

PYROLYSIS – AN ALTERNATIVE WAY OF RECYCLING

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Abstract In this article we will overview the possibility of using pyrolysis for the reuse of Municipal solid waste (MSW) and sludge. During the pyrolysis process, MSW is transformed to: i) fuel, ii) heat that can be used for drying the sludge or heat conversion to energy, iii) gases that can be burnt for electricity, stored and sold or used to move other generators and iv) some other products. Moreover, pyrolysis can be integrated with other technologies within a large integrated plant converting plastics, sludge and other waste materials into useful products. This work presents preliminary laboratory results of MSW pyrolysis and provide technological options for complex combinations of pyrolysis with other conversion processes. The combination of pyrolysis with other techniques shows promise in the alternative use of MSW and other waste in terms of energy and materials recovery.

Keywords:

Municipal solid waste, Pyrolysis, Gasification, Recycling, Integrated circular waste treatment facility.

1 Introduction

In line with advances in technology, we also invented new materials. One of these was plastic, which has many positive effects such as its light weight and versatile use, but it has serious implications for nature. For polymers, it seems logical to assume that we could convert plastic back to fossil fuel, reduce the production of new fossil fuels and also improve the conditions of natural habitats by reducing landfills and other dumping sites that have a negative effect on nature, because of plastic and other garbage that is hard to degenerate (Singh & Ruj, 2016; Thompson, 2009). The majority of plastic is produced for consumers and is sooner or later found in municipal solid waste (MSW). The production of MSW has increased over the years, but the technology to process it is advancing at a slower rate. In 2017, we produced 348 million tons of plastic (PlasticEurope, 2018), of which approximately 50 % is single use (Hopewell *et al.*, 2009).

2 Overview of technologies

With the advancement of technology and construction of cities, the pollution problem is becoming more and more complex. We need to advance in the field of waste management, because we create so much more waste every day that the old methods no longer work at the desired level.

2.1 Recycling

The best ecological treatment, that is commercial, is recycling. This is a process in which waste is treated and converted to new materials and products. With recycling, we have a source of raw materials. This reduces the greenhouse gasses, overall air pollution, water pollution and, above all, it reduces energy usage. The new raw materials can then be processed into new products (Geissdoerfer *et al.*, 2017). However, the quality of the recycled raw materials is not always good. Some materials are harder to recycle or even non-recyclable. Such materials cause problems in the recycling process and need to be separated before the rest is processed.

Plastic presents other problems, like additives which makes them into materials that are hard to recycle or even non-recyclable (Eriksen *et al.*, 2019). The non-recyclable materials are then transported to landfills. Some of the hard-to-recycle materials can be mechanically treated and used in lower quality products. For example, plastic can be shredded and then used in construction materials that can be used as a binder in paving blocks (Agyeman *et al.*, 2019).

2.2 Landfills

Landfills are another option for treating waste like MSW, although the waste loses some of its potential as an energy source and a source of new materials. According to some statistics, nearly 42 % of global waste is buried in landfills (Nizzeto *et al.*, 2016).

The garbage is processed, pressed as compact as possible and buried under the earth, then covered with different materials, to decompose. It decomposes through the process of thermo-oxidation and anaerobic degradation (Webb *et al.*, 2013; He *et al.*, 2019). The idea is that microbes in a controlled space, will decompose complex organic materials. The processes require time, workers and a correctly designed landfill.

One problem is the leaking of leachate. When rain inundates the landfill and percolates through it, it mixes with the dissolved materials and becomes leachate. If not contained properly, the adjacent soil and ground water can become contaminated, and microplastics can be released (Nizzeto *et al.*, 2016; He *et al.*, 2019).

Even if we unburied the waste, because of its physical-chemical properties, like contamination (Quaghebeur *et al.*, 2013) and partial degradation, it is mostly unsuitable, or at least uneconomical to recycle. (Canopoli *et al.*, 2018). There is also the option of using the excavated MSW and processing it to solid recovered fuel (SRF) with some profit (Ventosa *et al.*, 2014). On the other hand, pyrolysis offers an alternative, since some studies have shown that the excavated materials from landfills could be used as feedstock for a pyrolysis reactor. (Canopoli *et al.*, 2018).

2.3 Incineration

Another option is thermal disposal via incineration. In this process, the energy is recovered from the waste in the form of heat with combustion of the organic substances (Lino & Ismail, 2017). The heat can be used to produce electricity or to heat part of a city. From the residue, metals can be extracted and then recycled. During the combustion process, flue gas is formed, which contains many pollutants, such as CO₂, NO_x, and volatile organic compounds (VOC). Because of these, the flue gas must be cleaned before release into the atmosphere (Al-Salem *et al.*, 2009). However, some emissions are still present using current technologies, and the contamination of heavy metals persists (Wang *et al.*, 2019).

2.4 Gasification

Another form of recycling is gasification. In the process of gasification, organic matter or carbon-based materials are vaporized and transformed into a mixture of carbon monoxide, hydrogen and carbon dioxide at high temperatures. This mixture is also called synthetic gas (syngas). For this to happen, a controlled atmosphere with a gasification agent is needed, like oxygen or steam, and high temperatures of 700 °C or more (Panepinto *et al.*, 2015). Under this condition, syngas is formed, which can be used as a raw material for other products, like methanol or hydrogen, or, most of the time, as a fuel (Panepinto *et al.*, 2015, Higman & van der Burgt., 2008).

With the option of greater energy recovery, gasification plants are used in countries that lack other resources for electrical production, like Japan, or in some Scandinavian countries, which are among the leading countries in waste management (Panepinto *et al.*, 2015). Although it seems like a good alternative, gasification also has problems, like the tar problem. If tars, which can be up to 10 % of yield, are allowed to condensate, they can damage or disable pollution filter systems, and dangerous gases can be released into the atmosphere.

2.5 Pyrolysis

Pyrolysis is another alternative method to manage waste. The whole process happens at high temperature and in an inert atmosphere, like nitrogen. This causes thermal decomposition of the feedstock. The resulting gasses are then cooled to condense to form a liquid, tar, which can be used for fuel or as raw material. Some gases do not condense and can later be used as fuel or raw materials also. The reaction also leaves a solid residue, char, with a high carbon ratio (Sharuddin *et al.*, 2016). Pyrolysis is already used to some degree on a commercial level, but it has the potential to solve many waste problems, because it can be fueled with waste, like non-recycled plastic or MSW. Moreover, the products can then be used again in a different field with a positive energy balance (Abnisa *et al.*, 2014). Even waste excavated from landfills can be used as feedstock for pyrolysis, studies have shown (Canopoli *et al.*, 2018). Also, pyrolysis produces lower SO_x and NO_x emissions (Younan *et al.*, 2016), making the process more economical and ecological. The resulting liquid phases can be used as fuel directly or can be purified. A simple distillation may be enough for mixed plastic to be used as fuel (Wiriyampaiwong & Jamradloedluk, 2017).

Generally, for the pyrolysis process, we can use MSW, which differ in composition and type of contamination. Studies have shown that excavated MSW can be used (Canopoli *et al.*, 2018) and that metals like aluminum do not interfere with the process of pyrolysis (Ludlow-Palafox & Ha, 2001). Inorganic molecules can also function as a catalyzer, to reduce the temperature and the energy demand.

Additionally, new technologies are available, like microwave-assisted reactors. In this case, smaller particles of a microwave-absorbent material are mixed with the feedstock. This has the advantage that the heat is equally distributed. (Lam & Chase, 2012).

2.6 Other techniques

They are some new methods in the field of waste treatment that still lack proper research and large-scale applications. For example, there is plasma gasification. It uses plasma to convert organic materials into syngas (Moustakas *et al.*, 2005). Another method for treating MSW is hydrothermal treatment using subcritical water. The water used can have 234 °C and 3 MPa or 295 °C and 8 MPa to recover

MSW (Hwang *et al.*, 2012). Although there are many single methods available, it seems that most have flaws. So, it is only logical that we should combine some of these methods. Some facilities have already been built that first use pyrolysis and then gasification or incineration, for example, in Japan or Norway. Co-pyrolysis of biomass and plastic also shows promising results with lower consumption of energy (Breyer *et al.*, 2017).

2.7 Integrated waste treatment facility

For example, pyrolysis and gasification can be combined, as has been demonstrated in some plants in Japan. Moreover, the feedstocks can be mixed with two different wastes, e.g. the co-pyrolysis of biomass and plastic waste. In South Africa the paper industry creates 1200 tons of waste per month. Most of it consists of paper waste with sludge (PWS), which is rejected by the paper industry. It contains degraded fiber and some plastic waste (Brown *et al.*, 2019). Brown *et al.*, (2019) used rapid pyrolysis for the conversion of this waste. In rapid pyrolysis, the heating rate is a few hundred degrees Celsius per minute. The resulting oil has a higher heating value (HHV) of 22 MJ/kg at a yield of 60 %. The char remnants have an HHV around 20 MJ/kg (Ridout *et al.*, 2016). With this, they could reduce landfill and gain an economic resource.

Other studies have also shown the benefits of using co-pyrolysis, like Jin *et al.* (2019) in their studies of wheat straw and polyurethane. Some modeling has also been done by Oyedun *et al.* (2014), who proved that with the addition of biomass to plastic, we probably create a liquid heat medium and thus decrease energy inputs and costs.

Another modern waste type is sludge. With the increase in wastewater treatment, the amount of sludge is also increasing. For the drying of sludges, several new technologies have been developed, like vacuum drying. Moreover, with the synergistic effects of the biomass and the introduction of sludges into feedstock or pyrolysis, we could reduce energy costs while gaining a new resource. We can also perform pyrolysis of sludge in order to yield an additive for fertilizer (Samolada & Zabaniotou, 2014). With the implementation of these processes, we could also reduce waste water generation via a closed system and even recover heat.

In newer studies, the possibility of solar-assisted pyrolysis has also been analyzed. Ghenai *et al.* (2019) tested it in a laboratory reactor at 500 °C and atmospheric pressure. The tar had the density, dynamics, viscosity, calorific value and chemical structure of diesel fuel. They calculated that the pyrolysis process uses only 52 % of the created solar-grid energy. The rest could be sold at a decent price. With the use of solar panels, greenhouse emissions are also reduced. There is also the possibility of using geothermal energy or other alternative energy sources for the energy demands of the processing plant. With combinations like this, poorer countries with good solar days per year or alternative energy resources could decrease their waste and boost their economy with environmental benefits. For example, in Ghana, waste is a major factor in flooding, because it blocks the canals (Department, 2010) and consequently increases the likelihood of vector borne diseases (Clapp & Swanston, 2009). With pyrolysis, they could solve their problems, as demonstrated by Tulashie *et al.* (2019).

So, a combination of different methods should give better results. For example, the waste would arrive at an integrated processing facility, where it would be sorted, shredded and dried. With the addition of biomass, the energy demand would be decreased. Also, with solar panels or other alternative energy sources, the energy demand would be decreased.

3 Experimental

Pyrolysis could be an alternative in processing different wastes. In past projects we studied the possibility of treating MSW for the waste management company in Maribor. One option was pyrolysis of the solid recovered fuel (SRF). We studied pyrolysis of polyethylene (PE) and the MSW at different stages of the process. The hypothesis was that, if the SRF contains around 70 % plastic (or at least a major part), which the company claimed that it should have, it should behave like mixed plastic pyrolysis and yield positive results.

3.1 Set-up

We used a laboratory batch type reactor model for investigating the pyrolysis of SRF. Given the limitations of the project, we did not use any inert gases as the carrier gas. The liquid was sampled in the sample tube, where the gas products were burnt at the exit. For a comparison, we used PE.

3.2 Material

For the comparison with SRF, we used PE with high density (HD PE), which was bought at the store. All samples were chopped into smaller pieces to increase the surface for the reaction. For the SRF, we used undried SRF, dried SRF, SRF right after sorting and the SRF with a larger percentage of humus.

3.2.1 Drying

The drying process was carried out at 110 °C for 24 hours. The measured moisture content for SRF was 15 %. We also double-checked the results and dried the SRF in a vacuum oven at 30 °C for 1 day. This time the percentage was 12.61 %, from which we can conclude that the SRF normally have from 10-15 % moisture content.

3.2.2 Composition of SRF

For the optimization of the process, the feedstock composition should be known. For the physical and chemical properties, we used a homogenized air-dried sample with a moisture content of 2.761 %. The 3000 g of the sample were shredded into 2 mm pieces. The tests were carried out at Gorenje Surovina as a standard procedure.

4 Results of SRF and PE HD pyrolysis

All liquids were analyzed by a GC-FID and GC-MS. The gases were not analyzed but were only ignited.

4.1 Results of HD PE

We used 10.64 g of the HD PE feedstock. The temperature was measured at the connecting piece every 30 s to get a temperature trend. For safety reasons, the experiment was conducted at a maximum of 400 °C. According to the literature, we should increase the temperature up to 500 °C or even 600 °C.

We measured the mass of the feedstock and of the products, from which we calculated the mass efficiency (μ), which describes the ratio between the mass of liquid and gas products to the feedstock, and liquid efficiency, which describes the ratio between the mass of liquid products and the combination of liquid and gas products.

$$\mu = \frac{m_t + m_p}{m_s} = 55.55 \% \qquad \tau = \frac{m_t}{m_t + m_p} = 67.34 \%$$

The mass efficiency of HD PE was 55.55 % which is lower than in the literature. As said before, we were using laboratory distillation equipment and could not go above 400 °C. The liquid itself was clear orange in color, with a strong smell. The liquid and the gases burnt with an intense orange flame.

4.2 Results for SRF

For the calculations, we used dried SRF, although SRF with a larger percentage of biomass has shown great potential for further study.

$$\mu = \frac{m_t + m_p}{m_s} = 49.43 \% \qquad \tau = \frac{m_t}{m_t + m_p} = 55.83 \%$$

In the sample tube, we got two different phases: one a weak yellow and the other a brown-orange phase. We concluded that there was still some moisture left in the SRF, and depending on the type of SRF, we got a water phase with polar organic molecules. We separated the two phases and ignited the brown one, which burnt with an intense orange flame. We also got some solid products in an amount higher than in HD PE. These were ignited and burnt well. If we take the solid products into account, the overall yield increases.

4.3 Analysis of the liquid products

In order to determine the value of the liquid products, we analyzed them in a GC FID (Gas Chromatography - Flame Ionization Detector) and GC MS (Gas Chromatography – Mass Spectrometry). From a previous study, we obtained the caloric value of HD PE, which was 34.8 kJ/g (Roškarič, 2013). For the GC-FID, we used an HP-5 column (5 % phenylmethyl silicone). Then we prepared the temperature program: first heating for 3 min at 35 °C, followed by heating at a rate of 5 °C/min. After 10 min, we reached a final temperature of 280 °C. The whole program took 63 minutes.

The GC FID of HD PE has a typical graphic look, which was expected. From the temperature programs, we can calculate the boiling point of the components. We hoped HD PE and SRF would have similar results. However, the results show fewer components for SRF, which are more together. But we must also say that some of the peaks correspond to that of HD PE, so we do have some similarities.

We also conducted a GC-MS analysis with the same parameters as in GC-FID. The results are shown in the graphs below.

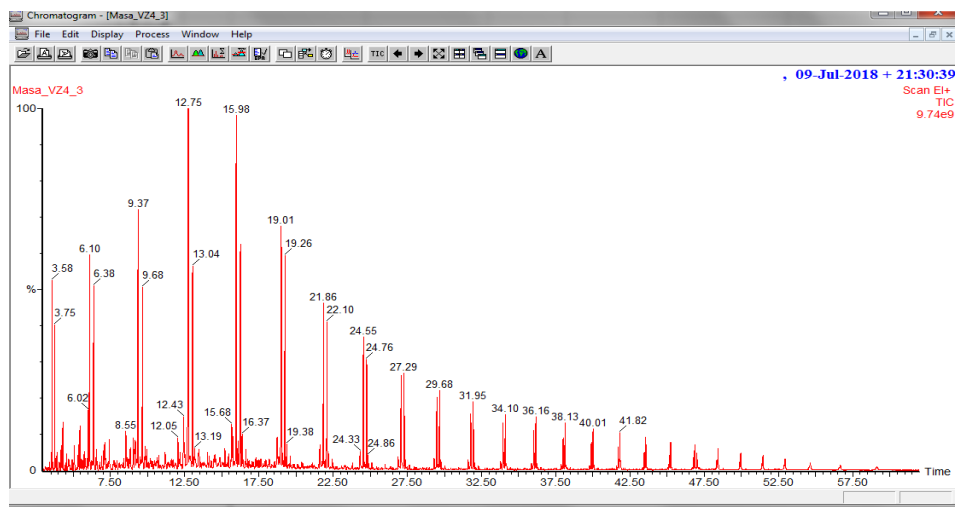


Figure 1: GC MS graph of HD PE.

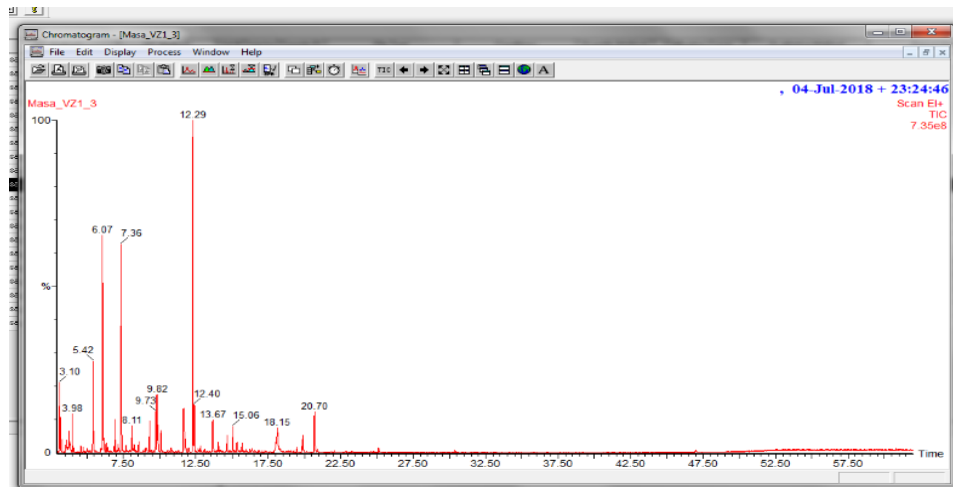


Figure 2: GC MS graph of SRF.

The GC MS of HD PE was a control group. Its results were, as expected, mostly C6 to C12 aliphatic components, much like in diesel oil. We analyzed SRF and a drop of SRF, which both gave nearly the same results. The library search showed that we got mostly an aromatic system or cyclic system with many heteroatoms, such as oxygen and nitrogen. An inert flow and a different rate of heating in pyrolysis could be more favorable for the aliphatic molecules which were desired. This also shows that the composition of MSW is not always what is said and that it is complex and unpredictable. Before using this oil, we would need to clean it or to install a proper filter for the gases that would be produced during incineration of this oil.

5 Integrated circular waste treatment facility

As shown before, the increase in waste plastic and MSW requires more advanced technologies for their processing. Figure 8 shows an integrated symbiosis plant for transforming wastes into energy, heat and products. The prices are estimated with the help of the computer programs Aspen Plus and Aspen Economics Analyzer and prices by the help of specifications provided by sellers following an internet search.

5.1 Input section

All the waste that comes in, undergoes a separation. With good separation at source, household waste would need only minimum separation. Several techniques can be used for separation, such as manual separation, robotic arms, near infrared (NIR) technology, gravity, magnetic, eddy current and others. If the stream goes to recycling, appropriate recycling procedures are taken. Rejected material then goes back with the other waste into the shredder and later to drying, if necessary.

Feedstocks with different compositions can be applied for different purposes. For example, with more PET in the plastic, more gases are created during pyrolysis (FakhrHoseini & Dastanian, 2013).

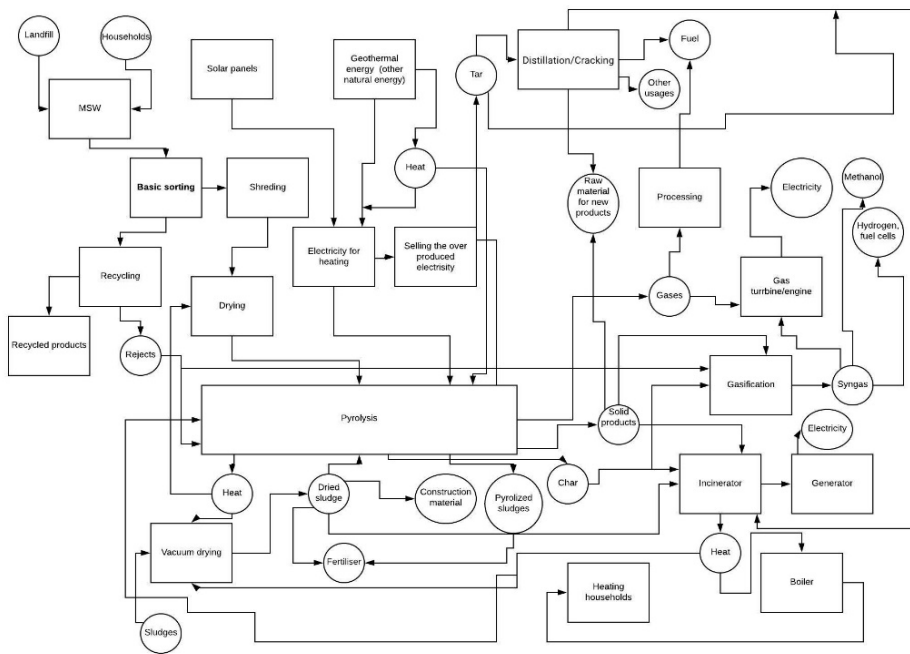


Figure 3: Integrated waste treatment plant (Several technologies can be integrated to achieve greater benefit. We can divide them in different sections.)

5.2 Drying of sludge

The investment for the drying facility would be around 1.2 million €, with a capacity of 100 tons per day for a 250 m³ reactor. The cost of the cooling water is 52 000 €/y and for the heating water 208 000 €/y. In an integrated circular waste processing facility, the required heat could be supplied from the cooling water from the pyrolysis or from other available sources in the plant.

5.3 Pyrolysis section

Depending on the size of the plant, different pyrolysis reactors could be used. With more medium-sized or small pyrolysis reactors, we could separately pyrolyze sludge for agricultural usage, MSW, sorted plastic or the recycled plastic, that cannot be recycled again. Two-step pyrolysis could also be an alternative.

The investment for a pyrolysis reactor starts at 500,000 € and can go up to 1,000,000 € for a medium-large reactor. With around 10 t per day, the electrical consumption would be 12,000 € a year. By using alternative energy and heating, we could reduce costs. For example, Lam *et al.* (2013) used microwave-assisted vacuum pyrolysis for plastic waste and cooking oil to achieve the synergistic effect. Moreover, they have proven that a vacuum system is better than a nitrogen flow, because it reduces the boiling point and lowers the temperature of pyrolysis, which saves money.

Investment in a pyrolysis plant that could process 100 t/h, which is 360,000 t per year, would cost 8 million USD \$ in total capital. The operational cost would be 30 million USD \$ per year. The estimated production of oil alone would be 122,927,000 L per year with a cost of production of 0.25 USD \$ per liter (with all the operational costs). With the diesel fuel price of 0.523 USD \$ per liter that they used, the yearly income would be 64 million USD \$. When we subtract 30 million USD \$ of production cost, and if we take into account unexpected costs, transportation and other costs, the profit should still be at least 20 million \$ per year. This is only a scale up of the scale up that they suggested. However, with the additional synergistic benefit of the integrated circular waste treatment plant, the annual cost should decrease.

5.4 Alternative energy sources

In order to reduce operating costs, alternative energy sources can be used. With electrical heating and solar panel systems, the cost of the pyrolysis and the carbon footprint would be reduced. Other alternative sources could be used, like geothermal energy.

5.5 Usage of the products

If sludge alone were pyrolyzed, active carbon could be produced and mixed with fertilizers to increase their effects. The liquid products can be burnt for electricity or used instead of fossil fuels. The tar could also be processed and then polymerized to produce new products. In this way, we would get products with the same high quality as those produced from fresh oil, and the plastic would recycle. Some research was done with promising results in this field (Achilias *et al.*, 2007). The gaseous products can be either processed or burnt for the electricity. The amount of waxes should be minimal, but they can also possess a high calorific value.

5.6 Gasification

The remaining char has two options. It can be burnt or used in the gasification process. With the gasification process, we could produce syngas from the remaining char. Some non-pyrolytic or unrecyclable waste could also be fed to the gasification reactor. It depends on the market demand whether more gaseous, liquid or solid products would be produced.

5.7 Incineration

The remaining waste, char or other products can also be incinerated in the thermal disposal section, where heat is generated and electricity produced. This step can be used for the remainder of the pyrolysis/gasification step and for the dried or pyrolyzed sludges.

6 Conclusions

With the increasing production of waste, like municipal solid waste or plastic waste, many new technologies have been developed to deal with the recovery of strategic resources. These technologies are emerging on the commercial level and will replace the old landfill system or incineration plants. Pyrolysis, gasification and hydrothermal treatment all possess serious potential but are lacking in other respects. Some need more pre-treatment, some are expensive, and for some operational costs are too high. On the other hand, most of them can process a large spectrum of MSW composition, which mechanical recycling does not offer. As has been shown in this study, different feedstock yields different results. The MSW sampled at Gorenje Surovina did not give great results, as compared to other mixed plastic studies. Nevertheless, a slightly different composition or a different reactor could provide better results. Additionally, if we process the MSW at the integrated circular waste treatment facility, many positive synergistic effects occur, so the recovery of resources is better, with lower operational costs. Further studies are required if such a facility is to be built, in order to find the optimum balance between the different techniques that yields the greatest benefit.

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