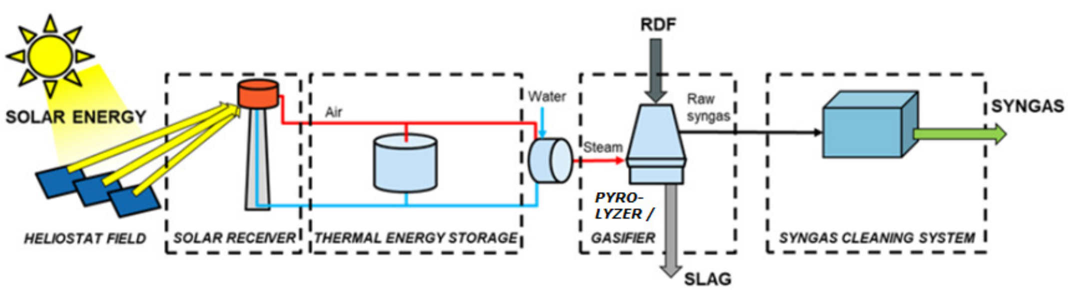
NEW-MINE

EU TRAINING NETWORK FOR RESOURCE RECOVERY THROUGH ENHANCED LANDFILL MINING

European Training Network for Resource Recovery Through Enhanced Landfill Mining (NEW-MINE)

D2.1 Final report on WP thermochemical conversion



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Confidential

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1. Introduction and objectives

Urban mining through excavation and processing of waste from landfill sites has been considered as a promising option to recover material and energy resources towards the production cycle. Therefore, urban mining could be a crucial factor for enabling a closed-loop cycle in the latest circular economy definition. Also, the demand for land redevelopment and pollution mitigation are the other factors that encourage the excavation and remediation of landfill sites. The Enhanced Landfill Mining (ELFM) concept emerges to overcome those challenges. ELFM can be achieved by implementing integrated valorisation of landfill waste, that consists of materials recovery through advanced sorting and separation processes of different waste streams, as well as the conversion of non-recyclable materials into gaseous fuels or/and vitrified slags through thermochemical conversion processes. As such, ELFM transforms a significant fraction of the excavated material into higher-added-value products, such as fuel-grade H_2 and alternative binders for low-carbon construction applications, and only a minor fraction needs to be re-landfilled.

One of the critical fractions from the advanced mechanical processing of excavated landfill waste is the Refuse Derived Fuel (RDF) fraction. In Classic Landfill Mining scenarios, common in Asia, this RDF fraction is further processed in grate incinerators or co-incinerators. In the EU-28 this is less straightforward as the quality requirements for such RDF (especially in the case of co-incineration) are stricter. NEW-MINE, however, envisages different thermochemical conversion processes. Based on two separate lines of successful research in Europe, NEW-MINE targets plasma-driven gasification, solar-driven gasification and hybrid combinations, allowing the flexible use of these technologies as a function of the climatic conditions (northern vs. southern Europe). The advantage of these technologies is that they can be tailored to process more variable RDF compositions while producing higher-value outputs compared to incineration.

Considering the factors mentioned above, WP2 of the NEW-MINE project aims to achieve the following objectives,

- to develop the high-temperature reactor technology for the thermochemical processing of landfill derived RDF into valuable synthetic fuels,
- to design, fabricate and demonstrate a plasma-driven reactor, a solar-driven reactor, and a hybrid plasma/solar-driven reactor for the efficient conversion of RDF, and
- to conduct fundamental thermodynamic and kinetic analyses, along with heat/mass transfer modelling, which would serve as tools for optimisation and scale-up.

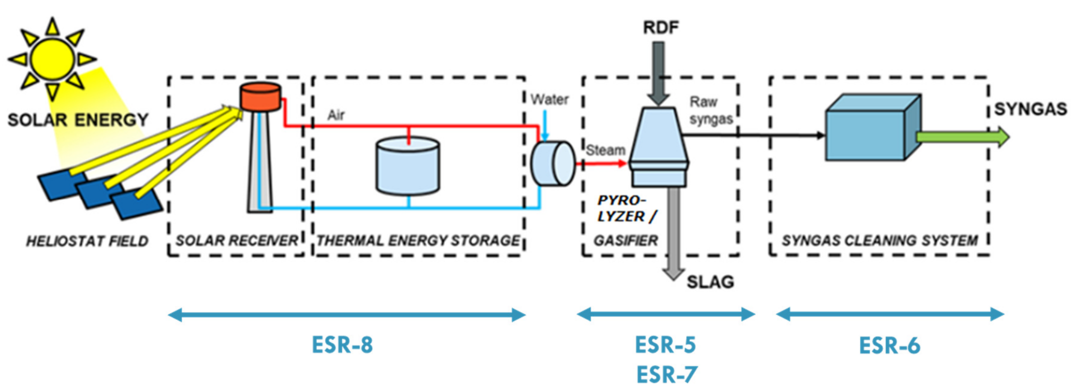

Figure 1. The scope of work within WP2.

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As shown in **Figure 1**, the research activities within the WP Thermochemical Conversion (WP2) can be divided into four subtasks, with an Early Stage Researcher (ESR) being responsible for each subtask. These subtasks are,

- ESR-5: Sustainable RDF treatment by high-temperature steam gasification,
- ESR-6: Syngas purification by plasma tar cracking,
- ESR-7: Production of valuable materials from excavated RDF by pyrolysis processes, and
- ESR-8: Thermal energy storage development for continuous solar-driven thermochemical conversion of RDF.

2. Methods

2.1 RDF samples from landfill sites

Within WP2, potential Refuse Derived Fuel (RDF) fractions obtained from pre-processed excavated landfill waste were used for the gasification and pyrolysis tests. Work Package 1 (WP1) of the NEW-MINE project performed the excavation and sorted the landfill waste into potential RDF fractions. Specifically, the used landfill RDF fractions originate from the excavation projects at two different landfill sites located in Halbenrain (Austria) and Mont-Saint-Guibert (Belgium). The RDF samples were received as pulverised materials with a particle size ranging between 3 – 5 mm.

2.2 High-temperature steam gasification of RDF from landfill

The investigations started with a kinetic study of char-steam reactions. This study was done by a thermogravimetric analyser (TGA) to obtain the mass degradation behaviour of char samples during steam gasification at various temperatures. The study was then followed by further gasification tests in lab-scale tube reactors. In these tests, approximately 4 – 10 g of samples were used to perform gasification in various atmospheres with operating temperatures ranging between 800 – 1100 °C. The produced syngas was collected and analysed by a Micro Gas Chromatography (Micro-GC). The tar samples were collected by Solid Phase Adsorption (SPA) and analysed according to internationally approved methods.

2.3 Pyrolysis process of excavated landfill RDF

The study started with the investigation of feedstock composition, which was performed using thermogravimetric and Py-GC/MS analyses for excavated waste fractions. This gave a detailed insight into the excavated waste composition. Subsequently, lab-scale pyrolysis was performed at six temperatures in the range of 400-700°C. The process products were thoroughly characterised, which includes their yields and composition. The last stage of the study is on-going and includes the lab-scale tests on catalytic cracking of volatiles produced during pyrolysis (pyrovapours). This stage also includes a complex characterisation of the products obtained.

2.4 Tar removal by plasma reactor

The experimental part was performed in a plasma reactor embedded in an oven able to reach 1200°C. The plasma power unit can reach up to 60kV and can produce plasma pulses of around 100ns at a rate of 70-72 pulses per second. The reactor is fed with a polluted synthetic gas using a set of flowmeters able to control the different gases fed and the residence time inside the reactor. The gas composition is monitored with an IR gas analyser. The level of pollutants is determined by GC-FID and GC-MS analysis ex-situ. The pollutants are sampled by the SPA/SPE method.

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2.5 Development of a thermal energy storage system

A thermal energy storage system based on high-temperature reversible gas-solid reactions was developed based on both experimental work and modelling techniques. The first task was to develop storage materials via material engineering techniques. Subsequently, a lab-scale heat storage reactor was designed, assembled, tested and modelled. The operation of the lab-scale heat storage system serves as a proof of concept and to acquire data useful to validate its numerical model which, in turn, is a useful tool to optimise and scale up the system.

2.6 Process simulation for solar-plasma hybrid gasification systems

This study aims to propose and evaluate the concept of integrated solar-plasma gasification systems for RDF conversion. The study began with the development of several novel concepts and scenarios for the integrated system. After that, technical assessments were done by evaluating the performance of the novel system through process modelling and simulation. This work is a collaboration between ESR-5, ESR-6, and ESR-8 in WP2.

3. Results

3.1 Kinetic behaviour and gasification performance of landfill-derived RDF

The results from the kinetic study imply that the char reactivity is mainly affected by the inorganic elements contained in the char. Char from finer fractions of landfill waste has higher reactivity due to the higher ratio value of alkali and alkaline metals which act as catalysts during gasification. The results from the gasification test of the landfill waste char demonstrate the importance of the process parameters during landfill waste sorting processes, especially the particle size distribution, on the thermal behaviour of the waste fuel.

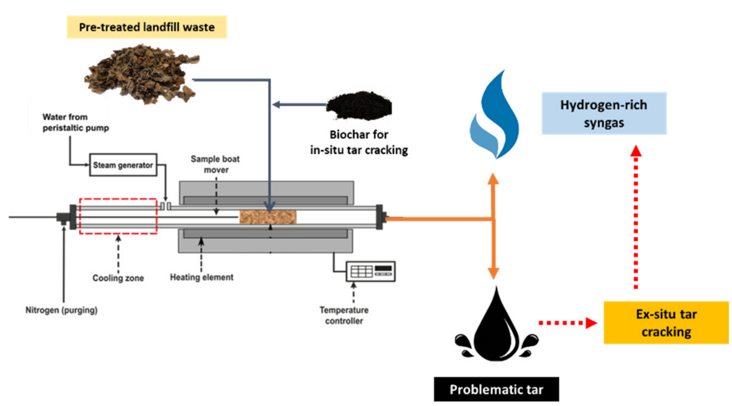

Figure 2. Process development of landfill RDF gasification using lab-scale facilities.

The results from lab-scale gasification tests of various fractions from landfill waste revealed that the gasification of landfill derived RDF may have a poor performance owing to high ash and high plastic contents. These properties may cause a lower gasification efficiency and a higher generation of problematic tar compounds. In this study, the potential of biomass or biochar addition during steam co-gasification of RDF obtained from excavated landfill waste is investigated, in order to improve

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gasification performance, especially concerning the H_2 and tar yield. It can be concluded that the addition of biochar to the gasification of RDF-landfill waste has better effects than that of biomass addition, as H_2 yields are higher and tar yields lower.

3.2 Pyrolysis of landfill-derived RDF

The first stage of the study included the thermogravimetric (TG) and Py-GC/MS analyses. The study revealed that the TG profile differs from the TG profile of fresh waste. Moreover, the feedstock's material composition was determined. This insight into feedstock's composition as well as its thermal decomposition pattern enabled planning the second stage of the study.

The second stage of the study revealed the fluctuations in pyrolysis products yields and their composition along with the temperature. Nonetheless, the produced pyrovapours show potential for catalytic cracking, but some of the potential risks of catalyst poisoning were identified as well. These findings serve as a basis for planning the third stage of the study (catalytic cracking of pyrovapours to improve gas quality).

The third experimental campaign is on-going; therefore, no conclusions can be drawn yet.

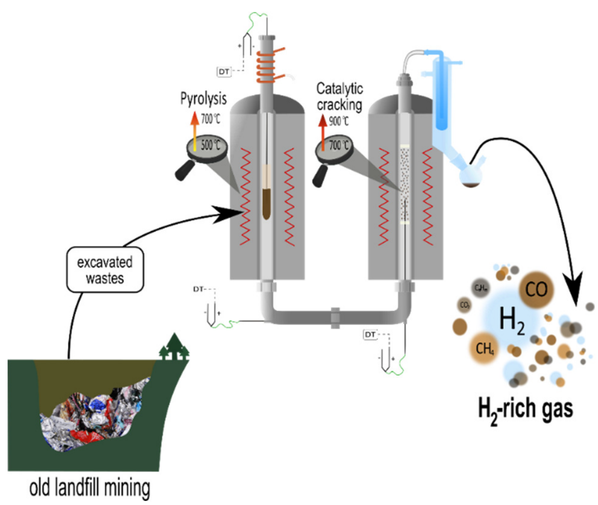

Figure 3. The general concept of the catalytic pyrolysis.

3.3 Performance and thermochemical mechanisms of plasma tar cracking

The main results show that the use of plasma can help to decrease the maximum temperature needed to crack naphthalene. **Figure 4** (right), shows that the temperature reduction is around 200°C vis-à-vis thermal cracking. In **Figure 4** (left), it is observed that the energy efficiency tends to increase with temperature, but once all naphthalene is cracked, the energy efficiency drops drastically. That means that plasma can be used to improve naphthalene cracking. Still, a careful optimisation is needed to get the best performance in terms of energy efficiency, by controlling the contribution of thermal tar cracking or by changing the plasma power.

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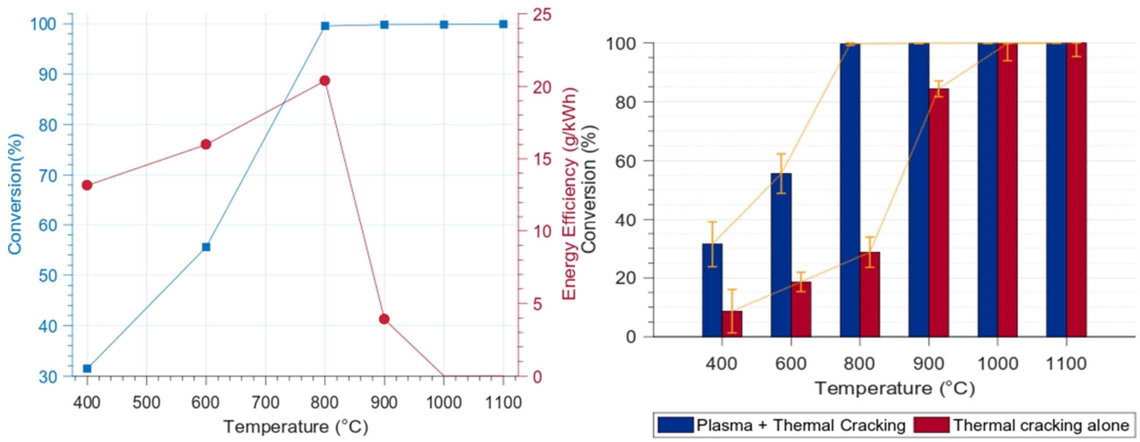

Figure 4. Left: Naphthalene conversion with plasma-assisted thermal cracking (blue) and energy efficiency of the naphthalene removal (red). Right: comparison of naphthalene removal by thermal cracking non-aided by plasma and thermal cracking aided by plasma.

3.4 Development of novel energy storage materials

All high-temperature gas-solid reactions suitable for thermal energy storage suffer from a loss of reactivity over consecutive cycles caused by thermal sintering. For this reason, the reactive materials were engineered by mixing them with support or dopant additives, necessary to achieve stable performance over long-term cycling. The investigated gas-solid reactions, based on oxidation-reduction or carbonation-calcination of metal oxides, are listed in **Table 1**.

Table 1. Characteristics of the investigated gas-solid reactions, based on oxidation-reduction or carbonation-calcination of metal oxides

Chemical couple	Temperature of the release of stored heat [°C]	Stable cycles
Fe_2O_3/Fe_3O_4	1290	30
$SrO/SrCO_3$	1090	100
Cu_2O/CuO	1025	100
Mn_2O_3/Mn_3O_4	820	100
Co_3O_4/CoO	790	100
$CaO/CaCO_3$	785	50
BaO_2/BaO	775	Unstable

The performance of the CuO-based granules was tested in a lab-scale packed bed reactor, which was also intended as a lab-scale demonstration of the real heat storage system. Thirty consecutive redox cycles were performed by switching the temperature of inflow air in the range 870-1150 °C. During the release phase of the stored heat, a spontaneous stabilisation of the temperature along the packed bed was observed at around the equilibrium temperature of Cu_2O/CuO in air, which is 1020°C. This event establishes the possibility of the heat storage system of providing heat at a constant value of temperature that also matches the requirement of steam gasification (i.e. above 800°C). The complete and fully reversible conversion was achieved for 30 cycles, without any evident sign of degradation of the system.

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3.5 The conceptualisation of solar-plasma hybrid gasification systems

The detailed concept of a novel integration of a concentrating solar power system, a solar thermochemical energy storage system, a high-temperature shaft gasification, and a plasma tar cracking process has been finalised. As seen in **Figure 5**, this integration will allow the flexible use of these technologies as a function of the climatic conditions. The technical assessment through process modelling and simulation is currently ongoing. Final results are expected soon.

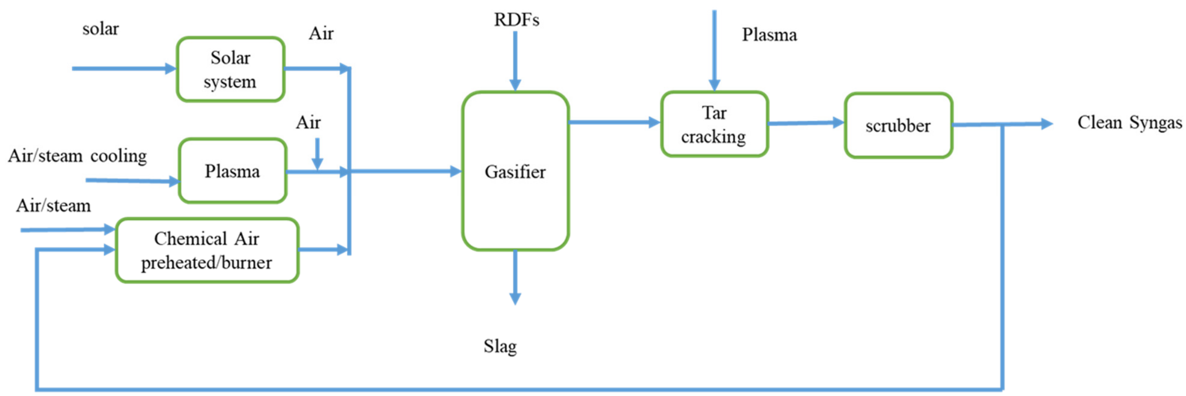

Figure 5. The proposed novel integration concept of a solar-plasma driven gasification process.

4. Conclusions and lessons learnt

Based on the activities within WP2, the following achievements have been accomplished:

1. Further exploration of the fundamental kinetic study and performance improvement of steam gasification for landfill-derived RDF.
2. Exploration of pyrolysis as a way for producing valuable materials from landfill-derived RDF.
3. Verification of tar removal processes for high-quality syngas productions through a plasma-based reactor.
4. Design and development of a solar thermal energy storage system for RDF conversion processes.
5. Conceptualisation and technical assessment of solar-plasma hybrid gasification systems for RDF conversion.

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5. List of publication

5.1 Published and submitted journal articles

1. Zaini I.N., Yang W., Jönsson P.G., *Steam gasification of solid recovered fuel char derived from landfill waste: A kinetic study*, Energy Procedia, Volume 142, 723-729 (2017) https://doi.org/10.1016/j.egypro.2017.12.118
2. Gomez-Rueda Y., Helsen L., *The role of plasma in syngas tar cracking*, Biomass Conversion and Biorefinery (2019) https://doi.org/10.1007/s13399-019-00461-x
3. Gigantino M., Kiwic D., Steinfeld A., *Thermochemical energy storage via isothermal carbonation-calcination cycles of MgO-stabilized SrO in the range of 1000 - 1100 °C*, Solar Energy, 188, 720-729 (2019) https://doi.org/10.1016/j.solener.2019.06.046
4. Parrodi J.C.H., Lucas H., Gigantino M., Sauve G., Esguerra J.L., Einhäupl P., ... & Krook J., *Integration of resource recovery into current waste management through (enhanced) landfill mining*, Detritus, 8, 141-156 (2019) https://doi.org/10.31025/2611-4135/2019.13884
5. Zaini I.N., García López C., Pretz T., Yang W., Jönsson P.G., *Characterization of pyrolysis products of high-ash excavated-waste and its char gasification reactivity and kinetics under a steam atmosphere*, Waste Management, Volume 97, 149-163 (2019) https://doi.org/10.1016/j.wasman.2019.08.001.
6. Gomez-Rueda Y., Zaini I.N., Yang W., Helsen L., *Thermal tar cracking enhanced by cold plasma -A study of naphthalene as tar surrogate*, Energy Conversion and Management, Volume 208 (2020) https://doi.org/10.1016/j.enconman.2020.112540.
7. Zaini I.N., Gomez Rueda Y., García López C., Ratnasari D.K., Helsen L., Pretz T., Jönsson P.G., Yang W., *Production of H_2-rich syngas from excavated landfill waste through a steam co-gasification with biochar*, Journal of Energy (under review after revision).
8. Gomez-Rueda Y., Zaini I.N., Yang W., Helsen L., *Purification of syngas obtained from MSW using recycled calcined shells in a secondary tar cracking unit,* Journal of Biomass and Bioenergy (under review).
9. Jagodzinska K., Zaini I.N., Svanberg R., Yang W., Jönsson P.G., *Pyrolysis of excavated waste - A comprehensive study on the process products*, Journal of Cleaner Production (under review).

5.2 Expected journal articles

1. Gigantino, M., Sas Brunser, S., Steinfeld, A. *Thermochemical energy storage via redox copper oxide couple: from particle design to packed-bed reactor engineering*. In preparation.
2. Gigantino, M., Notter, D., Steinfeld, A. *Pure and mixed metal oxides granules for high-temperature thermochemical energy storage via reversible redox reactions*. In preparation.
3. Gigantino, M., Notter, D., Bulfin, B., Steinfeld, A. *Redox kinetics and packed-bed reactor modelling of copper oxide-based granules for high-temperature thermochemical energy storage*. In preparation.
4. Gigantino, M., Zaini. I.N., Gomez-Rueda Y., Helsen L., Yang W., Steinfeld, A. *Energy analysis of hydrogen and liquid hydrocarbon production via hybrid solar-/plasma-assisted gasification of carbonaceous solid waste*. In preparation.
5. Jagodzinska K., Garcia-Lopez C., Pretz T., Yang W., Jönsson P.G., *Py-GC/MS and XRF characterisation of excavated waste fractions: Insights into a possible re-evaluation of excavated waste*, Waste management (to be submitted in June 2020).
6. Jagodzinska K., Yang W., Jönsson P.G., *Catalytic pyrolysis of excavated waste.* In preparation.

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7. Gomez-Rueda Y., Lieve H., *Steam and CO2 cracking of toluene and naphthalene as tar model molecules under plasma environment*. In preparation
8. Gomez-Rueda Y., Lieve H., *Real tar cracking and reforming under corona plasma at high temperatures*. In preparation.
9. Gomez-Rueda Y., Lieve H., *Simultaneous tar cracking and producer gas reforming in a plasma rector at high temperatures*. In preparation.
10. Zaini I.N., Jönsson P.G., Yang W., *Fragmentation behavior of refuse derived fuel pellets during thermochemical conversion processes.* In preparation.
11. Zaini I.N., Jönsson P.G., Yang W., *Integrated drying, high temperature gasification, and plasma tar cracking for conversion of high-ash landfill waste.* In preparation.
12. Zaini I.N., Nagy P., Jagodzinska K., Jönsson P.G., Yang W., *Synergistic effects on the syngas and tar formations during steam gasification of mixed plastic and paper waste.* In preparation.

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