

Background

In future fusion reactors, beryllium may be used both as a first wall material and as neutron multiplier in the breeder blanket. Neutron irradiation of the beryllium during reactor operation yields important amounts of tritium and, due to the presence of metallurgical impurities in the beryllium metal, leads to a high level of activation. At the end of its reactor life, a waste management solution for this beryllium should be available. In the light of societal acceptability of fusion power, it would be desirable to dispose of a method to recycle the irradiated beryllium for re-use in fusion applications. At present, however, no recycling route is available that can accommodate the high tritium content and level of activation characteristic of fusion-irradiated beryllium. Therefore, at SCK•CEN, we set out to contribute to the development of such a route.

Objectives

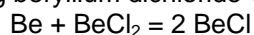
- Propose a generic recycling route for fusion-irradiated beryllium;
- Assess existing technologies that could be used for beryllium recycling;
- Identify processes that require further development.

Principal results

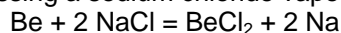
Taking into account the characteristics of fusion-irradiated beryllium, any recycling route will have to exist of the following three generic steps: detritiation, removal of radionuclides and other impurities, and production of recycled beryllium pebbles (or tiles in the case of first wall applications) by powder metallurgy. As refabrication of beryllium pebbles and tiles by powder metallurgy is a largely mature practice, focus of this study is on the first two steps of the recycling route: detritiation and purification.

Distinction has to be made between surface tritium and bulk tritium. The main mechanism responsible for the presence of a tritium-rich surface layer is co-deposition. Bulk tritium on the other hand, is mainly generated by neutron transmutation. The tritium-containing deposited film can be removed by in-reactor techniques, including the use of tokamak pulses possibly combined with ion cyclotron resonance heating, cleaning with a flash lamp, laser ablation, and plasma torch exposure. The bulk tritium can be removed by high-temperature annealing.

The second step, purification, can be achieved by several types of processes, including vacuum distillation, zone melting, and chlorine processes. The chlorine route consists in the chlorination of the irradiated beryllium and the subsequent reduction of beryllium chloride to highly pure metallic beryllium. The vapour pressure of beryllium is relatively low, as discussed above. Beryllium halides, by contrast, are fairly volatile at high temperatures. This fact forms the basis of the chlorination process: beryllium chloride is much more volatile than the impurities present in irradiated beryllium and therefore chlorination should be able to yield high purity beryllium. Several processes for the volatilisation of beryllium have been proposed, including passing beryllium dichloride vapour at a reduced pressure over heated impure beryllium:



and passing a sodium chloride vapour over the beryllium:



High purity beryllium chloride can be obtained by fractional distillation.

After the chlorination step, beryllium chloride can be reduced electrochemically, chemically, or thermally.

Electrochemical reduction of beryllium chloride

There are low and high temperature processes for the electrochemical reduction of beryllium chloride. The *low temperature* process consists in electrolyzing a mixture of approximately equal parts of beryllium chloride and sodium chloride at a temperature of 370°C in a nickel container which serves as the cathode for electrolysis. The anode is a centrally located graphite rod. A potential of 4 to 6 volts is used. The electrolysis is continued until the beryllium chloride is reduced to about 45% of the electrolyte, which after the removal of the beryllium flake is restored to 50% content to permit the continuation of the process. The metal thus removed is separated as completely as possible from any residual electrolyte and then quenched in ice water to effect a complete separation. After being quenched it is in turn separated from the quench liquor which is returned to the process for the recovery of beryllium as beryllium hydroxide. The flake is then alternately washed with dilute solutions of sodium hydroxide and nitric acid to effect, first, a reduction of aluminium content, and second, a removal of adhering surface oxides. As a final treatment the flake is washed with methanol and dried.

The *high temperature* process involves adding 13 to 15% beryllium to a mixture of equal parts of sodium and potassium chloride. The fusion temperature of this salt is about 760 to 790°C. This temperature must be increased during the electrolysis because of the depletion of beryllium chloride to a final temperature at completion of between 900 and 925°C. A direct current of about 6 volts is supplied between a graphite anode and the metallic container, which acts as cathode. Upon completion of the process, the entire content of the cell is transferred to another container where the salts are permitted to solidify. At this point, the reduced beryllium metal is in the form of flakes. After solidification, the flake is washed with dilute solutions of sodium hydroxide and nitric acid, to remove adhering surface oxides.

Thermal reduction of beryllium chloride

A potentially elegant solution for the reduction of beryllium chloride is to pass the gas over a SiC filament and decomposing it. Key questions remain the stability of the contact between the beryllium chloride gas and the filament (including the yield of the reaction, which was at best 'variable') and the volume of beryllium that could eventually be recovered by pursuing this method.

Past experience with the electrochemical and chemical reduction of beryllium chloride showed that these methods resulted in substantial amounts of impurities in the metallic beryllium. The main advantage of thermal reduction is that no new impurities are introduced into the beryllium chloride stream, but on the other hand, a stable contact between the gas stream and the filament seems to be difficult to achieve. In the light of this information, we propose the further development of thermal reduction, by optimising the physical geometry of this step, which should lead to both an increased contact surface, and an increased through-put of the thermal reduction step.

Main contact person

Frank Druyts, frank.druyts@sckcen.be

Main reference

F. Druyts, K. Dylst, J. Braet, "Beryllium recycling: feasibility and challenges," Proceedings of the 8th IEA International Workshop on Beryllium Technology, December 5-7, 2007, Lisbon, Portugal.