

Gasification-based thermal treatment of Low and Intermediate Level Waste containing organic matter

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Abstract. VTT has developed a gasification-based technology for processing low- and intermediate-level waste (LILW) rich in organic matter. VTT's process is based on well-controlled thermal fluidised-bed gasification followed by efficient gas cleaning, gas conditioning/oxidation, wet scrubbing of the oxidised gas (flue gas), and finally, filtering through a high efficiency particulate air (HEPA) filter, which acts as a backup cleaning system. In the case of an organic ion exchange resin (IXR) the primary product from thermal gasification is fine filter dust. In addition, the process produces some bottom ash, which consists mainly of the bed material. The wet scrubber liquid may also contain some activity. Demonstration trials were carried out using unspent organic IXR containing a small amount of added stable Cs in order to simulate Cs content in spent IXR. Gasification tests confirmed the capability of the process to remove organic matter from the IXR and clean the resulting off-gas as required. Simulated waste IXR was reduced to approximately 1 wt-% of its original mass before immobilisation. Filter dust and bottom ash have to be immobilised before final disposal. VTT has previously developed an advanced immobilisation technology based on geopolymerisation and this has been applied to samples from the THERAMIN test trials. VTT's technology has been designed as a compact process, which can be operated at the nuclear power plant site. Until now, all development and verification test trials have used simulated waste materials. The next step is the demonstration of the process using real radioactive waste.

1. Introduction

VTT has, over several decades, developed gasification and gas cleaning technologies for different applications and different feeds [1, 2, 3]. The primary focus has been on production of fuel gas or ultra clean synthesis gas from high quality fuels, but later in the 1990s development also focused on using different wastes and industrial by-products containing harmful contaminants that need to be removed from the resulting gas; this experience has enabled the development of thermal gasification technology for treatment of low- and intermediate-level radioactive waste (LILW).

The gasification-based process developed by VTT is based on well-controlled thermal fluidised bed gasification followed by efficient gas cleaning, gas oxidation and wet scrubbing of the oxidised gas. The process has been verified to be a suitable thermal treatment method for organic ion exchange resin (IXR). However, this technology can also be applied to any organic waste streams, including liquid organics. Removal of organic matter by thermal gasification results in significant volume reduction, which was demonstrated successfully in the THERAMIN project. The main final residue is filter dust, which requires further processing in order to immobilise it and enable safe disposal. Immobilisation can



be achieved by cementation. However, VTT has developed an advanced immobilisation technology based on geopolymers, which enables high waste loading and has shown lower leachability characteristics than Ordinary Portland Cement immobilisation. Geopolymerisation was used to immobilise the samples from demonstration test trials carried out in the THERAMIN project [4].

2. Gasification based LILW treatment technology

Thermal gasification is a mature technology that has been used for almost 200 years to produce energy from carbon-containing fuels. Gasification is a partial oxidation process. Conditions in the gasification reactor are reducing, and the gas produced still contains combustible compounds. Some oxygen is needed to produce the heat required for thermal gasification reactions, but the gas atmosphere is reducing which has an impact on inorganic chemistry in the reactor.

The major radionuclides in organic LILW are typically metals such as caesium and nickel, or carbon (C-14). Through gas cleaning, all solid material can be removed easily prior to gas combustion. Gasification temperature is significantly lower than high temperature vitrification and thus only a small proportion of some metals will be vaporised. These vaporised metals can be solidified by cooling the gas prior to particulate removal based on efficient high-temperature filters, and thus metals can be removed together with other solids. In addition, in radioactive waste treatment, the flue gases are cleaned after gas oxidation (combustion) by filtering through a High Efficiency Particulate Air (HEPA) filter.

VTT has Bubbling Fluidised Bed (BFB) and Circulating Fluidised Bed (CFB) gasification test rigs suitable for thermal gasification of organic IXR or other organic LILW. The demonstration presented in this paper was carried out using an atmospheric pressure pilot-scale CFB gasification test rig (Figure 1) at VTT's Piloting Centre. The bed diameter of the reactor was 102 mm and the freeboard diameter was 150 mm. Electrical heaters were situated around the reactor and gas pipes to compensate for heat loss. The test rig was equipped with versatile fuel feeders. In addition, the test rig was equipped with a separate feeder for the bed material/additive. The fuel feeding port was located above the dense bed. The particles from the recycling cyclone were introduced close to the air distributor. Electrically preheated air was used as a gasification agent.

The gasification product gas was cleaned in the recycling cyclone followed by a gas cooler and a high-temperature filter unit. The filter unit consisted of 12 metal candle elements, which were approx. 60 mm in diameter and 1000 mm in length. Nitrogen was used as a filter pulsing gas (back pulse cleaning). The role of the filter unit was to remove particles from the product gas. Easily volatile compounds condense into solid particles when the gas cools and when the filtering temperature is low enough, and harmful compounds (e.g. Cs in these test trials) are thus removed from the process along with the filter dust.

The test rig was equipped with an automatic data acquisition system to measure temperatures and pressures at different locations. The flow rates of gasification agents and purging nitrogen were continuously measured and recorded. The fuel feed rate was controlled adjusting the feed rate of IXR on the basis of the screw calibration curve, and the rotation speed of the metering screw was continuously measured. In addition, feedstock (IXR) batches were always weighed when loading into the feeding hopper in order to check the actual feedstock consumption after the test trial. The main gas components CO, CO₂, H₂, CH₄ and O₂ are monitored by on-line analysers. All mass streams were measured except product gas, which was calculated based on the other mass streams and the product gas composition. All solid residues (bottom ash and filter dust) were collected and analysed.

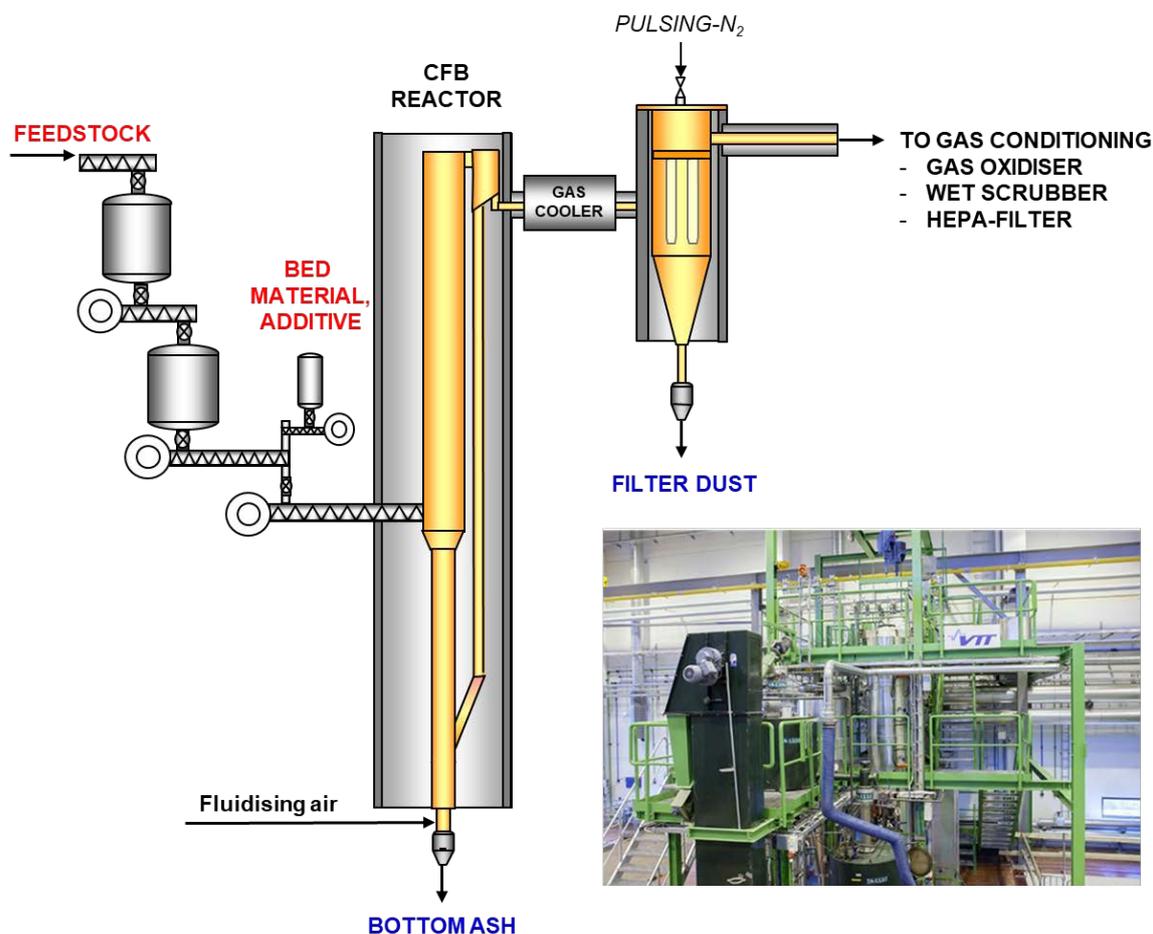


Figure 1. Pilot-scale Circulating Fluidised Bed (CFB) gasification test rig.

3. Demonstration trials

Demonstration trials were carried out in the pilot-scale CFB gasifier using IXR as a feed. The feed was impregnated with CsCl in order to simulate Cs content in a real spent IXR. A total of 325 kg of organic IXR was treated over the course of three trial days. The total duration of the trial was 26.5 hours. The average gasification temperature was 885-915°C, and the filtering temperature varied between 415 and 450°C. A total of 18 kg of inert Al₂O₃ (of particle size 0.18-0.25 mm) was used as a bed material. The dry gas composition prior to oxidation during the second test trial day is presented in Figure 2.

The success of the trial is primarily assessed by determining mass balances for the main elements involved in the reactions. The mass balances for the trials were calculated based on the average values of the measured data (including ash and ultimate analysis of the IXR and filter dust, dry gas composition as well as tar content in the dry product gas). The carbon conversion calculation is based on the carbon mass balance and it describes how efficiently input carbon is converted to gas. The carbon conversion to gas and tars was over 98.3 %, which means that the removal of the organic material from the IXR was successful. The product gas was dilute because the equivalence air ratio was kept deliberately rather high (0.49-0.54) since the main target was to remove the organic matter from the feedstock (IXR), not energy production. The mass and volume of waste to be disposed was significantly reduced: a total of 325 kg of organic IXR was gasified during three test trials, and 3.3 kg of filter dust was produced during the whole feedstock feeding time, which represents about 1 wt-% (1.4 wt-% dry) of the fed IXR.

The demonstration trials aimed to verify the effectiveness of gasification-based technology to reduce the volume of organic IXR and to produce treated waste (solid residue) for further characterisation tests. The product of thermal gasification was powder residues and therefore immobilisation was required to

enable final disposal. The solid residue can be immobilised by applying different technologies; in the THERAMIN project, geopolymerisation was used. The methodology for this process was developed during previous VTT projects and so far the results have been significantly better than immobilisation using Ordinary Portland Cement (OPC) [4]. Geopolymerisation enables high waste loading and simultaneously leachability of radionuclides is significantly lower compared to immobilisation by OPC.

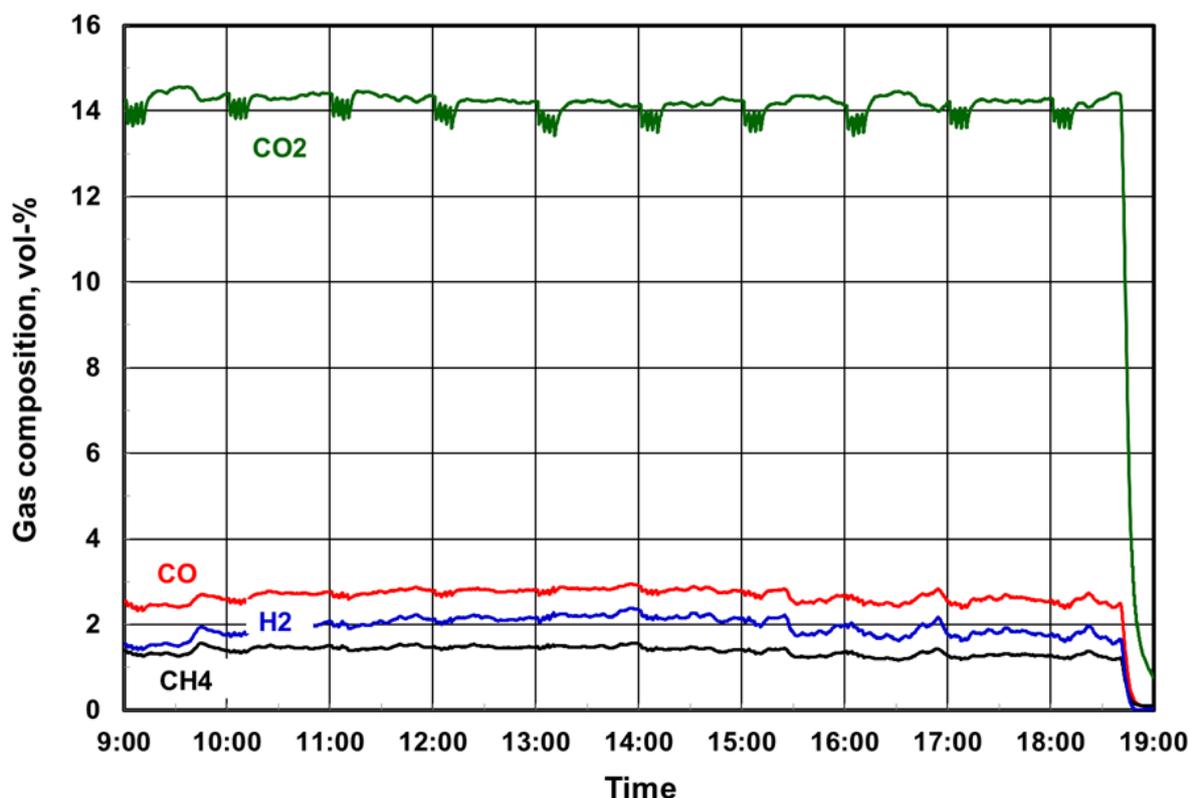


Figure 2. Main dry gas composition during the second trial day (the main gas component is nitrogen, which is not shown in the figure).

4. Conclusions

The performance of the gasification-based thermal treatment of organic ion exchange resins developed by VTT was demonstrated in the THERAMIN project. The treated waste (solid residue from the gasification treatment) was further immobilised by geopolymerisation. Demonstration trials including geopolymerisation verified that the developed thermal gasification, gas conditioning and geopolymerisation of treated waste effectively consigns all metallic radionuclides within the ash products, reduces waste volume significantly, does not cause any off-gas emissions, and demonstrates that the ashes can be immobilised effectively through geopolymerisation. The development will be continued further, in particular targeting demonstration with active wastes from nuclear power plants.

Acknowledgments

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