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

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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]  

[3] ITER - the way to energy [https://www.iter.org]
Experimental

**Samples:**
1. Be pebbles, **neutron-irradiated** (E > 0.1 MeV for 294 days, fluence 3-4 \( \cdot \) 10\(^{25} \) m\(^{-2} \), at a temperature range from 423 to 823 K) and **non-irradiated** pebbles of same size (Ø ~ 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%)

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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%.

\[
\text{Be}(s) + 0.5\text{O}_2(g) \xrightarrow{\text{temp.}} \text{BeO}(s) \\
\text{Be}(s) + \text{H}_2\text{O}(g) \xrightarrow{\text{temp.}} \text{BeO}(s) + \text{H}_2(g) \\
3\text{Be}(s) + \text{N}_2(g) \xrightarrow{\text{temp.}} 3\text{BeO}(s)
\]

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


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.

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 and 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)

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.

Fig. 1. Beryllium pebbles after thermal treatment under SEM at 4,5 $\cdot$ 10³ magnification:
a) non-irradiated DRY, b) neutron-irradiated DRY, c) non-irradiated HUMID, d) neutron-irradiated HUMID
Discussion: safety protocols for LOVA/ LOCA

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.

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

The participation in this conference was supported by the International Atomic Energy Agency (IAEA) project “Strengthening the Competence in Radiation Technologies and Safety for Biomedicine and Materials Science”, “To enhance the competence and infrastructure for providing training in radiation technologies and safety in Latvia”, project number LAT0004.