PYROHYDROLYSIS OF BITUMINIZED WASTE DRUMS

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ABSTRACT

NUKEM Technologies Engineering Services GmbH (NUKEM) has set up a R&D program for the treatment of bituminized waste stored in drums. This program is divided into two major phases, the lab-scale facility processing up to 30 kg of simulated bituminized waste and the demonstrator plant capable to treat one 200-liter drum per batch. In 2020 the R&D program was started with the lab-scale facility in 2022 it is planned to build the demonstrator based on the achieved results from the lab-scale phase.

INTRODUCTION

In the past, Bitumen was widely applied as matrix for conditioning of many kinds of low and intermediate level radioactive waste, such as

- Homogeneous conditioning of evaporator concentrates and sludge
- Homogenous embedding of spent ion exchange resins
- Heterogeneous embedding of technological waste (metal)
- Heterogeneous embedding of mixed waste (compacted drums, clothing, rubber, plastics)

Bitumen was seen a long-lasting material providing a safe enclosure of the radioactive substances and a good stability for the expected long storage and disposal time. However, test in France have shown that over the time radiolysis can occur, causing the formation of gas bubbles within the Bitumen matrix. This leads to acceptance problems in underground repositories due to risk of fire as well as the lack of long-time stability.

Due to the characteristics of the Bitumen matrix reconditioning of these waste package forms great challenges, such as

- Large amount of bituminized waste
- High calorific value
- Content of volatile radionuclides
- Difficult handling of molten Bitumen
- Potential spread of contamination

DESCRIPTION

Up to now no direct reconditioning process for the bituminized waste packages has been established at the market. There are ideas to co-incinerate the bitumen in existing facilities, but that would need

removal of the bitumen from the waste package prior to treatment. Additionally, that would still leave the original waste packages, having significant amounts of bitumen and radioactive material left inside, to be handled and treated somewhere.

NUKEM has already in the past executed test at the University of Freiberg, as well as with DBI-Virtuhcon showing in principle the suitability of pyrohydrolysis for the treatment of bituminized waste packed in whole drums.

From the results of these laboratory tests a concept idea for the treatment of drums containing bituminized radioactive waste was developed and NUKEM set up a R&D Project. The R&D Project for "Pyrohydrolysis of Bituminized Waste" support the way forward of the implementation of such a pyrolysis process in an industrial scale. The first step is to set up a so-called Lab-scale facility, processing up to 20 kg of bitumen, and a second step using a demonstrator unit able to treat a 200-liter drum in order to gain the full understanding of the pyrohydrolysis process involved in the treatment of the Bitumen matrix. In its final stage, the demonstrator unit should prove the process by using real radioactive bituminized waste.

It is assessed that the Technical Readiness Level (TRL – based on US DoD and EC assessment system) is prior to the start of the R&D program TRL3. When successfully performed the lab scale development TRL is increased to TRL5, while after completion of demonstrator work TRL7 should be achieved.

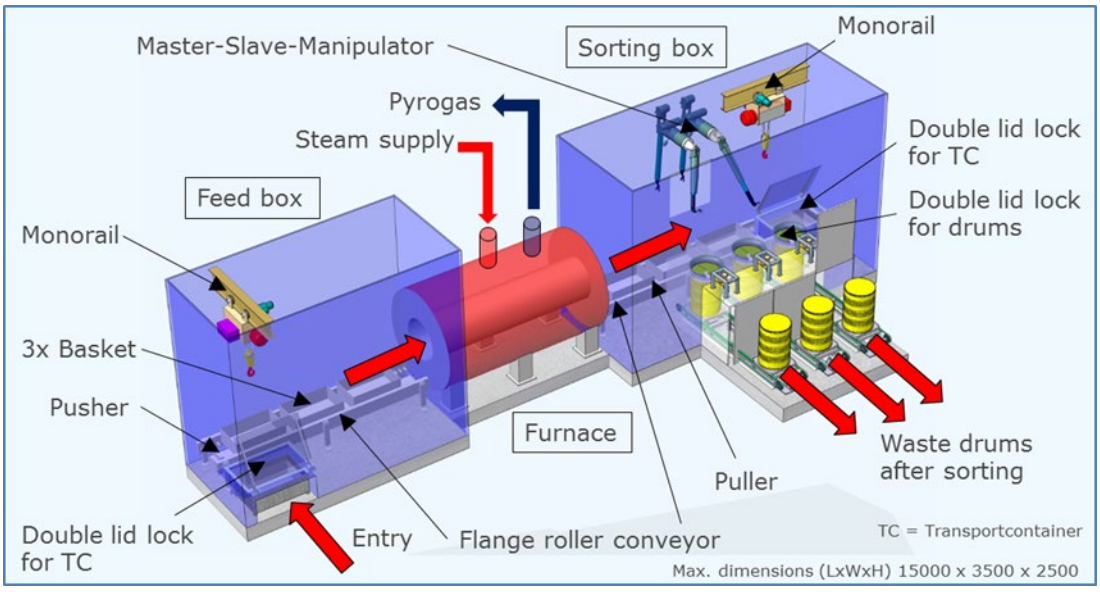

Figure 1: Illustration of an Industrial Scale Facility

LAB-SCALE FACILITY

In 2020 NUKEM has started its R&D Project "Pyrohydrolysis of Bituminized Waste". The first step was to design and procurement the lab-scale furnace able to process 30-liter drums containing simulated bituminized waste. Beginning of 2021 the furnace was delivered to DBI Virtuhcon and successfully installed and commissioned.

Parallel to the works on the lab-scale facility drain tests using 400 ml canister filled with pure bitumen were executed in order to investigate the behavior of bitumen and to simulate the necessary pretreatment step for the co-incineration. These results should serve as a benchmark for the pyrohydrolysis.

For the drain test the canister were punched around the circumference and to the height of the bitumen filling. After that the canister were slowly heated up with a heating rate of 10K/min to temperatures between 100 and 180 °C. Figure 2 shows exemplary pictures of the drain test.

Figure 2: Filled Canister, Drain of the Bitumen and Canister with Remains after Draining

After successful installation and commissioning of the lab-scale furnace first tests were performed using pure bitumen. The bitumen was filled into 30 liter stainless steel drums. The weight of the bitumen ranged between 10 and 20 kg. The value of 20 kg was chosen, because the remaining 10 kg will be used in the next step for the addition of salt, ion exchange resins or metal, simulating radioactive waste embedded into the bitumen matrix.

Figure 3 shows the installed lab-scale furnace, whereas Figure 4 shows a typical temperature profile used during the tests.

Figure 3: Lab-scale Furnace

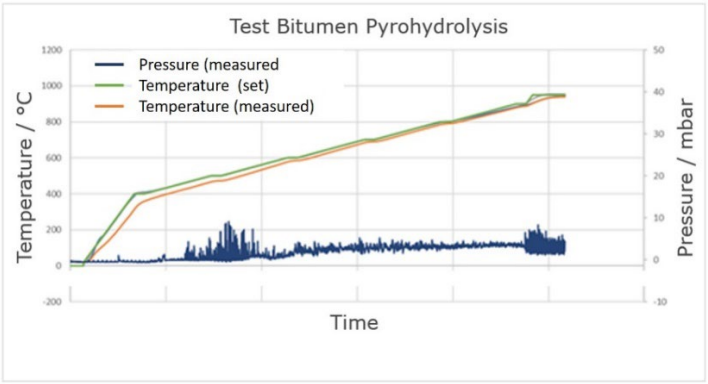

Figure 4: Typical Temperature Profile

As a first phase of the lab-scale furnace test program “approach-test” were executed to define the temperature profile and to get first impressions on the behavior of the bitumen in larger amounts. The bitumen was mainly decomposed during the pyrohydrolysis process. Nevertheless, at the bottom and at the surfaces of the 30-liter drum powdery remains of the original bitumen could be seen after the treatment. Also on the outside of the drum dust could be seen, which deposits itself during the pyrohydrolysis process on the surface.

By adjusting the temperature profile as well as the amount of steam used within the process the deposits could be removed completely. However, the tests showed that part of the drained bitumen, which is collected in a trip tray below the 30-liter drum, still remains (see Figure 5).

Figure 5: Inside and Outside View of the 30-liter Drum after Pyrohydrolysis Test as well as dust remains in the Trip Tray

In 2022 the test program will be continued by adjusting the steam flow to further reduce the remains in the trip tray. In the meantime, 30-liter drums were filled with salt or ion exchange resins embedded in bitumen matrix to simulate bituminized radioactive waste packages.

DEMOSTRATOR FACILITY

After the promising results from the lab-scale tests NUKEM decided to go ahead with the next phase in the R&D program. Therefore, NUKEM is searching for a partner for the establishment of a demonstrator unit. The demonstrator unit should be set-up at a bituminized waste owner institution / research center having the license to work with radioactive materials. The aim is to use bituminized waste from existing stocks to demonstrate the technical readiness of the process and its technical application. Since the demonstrator unit should also serve as a pilot plant for the treatment of bituminized waste it is crucial that radioactive bituminized waste can be treated by the institution.

Selection criteria for the site would be:

- License to work with radioactive material
- Permission and logistic to handle and to treat radioactive waste, including bituminized radioactive waste
- Available space for the demonstrator facility
- Ability to discharge treated off-gas, preferably with an existing post combustion system with connected off-gas treatment system

Goals and objectives of the Demonstration Unit

- Main target of the demonstrator unit is proving a reliable operation of the pyrohydrolysis facility for bituminized radioactive waste (target TRL 7)
- Other targets of the demonstrator unit:
    - Reliable set of operational and design parameters enabling precise calculation of investment and operational cost of an industrial scale facility
    - Pilot facility for the treatment of bituminized radioactive waste
    - Showing potential clients the successful operation
    - Verification of the process parameters from the lab scale facility
    - Information of the media consumption values

RESULTS

Bituminized radioactive waste seems to be at least partly excluded from the disposal at deep geological underground repositories. Owners of these waste package seek alternatives for the re-conditioning of the waste. Most of the established methods need to pretreat the waste package by draining the bitumen from the packages as the downstream processes are not capable to treat whole waste packages. When draining bitumen from waste packages prior to the treatment of the bitumen itself significant amounts of bitumen will remain in the waste package. These will also contain activity from the originally embedded radwaste. Due to the properties if the bitumen, e.g., sticky, a further handling of drained waste packages within glove boxes forms another problematic step in the treatment of bituminized waste packages.

The pyrohydrolysis process seems to be a promising technology to safely reprocess complete waste packages with bituminized radioactive waste without the need of draining first. The first test with 20 kg bitumen treated in the lab-scale furnace showed very good results in mass / volume reduction. After further optimizing the operational parameters, such as steam and temperature profile, the already high mass / volume reduction should be further increased.