Investigation of Neutron-Irradiated Beryllium Pebble Oxidation in Dry and Humid Air at Elevated Temperatures for LOVA/LOCA Estimation and Corresponding Safety Protocol Development

Rūdolfs Jānis Zabolockis[1,2], Elina Pajuste[1,2], Liga Avotina[1], Gunta Kizane[1]

[1]*University of Latvia, Institute of Chemical Physics, 1 Jelgava St., Riga, Latvia, LV-1004*
[2] *University of Latvia, Faculty of Chemistry, 1 Jelgava St., Riga, Latvia, LV-1004*

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Motivation

- Beryllium (**Be**), used as plasma facing and neutron-multiplying material in fusion reactors is exposed to neutron irradiation and temperature fluctuations.
- During extreme conditions Be can pose risks associated with the **release of fusion fuels** and toxic **beryllium compound dust formation** throughout loss of vacuum (LOVA) and loss of coolant (LOCA) accidents. [1, 2]
- **Thermal analysis** methods can be used to evaluate the processes beryllium could undergo in LOVA/LOCA conditions.

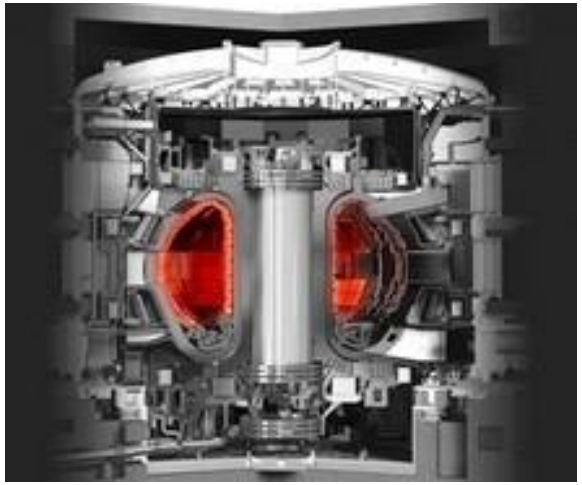

Fig. 1. ITER blanket zone [3]

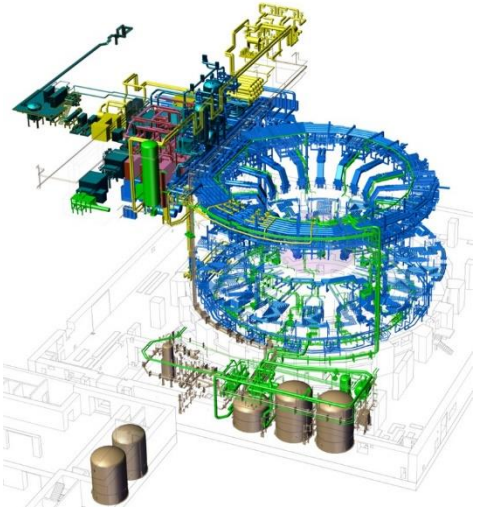

Fig. 2. ITER water cooling system [3]

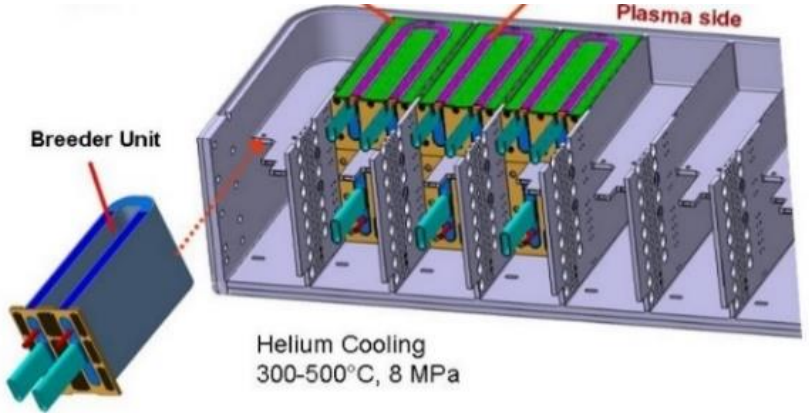

Fig. 3. Helium-cooled pebble bed tritium breeding units [4]

[1] Nakamura, M. M., *Fusion Engineering and Design*, **2018**, 136, B, 1484-1488.
[2] Ciparisse, J.-F., *Energies*, **2018**, 11, 856.
[3] ITER - the new way to energy https://www.iter.or
[4] Piazza G. et al., *Fusion Eng. Des.*, **2003**, 69, 227-231

Experimental

Samples:

1. Be pebbles, **neutron-irradiated** (E > 0,1 MeV for 294 days, fluence 3-4 · 10^{25} m^{-2}, at a temperature range from 423 to 823 K) and **non-irradiated** pebbles of same size (⌀ ~ 1mm) and grade for comparison, both produced by Rotating Electrode Process (REP) [1].

Methods:

1. Thermogravimetric/Differential Thermal Analysis (TG/DTA) with SEIKO Exstar 6300; air flow 12 L/h, heating rate 10K/min, up to 1548K.
2. Scanning electron microscopy with Hitachi S-4800 equipped with EDS system Bruker XFlash Quad 5040 123eV.

Sample analysis parameters:

- Non-irradiated
  - DRY (RH<5%)
  - HUMIDIFIED (RH>95%)
- Neutron-irradiated
  - DRY (RH<5%)
  - HUMIDIFIED (RH>95%)

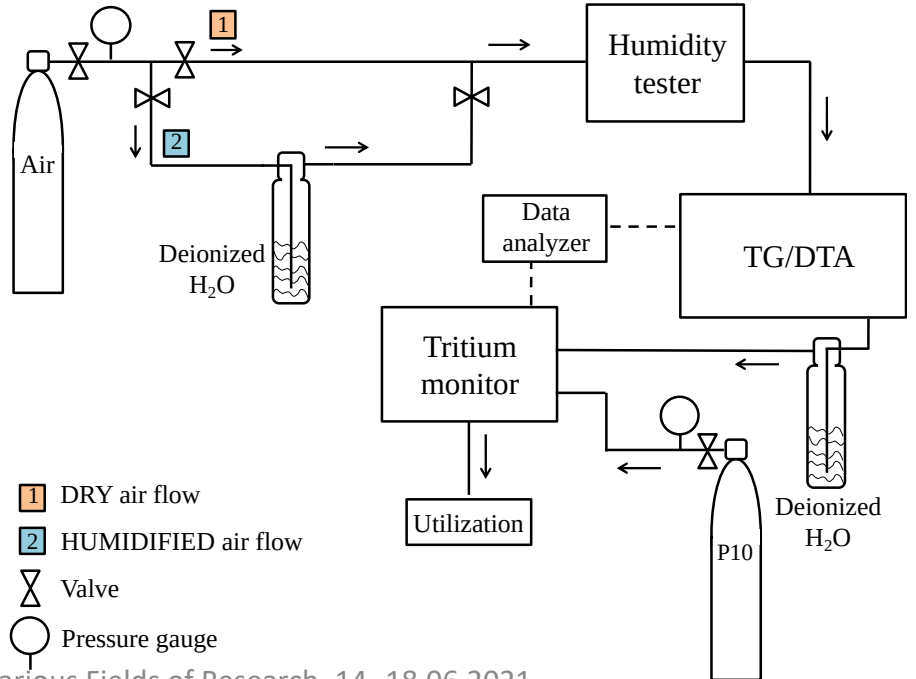

Fig. 1. Beryllium thermal treatment system with humidity control

[1] Pajuste, E., Kizane G., Avotiņa L., Zariņš A. *Journal of Nuclear Materials*, **2015**, 465, 293-300.

Results: thermogravimetry

TG/DTA shows an **increase of mass** which occurs due to the oxidation of beryllium in the presence of air and water moisture, ultimately leading to beryllium oxide formation. Most notably, neutron-irradiated pebbles in humid airflow exhibit a mass increase of almost **180%**.

$$Be(s) + 0{,}5O_2(g) \xrightarrow{temp.} \boldsymbol{BeO}(s)$$
$$Be(s) + H_2O(g) \xrightarrow{temp.} \boldsymbol{BeO}(s) + H_2(g)$$
$$3Be(s) + N_2(g) \xrightarrow{temp.} Be_3N_2(s)$$
$$Be_3N_2(g) + 1{,}5O_2(g) \xrightarrow{temp.} 3\boldsymbol{BeO}(s) + N_2(g)$$
$$0{,}5Be_3N_2(g) + 3H_2O(l) \xrightarrow{temp.} 1{,}5Be(OH)_2(s) + NH_3(g)$$
$$Be(OH)_2(s) \xrightarrow{temp.} \boldsymbol{BeO}(s) + H_2O(g)$$

Fig. 1. Chemical reactions between beryllium and corresponding compounds in air/ humid air [1, 2]

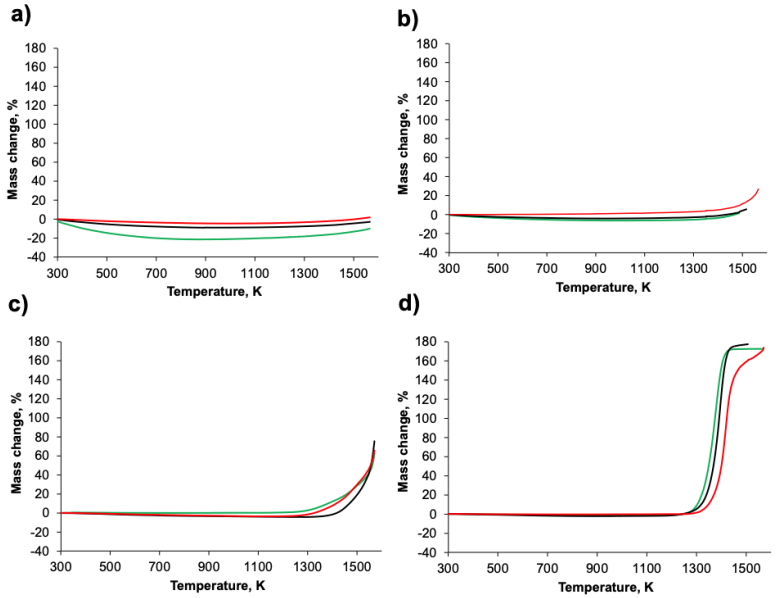

*Fig. 2. Mass change of 3 Be pebbles in: **a)** non-irradiated DRY, **b)** neutron-irradiated DRY, **c)** non-irradiated HUMID, **d)** neutron-irradiated HUMID airflow*

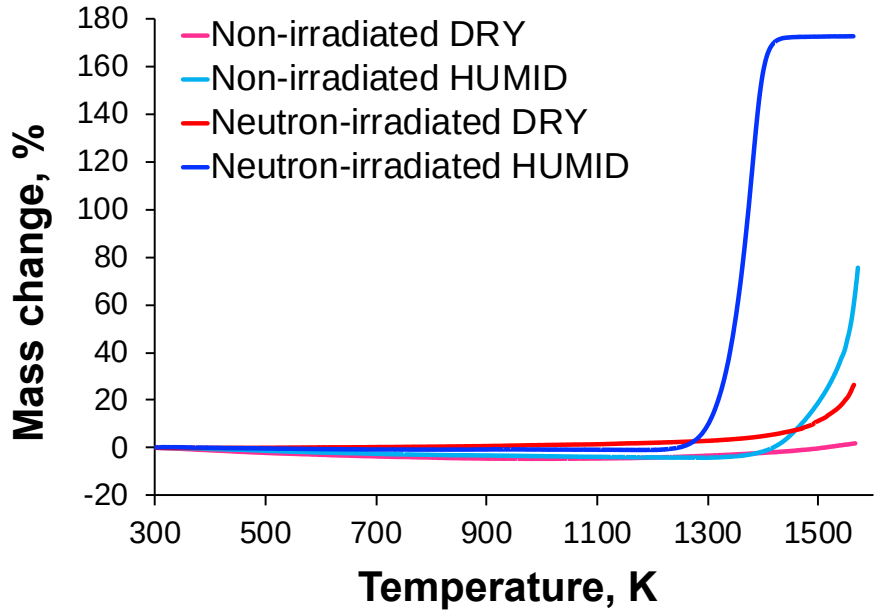

Fig. 3. Mass change of Be pebbles depending on treatment conditions

[1] Tomastik, C., *Nuclear Fusion*, **2005**, 45(9), 1061-1065.
[2] Ropp, R. C., Encyclopedia of the Alkaline Earth Compounds, *Elsevier*, **2013**, 116., 201.-202.

Results: differential thermal analysis

Beryllium undergoes various thermal processes upon/after commencing rapid oxidation. The temperature at which such processes take place is dependant on the treatment conditions.

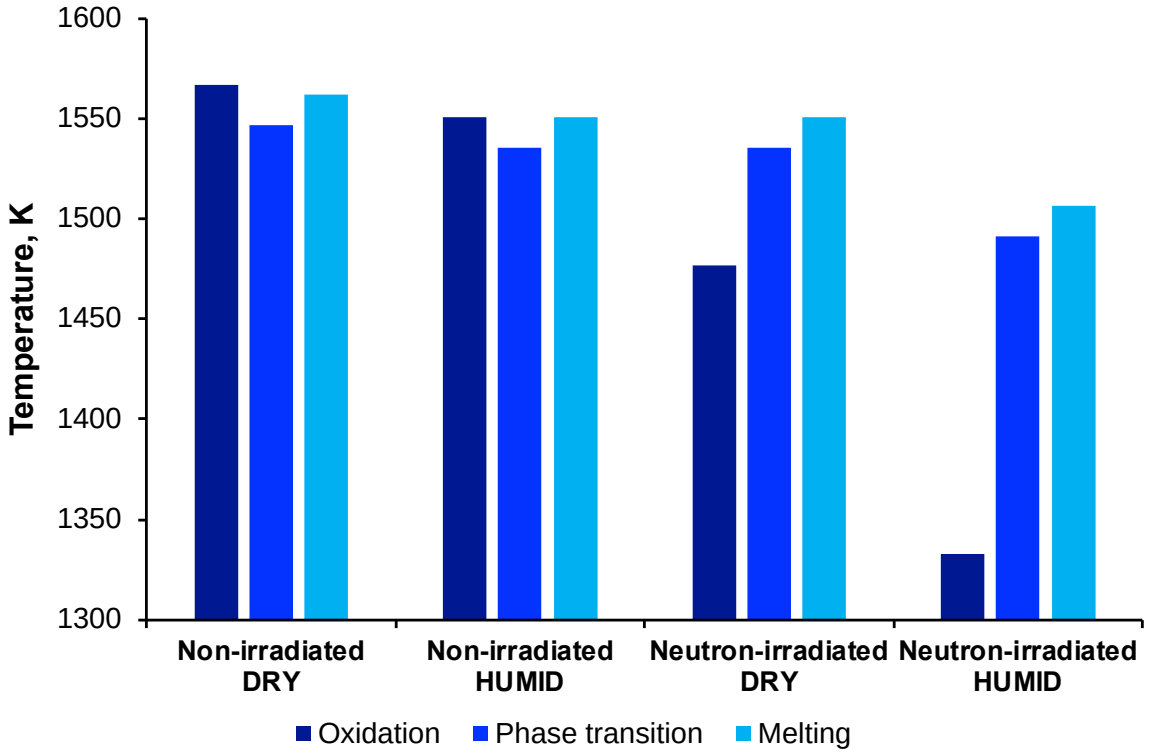

Fig. 1. Beryllium pebble behavior depending on treatment conditions

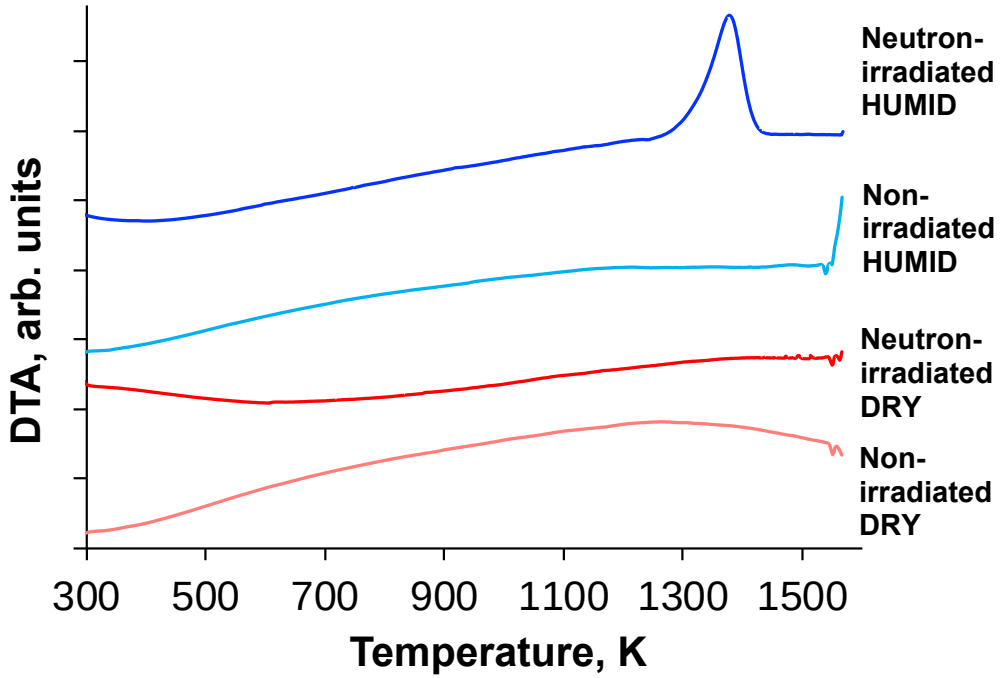

Fig. 2. DTA signal of non-irradiated and neutron-irradiated beryllium pebbles in humid and dry airflow.

It is evident that neutron irradiation can be deemed one of the most important factors impacting thermal process temperatures for Be. However, humidity also has a key role. Elevated humidity exhibits an **augmenting effect**, most significantly impacting the oxidation process.

Results: microstructure analysis (I)

Beryllium pebbles after thermal treatment have considerably increased in size and a complex layer of BeO on the surface is observed.

Furthermore, the complex structures include systems of cracks ard microcracks of varying depth and length.

Investigating the Be pebbles using a scanning electron microscope (SEM) revealed different types of micro- and nano-sized structures.

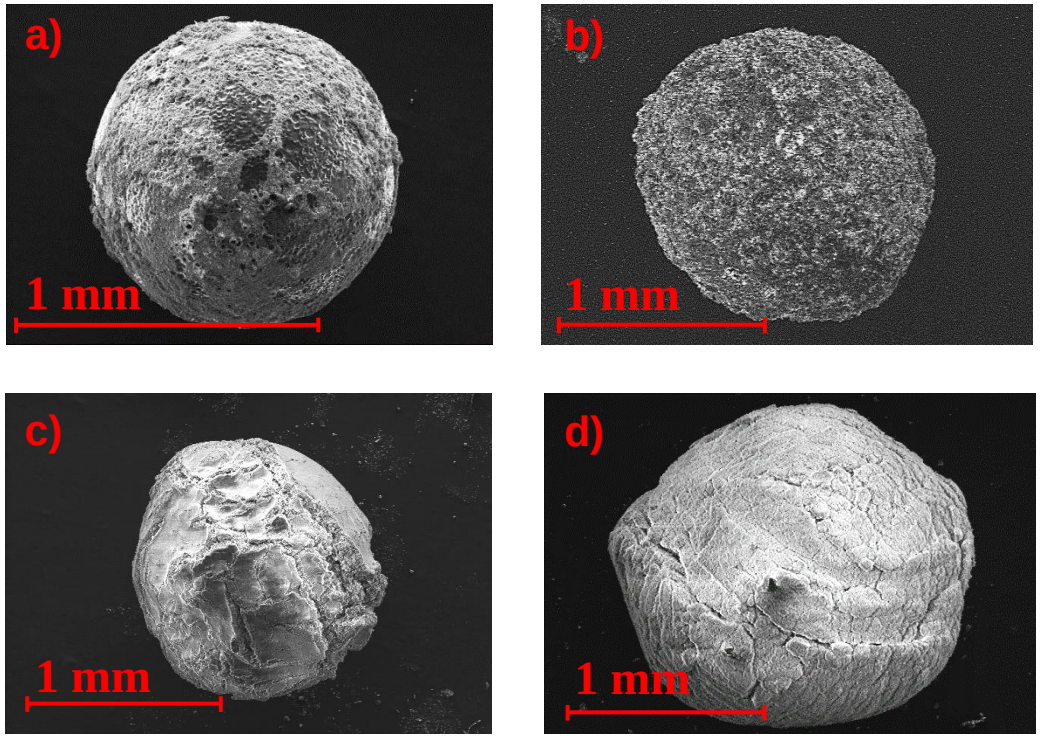

Fig. 1. Beryllium pebbles after thermal treatment under SEM: ***a)*** *non-irradiated DRY,* ***b)*** *neutron-irradiated DRY,* ***c)*** *non-irradiated HUMID,* ***d)*** *neutron-irradiated HUMID*

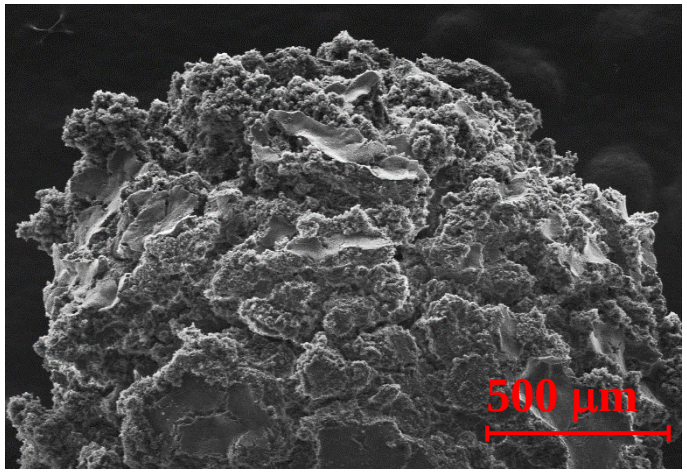

Fig. 2. Non-irradiated beryllium pebble after treatment in humid airflow

Results: microstructure analysis (II)

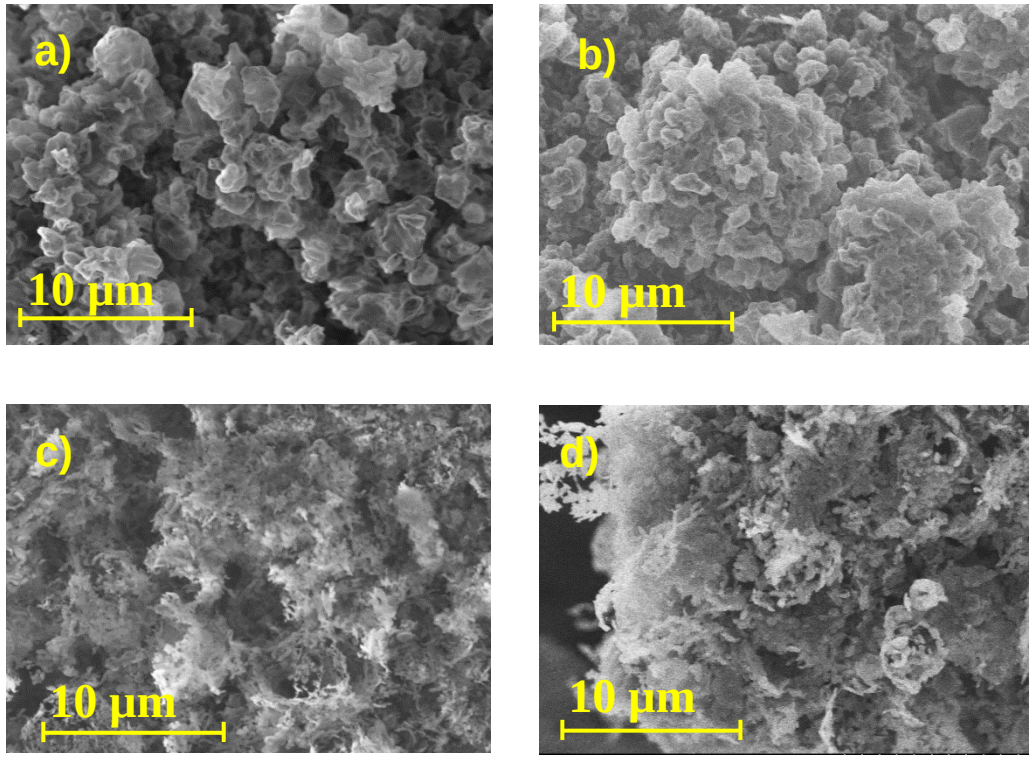

*Fig. 1. Beryllium pebbles **after** thermal treatment under SEM at* $4{,}5 \cdot 10^3$ *magnification:*
***a)** non-irradiated DRY, **b)** neutron-irradiated DRY,*
***c)** non-irradiated HUMID, **d)** neutron-irradiated HUMID*

Furthermore, magnifying the treated pebbles differences in the shape of BeO are observed.

Dry air appears to create cluster-like oxide particles, whereas humid air flow produces needle-like particles, which are substantially smaller and, thus, fragile.

In case of LOVA/LOCA such micro- and nano-sized particles are likely to form and pose **a hazard as they can form a toxic BeO aerosol** that causes berylliosis [1, 2].

Some spherical structures are visible on neutron-irradiated samples, indicating that a de-trapping of gas has likely occurred. Such structures **warn of probable trapped gas release** during LOVA/LOCA.

[1] Balmes, John R, *American Journal of Respiratory and Critical Care Medicine*, **2014**, 190(10), e34–e59.
[2] Shima, S., *Sangyo Igaku*, **1983**, 25(2), 91–105.

Discussion: safety protocols for LOVA/ LOCA

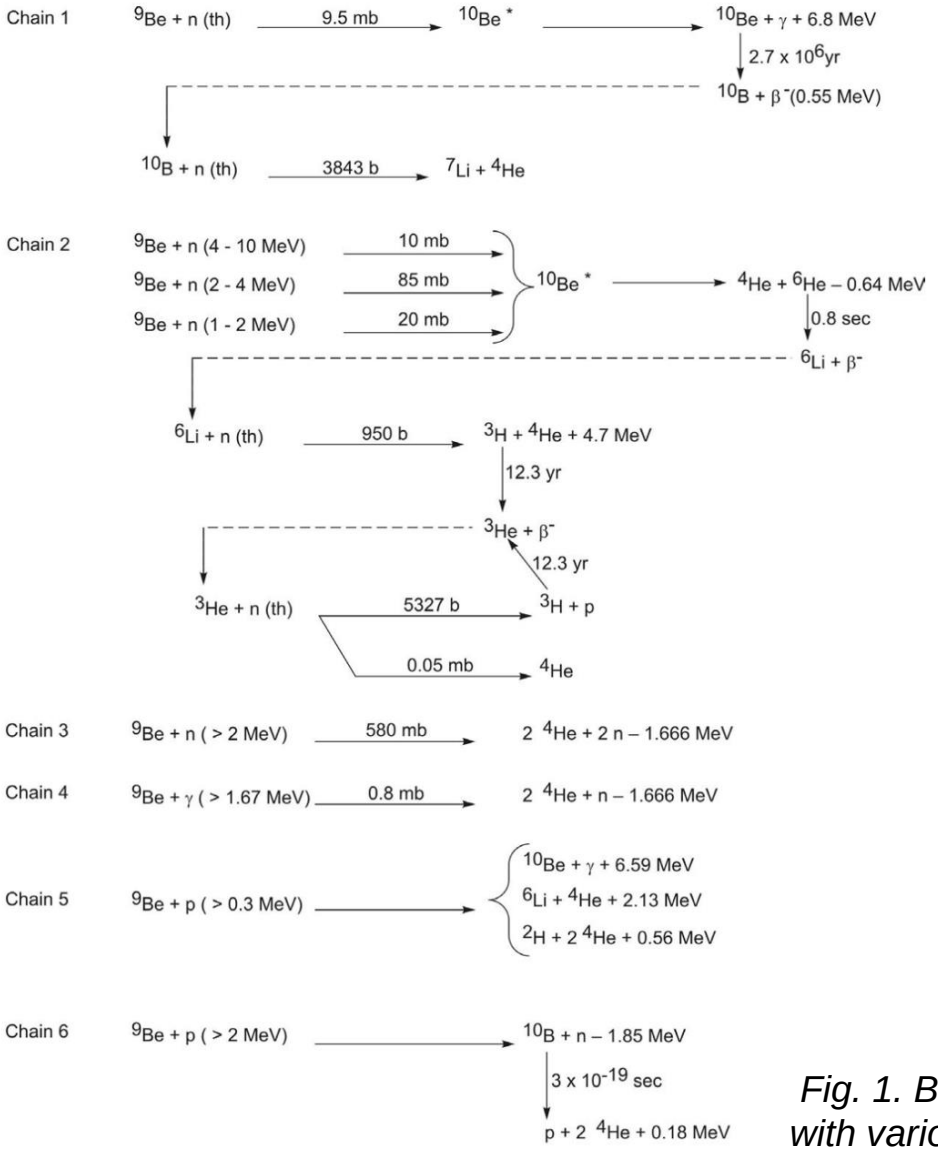

Fig. 1. Beryllium chain reactions with various particles [6]

Safety protocols regarding issues in nuclear fusion reactors [1] for extreme events such as LOVA/LOCA should further include process hazard analysis (PHA) evaluations on the necessary precautions regarding fusion fuel and other gaseous substance accumulation during operation and their subsequent release during LOVA/LOCA [2].

As beryllium can undergo violent oxidation and yield airborne beryllium compound particles [3, 4] together with the de-trapping of fusion fuels/other gases posing a hazard of an explosion [5], existing comprehensive safety protocols should be updated considering the conditions at which Be oxidizes and subsequently releases gaseous species depending on the severity of the LOVA/LOCA.

[1] Matthew Lukacs, Laurence G. Williams, *Fusion Engineering and Design*, **2020**, 150, 111377.
[2] Edgar Ebert, Jürgen Raeder, *Fusion Engineering and Design*, **1991**, 17, 307-312.
[3] R. Rossi, P. Gaudio, J.F. Ciparisse, L.A. Poggi, A. Malizia, *Fusion Engineering and Design*, **2018**, 126, 156-169.
[4] Riccardo Rossi, Pasqualino Gaudio, Luca Martellucci, Andrea Malizia, *Fusion Engineering and Design*, **2021**, 163, 112161.
[5] A. Denkevits, *Fusion Engineering and Design*, **2010**, 85(7-9), 1059-1063.
[6]. J. E. Evans, "Reaction Products in High nvt Irradiated Beryllium", **1956**, United States Atomic Energy Commission, IDO-16364.

Conclusions

1. Beryllium oxidation and thermal processes appear to be directly impacted by the thermal treatment conditions as well as neutron irradiation.
2. Cracking of the Be pebbles and an increase in size is visible after treatment. A micro- and nanosized layer of BeO has formed with some particles very weakly bonded to the pebble.
3. Results show the substantial cumulative effect of neutron irradiation and elevated relative humidity during thermal treatment on the thermal processes of beryllium, indicating a lowering of activation energies necessary for chemical reactions to take place.
4. Elevated humidity together with neutron irradiation yield the most significant mass increase (~180%) due to BeO formation.
5. The neutron irradiation causes structural defects as well as through nuclear reactions produces gaseous species within bulk material that ultimately lead to channel formation for the de-trapping of gas. In a fusion reactor, the gaseous mixture would contain fusion fuel that would also be released (hence posing a radiation hazard).
6. Results obtained herein this study are to be implemented in nuclear fusion device safety solution development as well as LOVA/LOCA impact assessment on fusion fuel release.

Thank you for your attention!

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