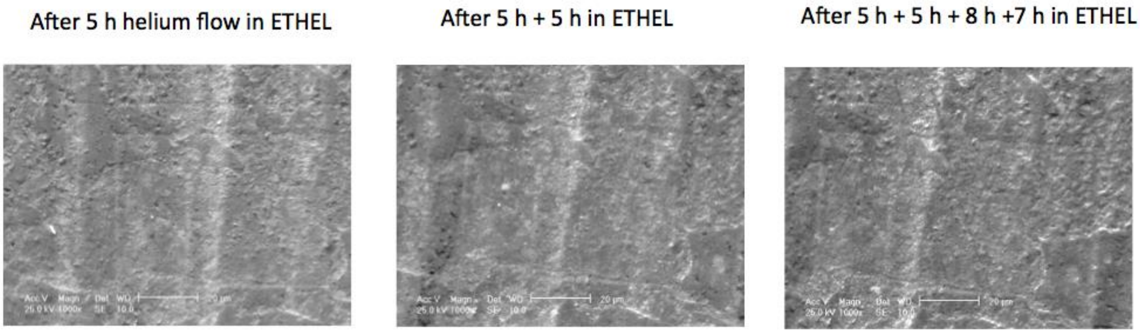
It is hard from the references mentioned in the TDR to verify the ablation rate of 10 µm/y. The TDR assumption could perhaps be considered as a figure that in any operational circumstances, in the ESS facility, will not be exceeded. Based on the references mentioned in the TDR the dust formation rate of 3-4 kg/y is a very conservative assumption

3. ETHEL

To understand if the helium cooling flow in the target wheel will have any impact on the oxide layer of the tungsten surface, and by that produce dust particles, a test equipment was introduced at LTH. Two samples were pre-oxidised at 500° C in He + 0.5 % O2 for 1 h. The experiment set up was a Helium jet of > 100 m/s at 9 bar and > 200° C blown perpendicularly onto the tungsten samples at above 300° C [3].

Scanning Electron Microscope (SEM) pictures, $0.01*10^{-6}$ m^2, taken at five positions on the samples after 5, 10 and 25 hour

s. Based on these pictures the conclusion is that the experiment could not reveal any dust formation what so ever with in the experiment basis. The erosion rate is by that 0 µm in this experiment. See figure 1


After 5 h helium flow in ETHEL

After 5 h + 5 h in ETHEL

After 5 h + 5 h + 8 h +7 h in ETHEL

Figure 1 ETHEL Pictures

Looking at the picture some protruding features are recognised. The features are estimated to be $(5\ \mu m)^3 = 125\ \mu m^3$. Some pictures have about 10 of these features. If you assume that this is the surface structure in general, and to make a conservative estimation, all of them are in some way eroded from the surface you can estimate a dust formation. These estimates will give an erosion amount of about 0,1 kg.

The calculation is :

> Features volume x tungsten density x total tungsten surface / SEM surfce
> $1250*10^{-18}$ m^3 * 19300 kg/m^3 * 43 m2 / $0.01*10^{-6}$ m^2 ~ 0.1 kg.

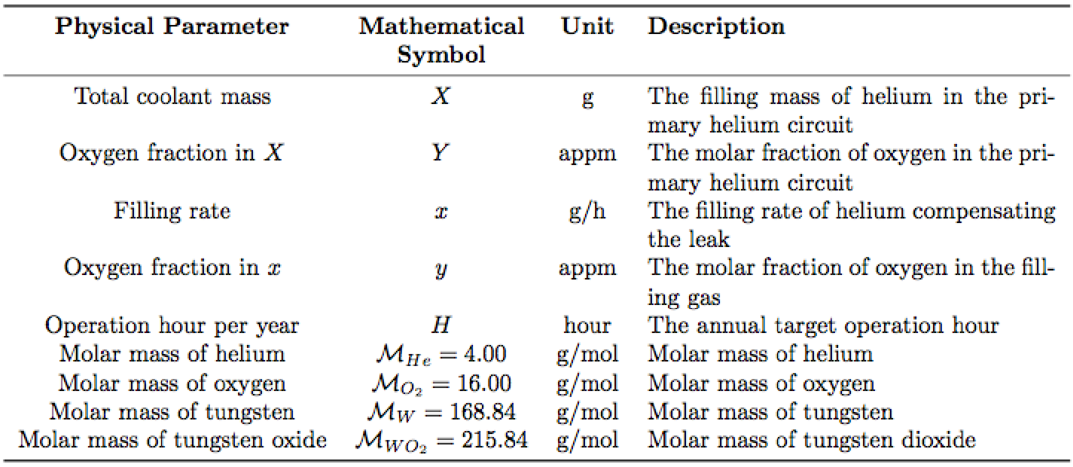
The test samples are pre-oxidized in 500°C He with 0,5% oxygen during 1h. Compared to the steady state operational condition this is far beyond the oxygen requirements in the Helium Cooling Loop. During operation the Helium is purified to 0,00001% (10 ppmV) oxygen. Taking this level into account the pre-oxidized test samples roughly could be compared to a five-year in operation tungsten brick. The 0.1 kg of particles derived from the eroded features is comparable to 20 g/y.

4. MAXIMUM OXIDE FORMATION

The ETHEL experiment could not display any erosion from the surface, neither from the tungsten it self nor the tungsten oxides on the surface. To still make a conservative estimation of the dust production the hypothesis is that all the oxides will in some way leave the tungsten surface and produce dust [4].

The tungsten will be operated in helium with a restricted amount of oxygen impurities. The following calculation assumes that all the oxygen is used for oxidisation of the tungsten surface, which also is a conservative assumption.

The physical parameters oxide formation calculation is listed in table 1.



Physical Parameter	**Mathematical Symbol**	**Unit**	**Description**
Total coolant mass	X	g	The filling mass of helium in the primary helium circuit
Oxygen fraction in X	Y	appm	The molar fraction of oxygen in the primary helium circuit
Filling rate	x	g/h	The filling rate of helium compensating the leak
Oxygen fraction in x	y	appm	The molar fraction of oxygen in the filling gas
Operation hour per year	H	hour	The annual target operation hour
Molar mass of helium	$\mathcal{M}_{He} = 4.00$	g/mol	Molar mass of helium
Molar mass of oxygen	$\mathcal{M}_{O_2} = 16.00$	g/mol	Molar mass of oxygen
Molar mass of tungsten	$\mathcal{M}_{W} = 168.84$	g/mol	Molar mass of tungsten
Molar mass of tungsten oxide	$\mathcal{M}_{WO_2} = 215.84$	g/mol	Molar mass of tungsten dioxide

Table 1

Formula used in the oxide calculation:

We consider a situation where all the oxygen contents supplied to the primary helium cooling circuit is used for oxidizing the tungsten blocks in the target wheel and the tungsten oxide layer so formed is made of tungsten dioxide. In this case, the total molar quantity of oxygen that is used for oxidation per operational year is given by

$$\Lambda_{O_2} = 10^{-6} \cdot \frac{(X \cdot Y + x \cdot H \cdot y)}{\mathcal{M}_{He}} \quad [\mathrm{mol}]. \tag{13}$$

The annual production of tungsten oxide layer in the target wheel is then given by the equation below,

$$M_{WO_2} = \mathcal{M}_{WO_2} \cdot \Lambda_{O_2} \quad [\mathrm{g}]. \tag{14}$$

With a conservative assumption that all the tungsten oxide layer is eroded away by the helium flow, the total loss of tungsten per operational year is estimated by the equation below,

$$M_W = \mathcal{M}_W \cdot \Lambda_{O_2} \quad [\mathrm{g}]. \tag{15}$$

For different oxygen impurity level, the annual production of tungsten loss from target wheel is listed in table 2.

Filling mass X [g]	**Filling rate** x [g/h]	O_2 **impurity** Y [appm]	O_2 **impurity** y [appm]	**Annual beam on target hours** [h]	**Annual W loss** [g/y]
$3.0 \cdot 10^4$	10.0	10.0	10.0	5400	38.6
$3.0 \cdot 10^4$	5.0	5.0	5.0	5400	13.1
$3.0 \cdot 10^4$	1.0	5.0	5.0	5400	8.14

Table 2

In the absence of a helium purification system, the pure industrial helium typically has an oxygen impurity level of 5 appm, and the estimated leak rate of the primary cooling loop is 1 g/h. Under the ESS helium coolant loop conditions, the release of tungsten via the erosion of the oxide layer is estimated to be 10 g/year, even in the absence of purification system.

The estimated 10 g/y correspond to an erosion rate of 0.01 µm/y. This is a considerably lower number than the number assumed in the TDR (10 µm/y). The number given in the TDR is however not taken into account the very low oxygen amount in the Helium Loop as in the calculation above.

If the hypothesis is correct that there will just be dust formation coming from the oxide layer, the calculated 10 g/y seems reasonable and still conservative.

5. VIBRATION TEST

ESS Bilbao has performed a vibration test [5]. The test assembly is a reconstruction of a part of the cassette, including 11 tungsten bricks. The bricks are placed in the test assembly with the same tolerance as in the originally designed cassette. The test assembly was placed on a vibration table. The following test data were used: 500 Hz – 1 hour – 1G - amplitude 0.1 mm.

Scanning Electron Microscopy (SEM) images was taken on two bricks (fig 2 and 3). Left picture is before and right is after vibration procedure.

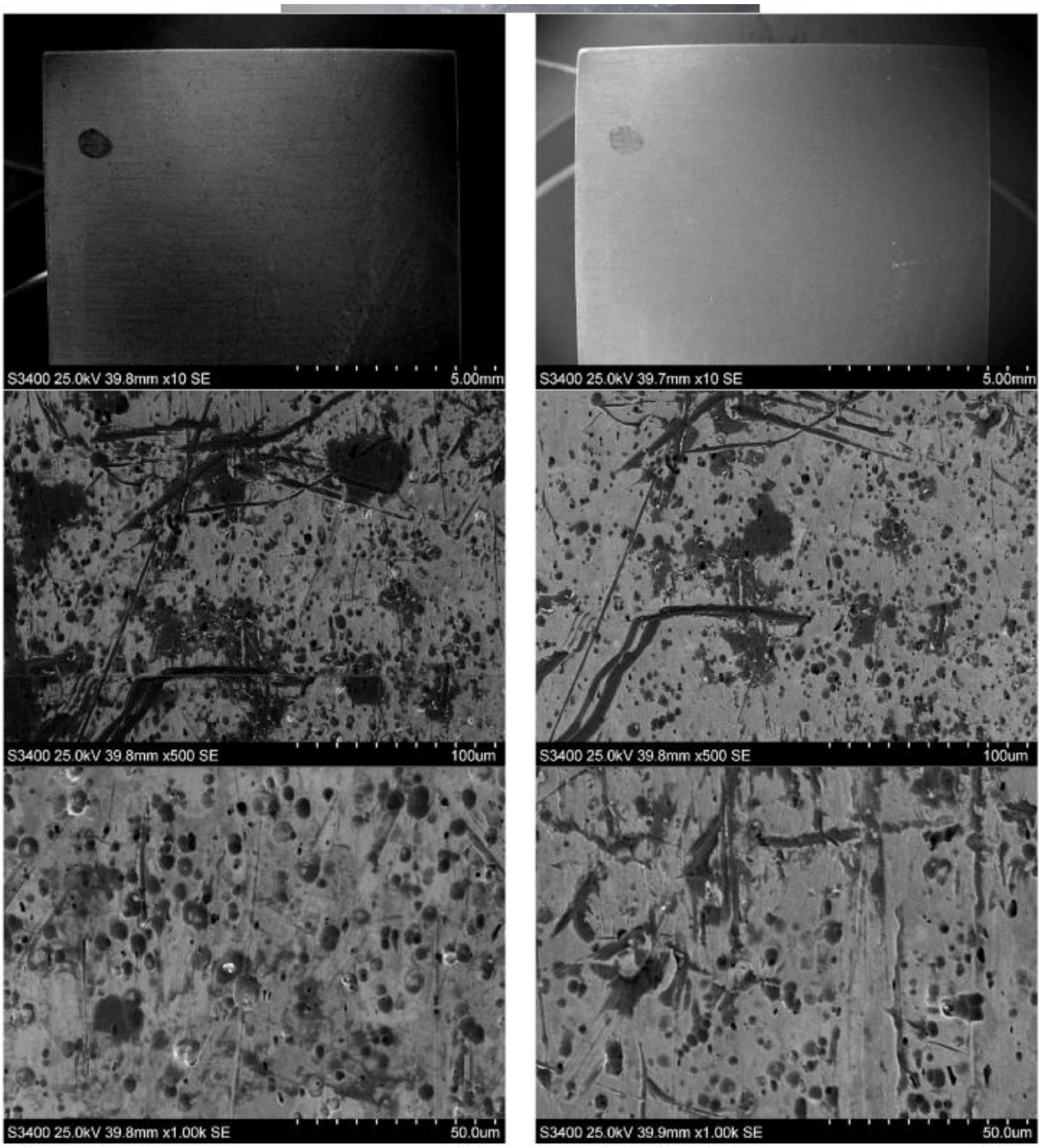


Fig 2 Vibration test sampl 1

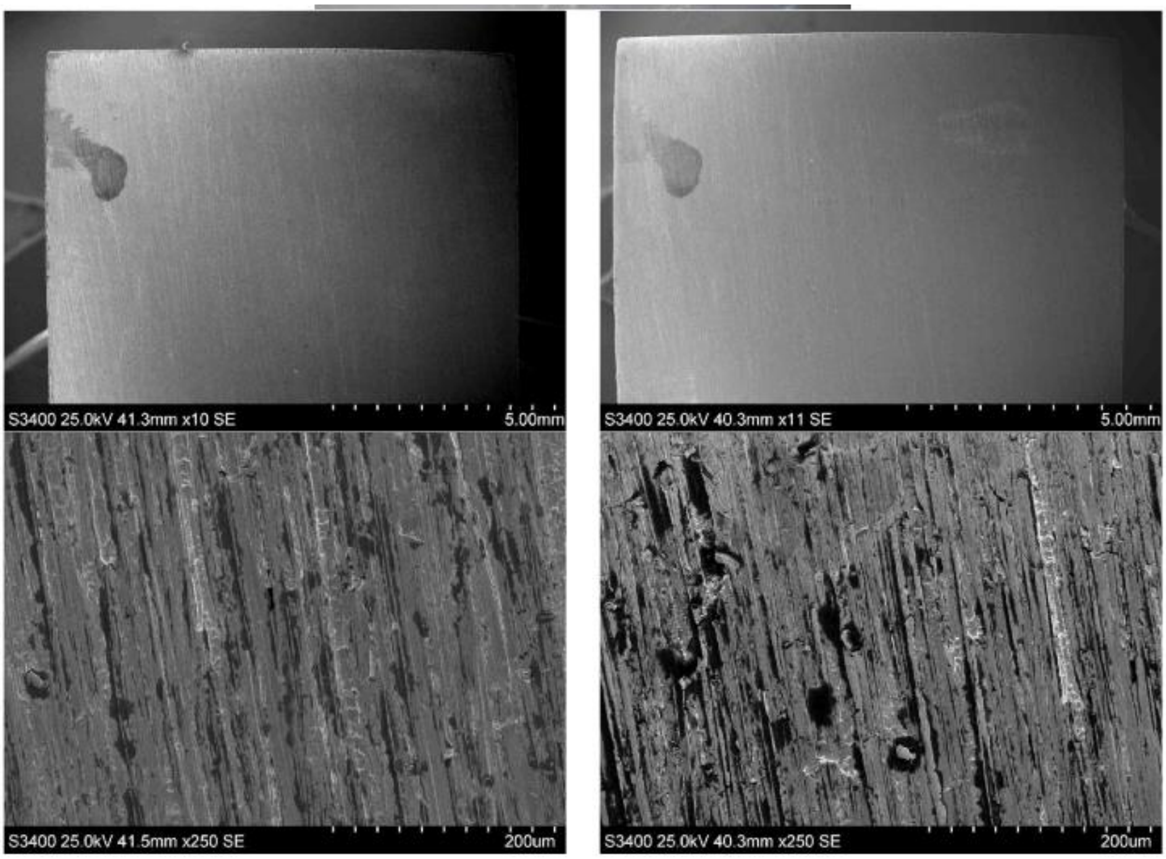


Fig 3 Vibration test sample 2

The main conclusion in the report is that there is no significant evidence of surface modification and/or degradation due to the vibration test.

During dismantling of the test assembly some debris were collected. The debris were weighed and analysed by Energy Dispersive X-Ray Fluorescence (EDXRF) equipment. The weight was 0.0364 mg and the EDXRF could not determine any tungsten particles in the debris and that all of the debris comes from the stainless steel cassette that holds the tungsten bricks. The report notices that some grains could have been lost during manipulation.

The test assembly included 11 bricks. The final Target Wheel will include about 7000 bricks. If the amount of dust from the test assembly is extrapolated to the 7000 bricks the amount of dust could be estimated to 23 mg.

How this result could be applied to justify the dust production in the cassette during normal operation is hard. The number of excitations in the vibration test is 2x10^6 (500Hz x 3600 s). As comparison, the number of proton beam pulses during one year of operation is 10x10^6 ((14 Hz / 36 cassettes) x 5400 h x 3600 s). If you assume that the Helium flow won't induce any vibrations the number of excitations in the vibration test are in same order of magnitude as the number of proton beam pulses. You could then assume that the amount of particles produced by mechanical wear is in the order of 10-100 mg, which is far below the suggested dust production of 10 g/y. However, the result indicates strongly that there won't be any production of pure tungsten particles. If there are any particles coming from mechanical wear it will be material from the cassette.

6. IMPACT FROM 5MW 2GEV PROTON BEAM

The impact on the tungsten from a 5 MW 2Gev proton beam could be analysed in a number of different cases. In this document thermal elongation and sputtering are discussed.

6.1. Impact from elongation

The thermal elongation is in the range of 0.01 mm. This is to the most heated bricks in the middle of the cassette. The proton beam hits the tungsten bricks with 0,39 Hz (14Hz/36 sectors). The number of elongation through one year is 9.7x10^6 (0.5 Hz x 3600s/h x 5400h of operation/year). This could be compared to the vibration test performed by ESS Bilbao of 1.8x10^6 (500Hz x 1h). However, the elongation of the bricks is much less then the tolerance between the brick and the brick holder (H9/h9 => max 0.1 mm) in the test assembly, and the max elongation is just accurate for a part of the total number of the bricks. Taking this into account the production of dust will be less than 23 mg.

6.2. Impact from beam pulse induced sputtering

In order to investigate the surface sputtering induced by beam pulse, pure and oxidized tungsten specimens have been irradiated by pulsed uranium beam at GSI. Specifically, a pulsed uranium beam with 4.8 MeV/nucleon, 150 µs pulse length, 1-2 Hz repetition rate and the particle flux 1.0e10 particles/cm^2/pulse has been used. This uranium beam deposits heat within 20 µm from the surface depth and heats up the tungsten sample temperature to the operational temperature range of the ESS target. The beam stopping power creates a thermal shear stress of more than 200 MPa on the surface, which is about factor 2 higher than the maximum stress applied to the tungsten blocks of the ESS target. With the uranium beam irradiation, the total fluence of 1.0e14 particles/cm^2 or 10000 pulses have been applied. With this maximum fluence level applied, the maximum dpa reached in the tungsten is about 0.05. The SEM image comparison of the irradiated tungsten surface has shown no noticeable change. Also no sputtering damage has been observed from oxidized tungsten samples under the equivalent irradiation conditions. This indicates that it is not likely to have a tungsten particle release from the tungsten blocks in the ESS target, due to the surface sputtering by the pulsed 5 MW beam.

7. HELIUM FLOW IMPACT ON PARTICLES

Radioactive particles will have an impact on dose to public and workers only if the particles leave the Target Wheel and Shaft. To do this the particles size and the Helium Flow have to be within some limits to be able to transport the particles through the vertical shaft. The assumed particles will be tungsten oxide not pure tungsten. The oxide has a much lower density than the pure tungsten and in the suggested conservative assumption in this document we assume that all particles will leave the Target Wheel and Shaft and are trapped by mechanical filters in the loop or deposited on internal surfaces of loop components where flow conditions allow (e.g., recirculation zones).

8. DOSE LIMITS

8.1. Hazard analysis

In the hazard analysis the radiation inventory calculated in [6] is used. In the calculation the total amount of contaminated Helium gas is 30 kg with 10 g/y particulates. The inventory calculated from these numbers is 3.12 10^{12} Bq (gases and smallest particles not captured in filters) and 1.47 10^{12} Bq (particles in filters). The dose outcome from this inventory during a mitigated event is 8 mSv/2 min to workers and 0.32 mSv/event to the public. During normal operation the dose to workers is 18 µSv/4h and dose to the public is 0.004 µSv/y.

If the number of particulates is one order of magnitude higher (100g/y) the radioactive inventory in the filters will rise to 2.4 10^{12} Bq and the dose to workers is calculated to be 11 mSv/2 min. This is close to the figures for 10g/y. The dose from erosion will not have any major impact to the total dose until the erosion is closer to 1 kg/y. Until then the major part of the dose comes from diffusion and ejection.

8.2. Maintenance

The different rooms where the Helium Cooling System is installed will be closed during operation. During maintenance period the rooms will be opened up for access of the system. The system will be designed to avoid hot spots but a general contamination is foreseen. This will be handled by normal preventive radioactive working protocol.

However, the filter room will be of main concern regarding radiation to workers. A preliminary calculation taking into account 5% of the most radioactive nuclides in the filters (about 10% of the radioactive inventory) will produce 0.8 Sv/h standing 1 m away from the filters. Assuming this is 10% of the total dose you can estimate a total dose to 8 Sv/h. These figures needs to be confirmed. In any way shielding and remote handling of filters are foreseen.

The dose will be measured on line and operation stopped if the stipulated limit is exceeded.

9. WHAT IF THE PARTICLE ESTIMATION IS NOT CONSERVATIVE?

The assumptions used in chapters 3, 4 and 6 can be considered conservative. However, the calculations are based on current knowledge and the operations might disclose facts not included in the performed calculations. If the operation of the Target System will produce particles far beyond the assumed 10 g/y this will be captured by the radiation measuring system of the filters. The filters then need to be changed out more frequently to fulfil the maximum radiation inventory used in the hazard analysis. Preliminary calculation, table 3, indicates that if the particle production is 100 g/y we still just need to replace the filters once per year. If the particle production is 1000 g/y we probably need to revise the replacement frequency. The particles are highly radioactive and even with the assumed number of 10 g/y the filter exchange have to be performed by remote handling.

10g/y	**100g/y**	**1000g/y**	
9,40E+04	1,30E+05	4,60E+05	mSv/4h; Unmitigated dose to person in rooms
3,70E+04	5,00E+04	1,80E+05	mSv/4h; Unmitigated dose to person in ADJACENT rooms

Table 3 The dose values are based on the amount of isotopes produced in the target during one year of operation, accumulated and decayed in the filters. Isotopes with short half-life, e.g. < 5 days affects the dose to a limited extent even if eroded 100 times more. As seen in the table above, the total dose is appr. 5 times higher with 1000g/y compared to 10g/y

10. CONCLUSION

The TDR assumption of 3-4 kg/y particle and dust production seems to be overestimated. This very conservative number will put the design of the Target Helium Cooling System under stress and make the system solution and maintenance preparation more complicated and expensive than necessary. The dust will be highly radioactive and taking 3-4 kg/y into account this will require extensive maintenance and filter changes to keep the radioactive inventory in the loop within values of the hazard analysis.

Looking into the references mentioned in the TDR it is hard to verify the number of 3-4 kg/y. It seems like this number is a very first conservative estimation and as mentioned in one of the references “a more detailed procedure has to be started”.

The ETHEL experiment could not demonstrate any erosion at all on a pre-oxidised surface. The oxide layer that will be produced under the conditions of the Target Helium Cooling System specifications seams to be stable and will not generate any loos particles.

Vibrations test performed by ESS Bilbao strongly indicates that particles coming from the mechanical wear will be from the cassette material and not from the pure tungsten. The number of debris produced in the vibration test is extrapolated by general engineer assessment to normal operation conditions taking the proton beam pulses into account. The particle production produced by mechanical wear during the assumed circumstances is estimated to be in the order of 10-100 mg/y.

To still make a conservative assumption regarding oxidation of the tungsten surface you could calculate the possible amount of oxides formed. This layer is, in some way, assumed to be released into the helium loop. The helium gas is continuously purified and the oxygen content will be 5 appm. If all of this is used to form the oxide layer, the formation of oxide will be in the range of 8 g/y. This number of 8 g/y, taken into account that all oxides are released and all oxygen will be involved forming the oxide layer on the tungsten surface, is still conservative.

Also sputtering is discussed in the document. Based on test at GSI we don’t expect any particles produced by this phenomena.

The filters will be monitored during operation and if the upper limit of radiation is exceeded the filters will be changed to fulfil the maximum radiation inventory used in the hazard analysis.

This document suggests using a maximum particle production rate of 10 g/y in the final design of the Target Helium Cooling system and in the hazard analysis.

11.GLOSSARY

Term	Definition
<<Sample term>>	<<Sample explanation >>

12.REFERENCES

[1] C. Kharoua. `Filtering system for the potential tungsten dust.' Technical Report EDMS 1192485,European Spallation Source, 2012 (ESS-0007545)
[2] Dust borne activities in gas-cooled spallation sources, experience from gas cooled reactors and from fusion devices (ESS-0007614)
[3] Per Nilsson, ETHEL Erosion Tests (ESS-0051445)
[4] Yong Joong Lee, Integrity of spallation material in the target wheel during operation and accidents (ESS-0051742)
[5] Fernando Sordo, Vibration Test (ESS-0055645)
[6] Jens Harborn, Dose Calculations system 1010 & adjacent rooms (ESS-0044590)

DOCUMENT REVISION HISTORY

Revision	**Reason for and description of change**	**Author**	**Date**
1	First issue <<Keep only full number revisions when approving document>>	Ulf Oden	2016-06-03