Abstract

The contribution aims to evaluate the effect of a cellulose-based bio-plastic waste (cellulose acetate) on the current Italian Municipal Solid Waste (MSW) management system. Three main scenarios were proposed, assuming the collection of cellulose acetate waste with organic, plastic and mixed waste streams respectively. The treatment routes of organic, plastic and mixed waste were considered as a combined anaerobic digestion and composting process, mechanical sorting and recycling and mechanical-biological treatment followed by incineration, respectively. For the scope, experimental activities have been carried out in order to estimate the performance of each treatment from a technical point of view, described as performance of the process and performance of the outputs. Based on the technical results, environmental and economic assessments have been carried out in order to evaluate the carbon footprint and the total cost of the whole waste management system for each scenario. From each evaluation, three indices were calculated and combined through a multi-criteria decision analysis to identify the preference of the different scenarios according to four decision-makers. Results have revealed how, despite the presence of cellulose acetate resulted in no effect or small improvement in the process for organic and plastic treatment, the outputs were strongly affected by the presence of bio-plastic. On the other hand, during mixed waste treatment, cellulose acetate did not influence the process as well as the output was slightly improved. These achievements were applicable also to environmental and economic analysis. Both evaluations revealed that cellulose acetate performed worst when handled with organic waste, while the treatment of plastic and mixed waste with cellulose acetate gave similar and better results. The main reason for this result was the non-conformity of compost quality when treating cellulose acetate with organic waste, which limited the use of compost in agriculture. These results were combined through the multi-criteria decision analysis approach, which showed a preference for the treatment of cellulose acetate with mixed waste, while the one in organic one was significantly low. Finally, the contribution suggests the necessity to upgrade the current organic waste management system to increase the performance of bio-plastic waste treatment, which is the strategy pursued by Italy.

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Giovanni Gadaleta
Treatment technologies of cellulose-based bio-plastic waste: an experimental and theoretical investigation
13

D. R. R. S.

Doctor of Philosophy in Risk and Environmental, Territorial and Building Development

Coordinator: Prof. Michele Mossa

XXXV CYCLE
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Department of Civil, Environmental, Land, Building Engineering and Chemistry

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POLITECNICO DI BARI

13
2022

Politecnico di Bari

LIBERATORIA PER L'ARCHIVIAZIONE DELLA TESI DI DOTTORATO

Al Magnifico Rettore
del Politecnico di Bari

Il/la sottoscritto/a GIOVANNI GADALETA nato/a a MOLFETTA (BA) il 30/06/1995 residente a MOLFETTA (BA) in via G. MARCONI N. 09 e-mail giovanni.gadaleta@poliba.it, iscritto al 3° anno di Corso di Dottorato di Ricerca in RISCHIO E SVILUPPO AMBIENTALE, TERRITORIALE ED EDILIZIO ciclo XXXV, ed essendo stato ammesso a sostenere l'esame finale con la prevista discussione della tesi dal titolo: TECNOLOGIE DI TRATTAMENTO DI RIFIUTI BIO-PLASTICI A BASE CELLULOSICA: INVESTIGAZIONE SPERIMENTALE E VALUTATIVA

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XXXV CYCLE
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Treatment technologies of cellulose-based bioplastic waste: an experimental and theoretical investigation

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DICATECh Dipartimento di Ingegneria Civile, Ambientale, del Territorio, Edile e di Chimica		
	Tecnologie di trattamento di rifiuti bio-plastici a base cellulosica: investigazione sperimentale e valutativa	
	Prof. Sabino De Gisi (relatore) Prof. Michele Notarnicola (relatore) DICATECh Politecnico di Bari, Italia Prof. Vito Iacobellis (tutor accademico) DICATECh Politecnico di Bari, Italia	
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EXTENDED ABSTRACT *(Eng)*

The effect of a growing use of conventional fossil-based plastic packaging has led to several concerns for human health and the environment. In order to prevent this pollution, bio-plastics have been identified as possible substitutes of conventional polymers. On the other hand, the effect of these materials on the current waste management system is still unclear and it differs for each single bio-plastic.

The present thesis aims to identify the most suitable treatment of a cellulose-based bio-plastic (cellulose acetate - CAT) waste. For the scope, three treatment routes in line with the current Italian waste management system have been hypothesized by collecting cellulose acetate with the organic, plastic, and mixed municipal waste stream and treated as follows: conventional biological process as anaerobic digestion and composting (organic waste treatment route); sorting and mechanical recycling (plastic waste treatment route); aerobic biostabilization and incineration (mixed waste treatment route). Technical, economic, and environmental assessments have been carried out through laboratory experiments, expenses-revenues analysis and Carbon Footprint value, respectively. The results from each assessment have been combined through a multi criteria decision analysis (MCDA) approach.

Results have revealed that anaerobic digestion and composting (organic waste treatment route) for cellulose acetate treatment was not efficient from a technical point of view since residues of non-degraded bio-plastic exceed the compost limit. In addition, economic and environmental assessment of this treatment have achieved the highest cost and impact respectively due to the landfilling of the produced compost.

The plastic waste treatment route has showed a strong effect on the quality of the recyclate, despite environmental benefit and the lowest cost have been achieved by this scenario. Finally, the mixed waste treatment route has not revealed any difference in the process of mixed waste with or without cellulose acetate, as well as the output of the system. The cost and the environmental benefit were nearly the same of the ones achieved by plastic treatment route, confirming how the main contribution of these assessments has been given by the landfilling of contaminated composting.
Combining these results, the MCDA have pointed out how mixed waste treatment route of CAT was the most suitable and efficient option, with a preference index of more than 90% for all the decision makers considered in the analysis.

These results have confirmed the necessity of upgrading the current anaerobic digestion and composting processes as well as a re-adaptation of standard regulations concerning the future bio-plastics trends framework. In addition, this work opens to other possible treatment for bio-plastics, which is possible only with a coordinate strategy.

Keywords: *Bio-plastics, Waste management system, Cellulose acetate, MCDA, Waste treatment.*

EXTENDED ABSTRACT *(ita)*

L'effetto di una continua crescita nell'uso delle plastiche fossili convenzionali da imballaggio ha portato a diversi problemi alla salute umana e all'ambiente. Per combattere tale inquinamento, le bio-plastiche sono state individuate come un possibile sostituto dei polimeri convenzionali. D'altra parte, l'effetto di tali materiali sul sistema di gestione dei rifiuti non è ancora completamente chiaro e risulta essere diverso per ogni singola bio-plastica.

La presente ricerca si è posta come obiettivo l'identificazione del trattamento maggiormente efficiente e sostenibile di una bio-plastica a base di cellulose (acetato di cellulosa) al termine del suo ciclo di vita. Per tale scopo, tre percorsi di trattamento sono stati ipotizzati in linea con il sistema attuale di gestione dei rifiuti in Italia associando l'acetato di cellulosa rispettivamente con i flussi di rifiuto urbano organico, di plastica e misto e soggetti ai seguenti trattamenti: processi biologici convenzionali come digestione anaerobica e compostaggio (percorso di trattamento del rifiuto organico); selezione e recupero meccanico (percorso di trattamento del rifiuto in plastica); biostabilizzazione aerobica e incenerimento (percorso di trattamento del rifiuto misto). Successivamente, valutazione tecniche, economiche e ambientali sono state svolte attraverso rispettivamente test in laboratorio, analisi di costi-guadagni e valore della Carbon Footprint. I risultati ottenuti dalle precedenti valutazioni sono stati combinati attraverso un approccio di decisione multicriteriale.

I risultati hanno mostrato come la digestione anaerobica e il compostaggio (percorso di trattamento del rifiuto organico) non hanno mostrato una efficienza tecnica adatta al trattamento dell'acetato di cellulosa in quanto diversi residui non degradati di bio-plastica superavano i limiti di qualità del compost finale. In aggiunta, le analisi

economiche ed ambientali di tale percorso hanno raggiunto i valori più alti di costo del sistema di gestione dei rifiuti e di impatto sull'ambiente a causa dello smaltimento in discarica del compost fuori-specifica.
Il percorso di trattamento del rifiuto in plastica ha mostrato un forte impatto sulla qualità del riciclato, nonostante tale scenario abbia raggiunto dei benefici ambientali e il costo minore di gestione. Per ultimo, il percorso di trattamento del rifiuto misto non ha rivelato alcuna differenza nel processo del rifiuto indifferenziato con o senza bio-plastiche, così come gli output del sistema. Il costo e i benefici ambientali sono approssimativamente simili a quelli ottenuti dal percorso di trattamento del rifiuto in plastica, confermando come il contributo principale di queste valutazioni è stato dato dallo smaltimento in discarica del compost fuori-specifica.
Combinando i risultati ottenuti, l'analisi decisionale basata su criteri molteplici ha mostrato come il percorso di trattamento del rifiuto misto per l'acetato di cellulosa risulta essere l'opzione di trattamento più adatta ed efficiente, ottenendo una preferenza superiore al 90%.
Tali risultati hanno confermato la necessità di aggiornare e potenziare i processi attuali di digestione anaerobica e compostaggio del rifiuto organico per il trattamento delle bio-plastica così come un riadattamento della normativa attuale riguardante lo scenario di gestione delle bio-plastiche al loro termine vita. Questo lavoro apre ad altri trattamenti per le bio-plastiche, possibili solo grazie ad un'azione coordinata di autorità internazionali.

Keywords: *Bio-plastiche, Sistema di gestione dei rifiuti, Acetato di cellulosa, MCDA, Trattamento dei rifiuti.*

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1. INTRODUCTION

1.1. The problem of plastic pollution

Nowadays, plastics are the most used material all over the world: due to its properties and the low production cost, plastics increased in this last centuries reaching a production of 57.9 million tons and a demand of 50.7 million tons only in Europe (PlasticsEurope, 2020). Plastics cover a wide range of applications like building, automotive, and agriculture but the largest use market (by weight) is packaging. Among these applications, the most common polymers are Low-Density Polyethylene (LDPE), High-Density Polyethylene (HDPE), Polypropylene (PP), Polyvinyl-chloride (PVC) and Polyethylene Terephthalate (PET) (Figure 1.1).

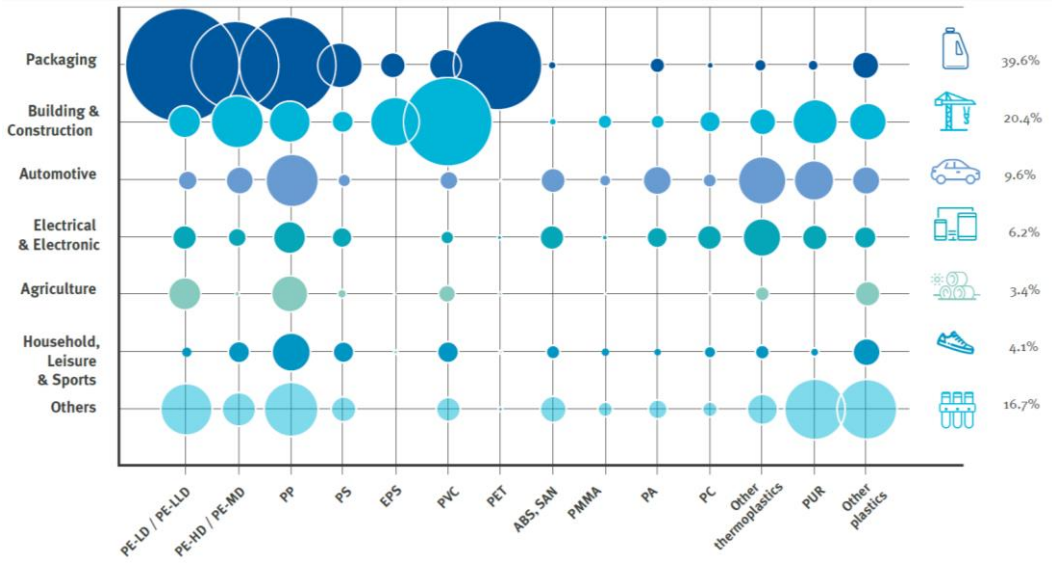


Figure 1.1 - Plastics demand by application and polymer type (PlasticsEurope, 2020).

In the last decades, Europe has implemented Municipal Solid Waste (MSW) management strategies with the aim of collecting this amount of plastic waste and recover them into new raw materials or energy. However, the amount of plastic waste disposed onto the landfill is still high (24.9% of the collected post-consumer plastic waste) (Geyer et al., 2017). In addition, waste management remains inadequate in many European countries, leading to a leakage of plastic waste in the environment. Once plastics reach the environment, their resistance and durability allow them to persist for hundreds of years. This leads to a higher likelihood of ecosystem exposure, and thus a higher chance of interaction, ingestion and hazardous effects across food webs (Horton and Barnes, 2020). If the climate change related to plastic production and management and the limited fossil fuel resources are considered too, it is clearer how governments, companies and scientists have to find alternatives to conventional plastics. At this point, rethinking plastic production, use and disposal is a key aspect for sustainable product development.

1.2. Bio-plastics

One possible solution to overcome the plastic pollution issue relies in bio-plastics. Bio-plastics are a wide range of materials with different properties and applications. According to the source and the biodegradability, the term bio-plastic means:

- Bio-based and biodegradable plastics: all that materials that are made from natural biomass as corn, sugarcane, or cellulose and present biodegradability trend;
- Bio-based and non-biodegradable plastics, as bio-PET, bio-PE or bio-PP, with identical features as their petrochemical ancestors but made from natural sources;
- Fossil-based and biodegradable plastics, which can be rapidly converted into simple compounds despite their fossil source.

Thus, the main advantages of bio-plastics are the saving of fossil resources by using biomass that regenerates and/or a faster degradation compared to their conventional counterparts.

In 2020, global bio-plastics production was around 2.42 million tonnes, about one percent of the 367 million tonnes of annual plastic produced. Bio-plastics market is continuously growing in amount and diversifying in possible application, which market value is expected to increase from US$3.02 billion in 2018 to US$12.4 billion in 2027 (Mazhandu et al., 2020). These materials are mostly produced in Asia (50%), but Europe strengthens its position as a major hub for the entire bioplastics industry (24% of the bio-plastics production) due to the high field of research and market.

Bioplastics cover a wide range of applications: from packaging, catering products, consumer electronics, automotive, agriculture/horticulture and toys to textiles and several other segments (Figure 1.2). Packaging, especially flexible one, remains the largest market for bioplastics with 48% of the total bioplastics market in 2021 (European Bioplastics, 2021). Indeed, the best-selling products are disposable garbage bags, soft packaging, rigid packaging and agricultural film but also sponge cloths, electronics applications, rigid materials (such as plates and cutlery), medical and agriculture products could be easily found on the market (Rujnić-Sokele and Pilipović, 2017).

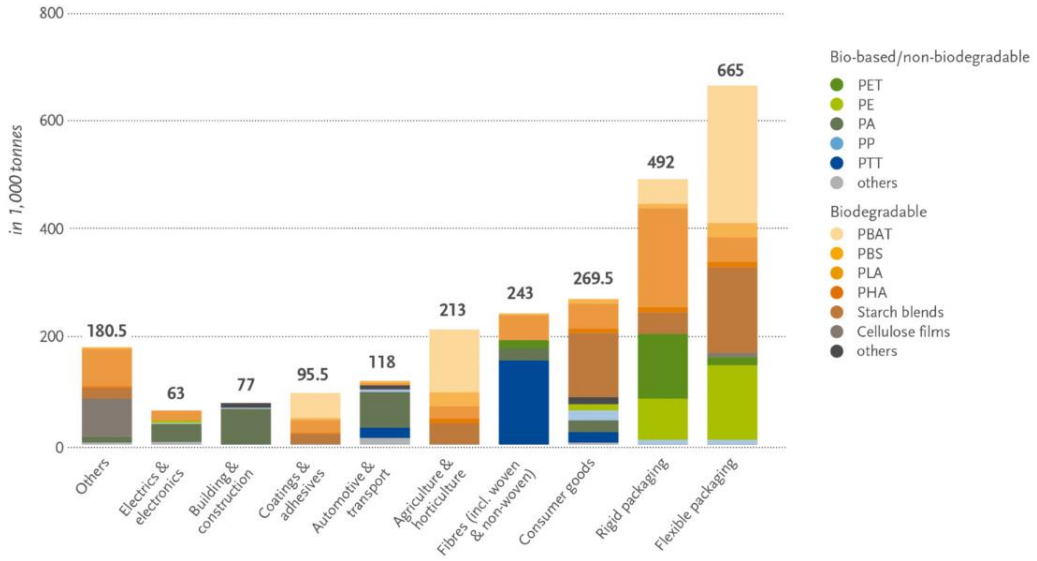


Figure 1.2 - Global production capacities of bioplastics in 2021 by market segment (European Bioplastics, 2021).

The number of bio-plastics available on the market is wide and continuously increasing but the most common ones are:

- Polylactic acids (PLA), polyhydroxyalkanoates (PHAs), polybutylene succinate (PBS), starch- and cellulose-based plastics for bio-based and biodegradable plastics;
- Bio-polyethylene (bio-PE), bio-polyethylene terephthalate (bio-PET) and bio-polypropylene (bio-PP) for bio-based and non-biodegradable plastics;
- Polybutylene adipate terephthalate (PBAT), polybutylene succinate (PBS) and polycaprolactone (PCL) for fossil-based and biodegradable plastics.

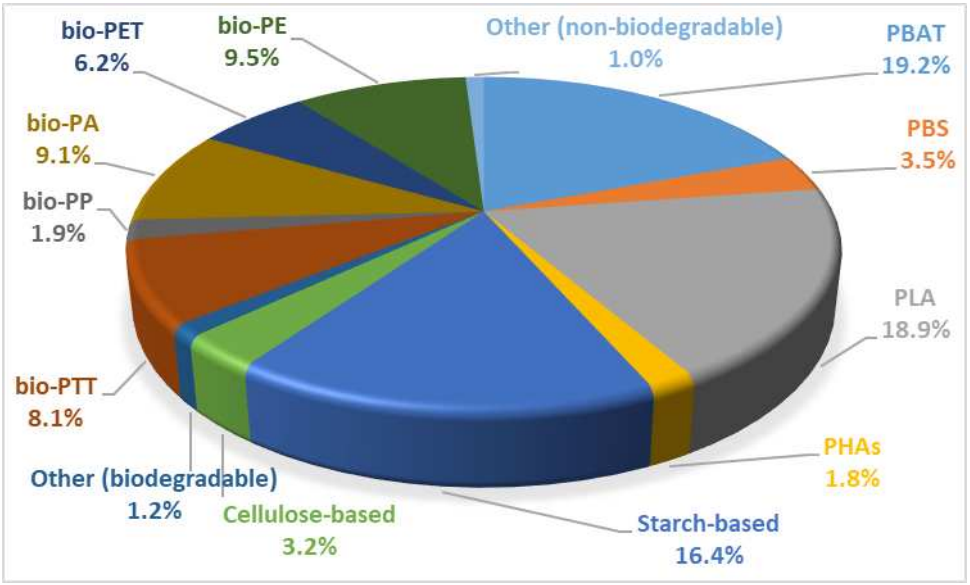


Figure 1.3 - Global production capacities of bioplastics in 2021 by material type (European Bioplastics, 2021).

Biodegradable plastics, regardless of natural or fossil source, account for 64.2% of the global bio-plastics production: among them, the most produced are starch-based plastics (16.4%), PLA (18.9%) and PBAT (19.2%). Despite PHAs cover only 1.8% of bio-plastics production (Figure 1.3), its capacity is estimated to increase almost tenfold in the next five years due to strong investments in Europe and USA. On the other hand, non-biodegradable and bio-based bio-plastics cover 35.8% of bio-plastics production. Along with bio-PE and bio-PET, which represent 9.5% and 6.2% of global bio-plastics production respectively, bio-polyamides (bio-PA) and bio-polytrimethylene terephthalate (PTT) cover an important segment of this amount (9.1% and 8.1% respectively). Non-biodegradable bio-plastics are predicted to decrease as the forecast for biodegradable plastics production shows a higher level of growth. This decrease will be more evident for bio-PET due to the introduction in 2023 of bio-PEF (polyethylene furanoate), a new polymer comparable to PET but with superior barrier and thermal properties for packaging application.

In economic terms, bio-plastics cost more than oil-based plastics, ranging between 2–6 €/kg, which is 2 to 5 times higher compared to the conventional plastics (Folino et

al., 2020). Nevertheless, some producers are willing to pay more for bio-plastics to protect the environment. Therefore, bio-plastics usage is going to increase rapidly in the next few years.

1.3. Biodegradability & compostability

Currently, there is still confusion regarding the link between bio-plastics and biodegradability. Considering the definition of ASTM Standard D-5488-84d, plastics can be considered "biodegradable" only if they are "capable of undergoing decomposition into carbon dioxide, methane, water, inorganic compounds, or biomass in which the predominant mechanism is the enzymatic action of microorganisms, which can be measured by standardized tests, in a specified period of time, reflecting available disposal condition". This means that the biodegradation refers to a biological process from microorganisms (bacteria, fungi, algae etc.) and not to all the physical degradation as mechanical fragmentation, photo-oxidation and thermal degradation. Many polymers that are claimed to be 'biodegradable' are in fact 'erodable', 'hydrodegradable' or 'photo-degradable'. It is clear that the biodegradation can occur in different environmental conditions: to avoid confusion regarding the biodegradability assessment and to normalize this wide field, several test methods have been developed according to the specific environment where the biodegradation is assessed (Table 1.1).

Table 1.1 - Main standards and principal parameters utilized to assess biodegradation in different environments.

Standard	Environment	Biodeg. Parameter	Volume [l]	N. samples	T [°C]	Days
Aerobic environment						
ISO 14855-1:2012: Determination of the ultimate aerobic biodegradability of plastic materials under controlled composting conditions — Method by analysis of	Composting	CO_2	>2	3	58	<180

evolved carbon dioxide — Part 1: General method						
ISO 14855-2:2018: Determination of the ultimate aerobic biodegradability of plastic materials under controlled composting conditions — Method by analysis of evolved carbon dioxide — Part 2: Gravimetric measurement of carbon dioxide evolved in a laboratory-scale test	Composting	CO_2	0.5	2	58	<180
ISO 17556:2019: Plastics — Determination of the ultimate aerobic biodegradability of plastic materials in soil by measuring the oxygen demand in a respirometer or the amount of carbon dioxide evolved	Soil	BOD/CO_2	-	3	20-28	<180
ASTM D5988-18: Standard test method for determining aerobic biodegradation of plastic materials in soil	Soil	CO_2	2-4	3	21	<180
ASTM D5209-91: Standard test method for determining the aerobic biodegradation of plastic materials in the presence of municipal sewage sludge	Sludge	CO_2	4	6	23	-
ASTM D5338-98: Standard test method for determining aerobic biodegradation of plastic materials under controlled composting conditions	Composting	CO_2	2-5	3	58	45
ISO 14851:2019: Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium — Method by	Aqueous	BOD	-	2	20-25	<180

measuring the oxygen demand in a closed respirometer						
ISO 14852:2021: Determination of the ultimate aerobic biodegradability of plastic materials in an aqueous medium — Method by analysis of evolved carbon dioxide	Aqueous	CO_2	-	2	20-25	<180
Anaerobic environment						
ISO 14853:2016: Plastics — Determination of the ultimate anaerobic biodegradation of plastic materials in an aqueous system — Method by measurement of biogas production	Liquid	Biogas	0.1-1	3	35	60
ISO 15985:2014: Plastics — Determination of the ultimate anaerobic biodegradation under high-solids anaerobic-digestion conditions — Method by analysis of released biogas	Solid	Biogas	>0.75	3	52	15
ISO 13975:2019: Plastics — Determination of the ultimate anaerobic biodegradation of plastic materials in controlled slurry digestion systems — Method by measurement of biogas production	Liquid	Biogas	1.5	2	35-55	60
ASTM D5210-92: Standard test method for determining the anaerobic biodegradation of plastic materials in the presence of municipal sewage sludge	Liquid	Biogas	0.16	3	35	14
ASTM D5511-02: Standard test method for determining anaerobic biodegradation of plastic materials under high-	Solid	Biogas	2	3	52	15

solids anaerobic-digestion conditions						
ASTM D5226-18: Standard test method for determining anaerobic biodegradation of plastic materials under accelerated landfill conditions	Landfill	Biogas	4-6	3	35	-

Among this wide range of standards, the aerobic biodegradability under industrial composting conditions and the anaerobic biodegradability under aqueous or high-solid state concretely concerns the waste management system. Each standard allows to assess different environmental conditions, and specific differences could be found among them. For example, ASTM D5338 and ISO 14855-1 recommend the use of compost and sample in a ratio of 6:1 and the commonly used quantity is 600 g of compost to 100 g of sample, whereas in ISO 14855-2 a 1:10 ratio of compost/sample is suggested. The basic concept is that aerobic and anaerobic biodegradability is calculated through the conversion of the plastics' organic carbon to CO_2 or biogas (CO_2 and CH_4), respectively.

In order to collect bio-plastics with the organic fraction of municipal solid waste, the biodegradability is not the only parameter that needs to be assessed: this is possible only if the plastic is certified as compostable. Compostable plastics have to be compliant to three criteria:

- Biodegradation: plastic biodegradability under controlled conditions should be greater than 90% within 6 months according to specific test methods (ISO 14855-1 or ASTM D5338);
- Disintegration: at the end of 12 weeks of industrial composting, no more than 10 % of the original dry weight of the sample remains after sieving on a 2.0-mm sieve, according to the international standard ISO 16929:2021 for pilot-scale disintegration test;
- Compost quality: the tested materials shall not adversely impact on the ability of compost to support plant growth by fulfilling the eco-toxicity test as described in OECD Guideline 208 (the germination speed and the vegetal

biomass of two species of plant from the tested compost have to be no less than 90% of the ones from a blank compost).

If plastics fulfil these requirements, they can be certified and marked as compostable. Nowadays, there is no unique label for compostable plastics, and various systems have emerged which are mostly limited to one or at best a few countries: DIN CERTECO in Germany, Consorzio Italiano Compostatori (CIC) in Italy, TÜV AUSTRIA in Austria and Biodegradable Products Institute in USA.

Thus, it is possible to note that, for example, a bio-plastics could be compliant to the biodegradability requirements but not to the disintegration ones or a bio-plastics could be disintegrable but not biodegradable. This means that a compostable product is always biodegradable, but a biodegradable product does not have to be compostable.

1.4. State of art on bio-plastics' biodegradability

Apart from bio-based and non-biodegradable bio-plastics as bio-PET, bio-PE and bio-PP, all the other bio-polymers present different degradation trends according to the material type and the environment where the biodegradation occurs.

1.4.1. Polylactic acid (PLA)

Due to its high mechanical strength, ease of processing and low cost, PLA is currently used in various sectors. The PLA monomers are produced through the fermentation of sugar extracted from corn, cane molasses, potatoes, sugar beets, etc. As investigated since a long time in both full and laboratory scale, the biodegradability of PLA is mainly influenced by temperature (Kijchavengkul et al., 2006; Rudnik and Briassoulis, 2011). In thermophilic condition (>58°C), PLA can be rapidly degraded under aerobic environment but slowly at ambient or mesophilic temperature (35-40 °C), even slower than other bio-polymers. Indeed, at 58°C PLA achieved a level of biodegradation of 84% in 58 days (Kale et al., 2007a) or even fully biodegraded in 28 days (Arrieta et al., 2014). Instead, the PLA weight loss achieved after 28 days were 39% and 13.8% at 30°C (Massardier-Nageotte et al., 2006) and 25°C (Adhikari et al.,

2016) respectively. In anaerobic environment, the results appears not clear as for the one under aerobic conditions. First studies demonstrated that under anaerobic conditions, no sign of degradation was found for PLA in long and short assessment (Bátori et al., 2018). On the other hand, recent studies have showed how PLA can reach high level of anaerobic biodegradation. At mesophilic conditions, more than 60% of PLA was biodegraded after 40 days (Itävaara et al., 2002) instead, at thermophilic conditions, over 90% of biodegradation was reached by PLA samples after 75 days (Yagi et al., 2009).

1.4.2. Polyhydroxyalkanoates (PHAs)

PHAs are thermoplastic polyester polymers synthesized by various kinds of bacteria through the fermentation of sugars or lipids present in agricultural raw materials (Tsang et al., 2019). Poly(3-hydroxybutyrate) (P3HB or PHB) is the major polymer belonging to the PHA class, which has been proposed in several short-term applications such as food packaging. Compared to other bio-polymers, PHB achieves the highest degradation rates in both mesophilic and thermophilic conditions in an anaerobic and aerobic environments, with lower values in the latter case. In aerobic environment, the degradation can be fast: after 15 days at 25°C and 65% relative humidity, a biodegradation level of 40-50% was reached (Arcos-Hernandez et al., 2012). This trend can continue over time until a fully degradation occurs (Boyandin et al., 2013). Increasing the temperature to mesophilic or thermophilic values, the degradation time decrease, achieving almost 80% of biodegradation in 28 days (Tabasi and Ajji, 2015). In anaerobic environments, the degradation is faster: 90% of biodegradation can be achieved after only 10 days of mesophilic anaerobic digestion (Yagi et al., 2014). Despite these results, there are not many other commercial PHA-based products for the narrow processing window, the high fragility and price, which is 5 to 10 times higher than those of petroleum-based plastics (Mazhandu et al., 2020).

1.4.3. Polybutylene succinate (PBS)

PBS is a biodegradable polymer produced from both renewable or non-renewable carbon sources, used as a substitute for conventional petrochemical polymer in the agricultural, medical, and dairy sector. The degradation of PBS is completely different according to the environment: in compost or soil, PBS is claimed to be biodegradable, but it does not degrade under anaerobic conditions. In 100 days, the aerobic degradation was 90% (Anstey et al., 2014) instead, after the same time, the anaerobic biodegradation of PBS was 2% (Cho et al., 2011) or even less (Yagi et al., 2013).

1.4.4. Poly (butylene adipate-co-terephthalate)(PBAT)

PBAT is an aliphatic–aromatic biodegradable polyester with excellent physical properties and significant degradation in several conditions. PBAT has been widely used in the production of blown film and its associated membrane products, especially in the last few years. The aerobic biodegradation of PBAT is well known, enough to fulfil the compostability requirements of EN 13432 and ASTM D6400: PBAT reached 80% of biodegradation after 45 days (Jian et al., 2020). In soil, PBAT can reach high biodegradation level: approximately 9% after 60 days and 21% after 180 days (Palsikowski et al., 2018). Unfortunately, research on the anaerobic biodegradation of PBAT is still limited, yet it is known that anaerobic biodegradability of PBAT is possible: after 25 days of anaerobic digestion in mesophilic conditions, 5.9% of PBAT samples were biodegraded (Ren et al., 2019) and with thermophilic temperature this value increased (Svoboda et al., 2019).

1.4.5. Polycaprolactone (PCL)

PCL is synthetic polyester with many industrial applications. PCL is degradable under aerobic conditions: at 55°C biodegradation of PCL achieved 38% after only 6 days (Nakasaki et al., 2006) but at 30°C it dropped to 7.6% in 28 days (Massardier-Nageotte et al., 2006). On the other hand, the anaerobic (bio)degradability of this polyester has been controversial. Some authors did not find any anaerobic biodegradation of PCL (Abou-Zeid et al., 2001; Kale et al., 2007b). Recent studies found that PCL is actually anaerobic biodegradable: in 139 days PCL was biodegraded

at 83% (Cho et al., 2011) and after 47 days at 92% (Yagi et al., 2009). In general, PCL degradation was slower than other polymers one (e.g., PHB) but it increased with a reduction in particle size.

1.4.6. Starch-based bio-plastics

Starch is a biopolymer with high availability, low cost, and biodegradability. The hydrophilicity and brittleness of natural starch require to convert it in a thermoplastic starch through the use of various plasticizers (glycerol, citric acid, urea etc.). Starch-based bio-plastics were the first biodegradable plastics to be commercialized and nowadays examples as MaterBi (composed of 60% starch and 40% biodegradable polymers) are worldwide known (Narancic et al., 2020). In a 12 week composting lab experiment with starch-based carrier bags a mass loss of 94–99% was observed (Adamcová et al., 2019). On the other hand, starch degradation can decrease up to 50% when the starch present is more than 10% of the organic matter in the substrate due to a decrease in enzymatic activity (German et al., 2011). In soil, the degradation can still achieve significant values: 14.2% in 110 days or 26.9% in 72 days (Gómez and Michel, 2013; Mohee et al., 2008). Anaerobically, the degradation of MaterBi products showed approximately 90% anaerobic degradability within 30 days (Bátori et al., 2018).

1.4.7. Cellulose-based bio-plastics

The presence of OH groups of cellulose results in a difficult processability of the biopolymer. For this reason, cellulose fibres from different plant residues such as rice straw, cotton, wheat or wood are converted through acetylation in an organic bio-polymer called cellulose acetate (Yadav and Hakkarainen, 2021). During the acetylation, a hydrogen atom in the cellulose is replaced with an acetyl group: the amount of acetyl groups per monomers in the substitution is called degree of substitution. A maximum degree of substitution of 3 could be done, generating cellulose acetate, diacetate and triacetate respectively. Thanks to its barrier capacity, transparency and impact strength, cellulose acetate could be used in the food

packaging sector. On the other hand, the acetylation does not always reach a suitable processability, which implies the use of plasticizer. In some cases, plasticizers do not link with the polymer matrix and they are expelled on the surface of the polymer (sweating).

Generally, in aerobic environment, after a first increase, the cellulose-based bio-plastics biodegradation proceeds slowly, requiring even more than 100 days to degrade completely. A weight loss of 33-41% was measured after 14 days of incubations under standard composting conditions (Mostafa et al., 2018) and cellulose-based sponge clothes were aerobically biodegraded for more than 80% at 58°C after 154 days (Vaverková and Adamcová, 2015). In soil, a complete degradation can occur after 105 days (Bilo et al., 2018). The degradation rate is dependent on the degree of substitution: high degree of substitutions shows lower biodegradation (Yadav and Hakkarainen, 2021). Biodegradation rates of 60% and 45% were achieved in 20 days for cellulose acetate with a degree of substitution of 1.8 and 2.3, respectively (Polman et al., 2021). The biodegradation of cellulose acetate can be increased with the addition of plasticizer as triacetine: the biodegradation of a cellulose acetate and triacetine (70/30) has increased of 100% the biodegradation of pure cellulose acetate (Phuong et al., 2014). The anaerobic degradation of cellulose-based bio-plastic is still an unexplored field. The disintegration of cellulose-based film can range from 57.4% to 93.4% in a liquid-state anaerobic digestion, resulting in a methane yield of about 0.4 l/gVS after 65 days (Zhang et al., 2018). On the other hand, the biodegradation is no more than 20% (Shrestha et al., 2020), revealing that the degradation is mainly physical, especially in environment rich of water where the cellulose can dissolve in. Cellulose-based biopolymers are mainly used to produce engineering plastics and sheets for electronic applications as they can be used to obtain either shrink films and/or sheets or to drive shape memory effects (Folino et al., 2020). Due to its hardness, good impact resistance and optical transparency, cellulose-based bio-plastics applications ranged from textile industry to packaging, photographic films and cigarette filter tows.

1.5. Bio-plastics & waste management system

At their end-of-life, bio-plastics become a post-consumer waste potentially suitable for mechanical recycling, organic recycling, and energy recovery. European countries have adopted for years MSW management strategies aimed at material and energy recovery (European Commission, 2018). Although the amount of bio-plastics waste is still low if compared to other waste fractions, the introduction into the current MSW management system of bio-plastic materials would have an impact on the entire chain (De Gisi et al., 2022). Indeed, beside the positive appeal of bio-plastics for consumers, lower disposal rates were registered for bio-plastics compared to conventional plastics (Taufik et al., 2020). Today, bio-plastics are not collected in a separate stream and, generally, they can be collected with other plastics, with organic waste, or in the residual waste (Hottle et al., 2017).

Generally, plastic waste requires sorting process in material recovery facility and then mechanical recycling. Nowadays, if bio-plastics were collected with plastic waste, they would impact this well-established system. Indeed, even the presence of 5% w/w of bio-plastics in a homogeneous plastic waste stream can affect in different degree the mechanical and thermal properties of the recycled polymers and the recycling process itself (Alaerts et al., 2018; Kuciel et al., 2018). Therefore, bio-plastics have to be removed during the sorting process and collected with other non-recyclable plastics. Despite adapted sorting equipment are available, bio-plastics sorting for mechanical recycling is rarely used on a commercial scale for different reasons:

- Only optical sensors can be adopted because other more common sorting technologies as visual discrimination and mechanical separation are not applicable for the similarity in appearance, weights, and densities between conventional and bio-plastics (Rujnić-Sokele and Pilipović, 2017);
- The number of bio-plastics is very wide and a specific sorting of each bio-plastics is not economically sustainable;
- The possible household-sorting of bio-plastics would require extra labeling, generating more confusion for the customers (Dilkes-Hoffman et al., 2019);

- The loss of mechanical properties affects the use of a fully recycled bio-plastics requiring a blend of virgin and recycled materials (Zhao et al., 2018);
- China's announcement that it would no longer accept international plastic waste for recycling from the beginning of 2018 has exacerbated the problem of mechanical recycling (Brooks et al., 2018).

If bio-plastics meet the industrial compostability criteria (UNI EN 13432 and UNI EN 14995, as described above), they can be treated with the organic fraction of MSW through anaerobic digestion and/or composting (Girotto et al., 2017).
Composting is an aerobic treatment that converts the organic waste in heat, humidity, and carbon dioxide (CO_2), producing a nutrient-rich compost that can be used as an agricultural amendment.
Anaerobic digestion treatment consists in a conversion of organic matter to biogas and digestate. Biogas is a blend of 50–70% methane (CH_4), 25–45% CO_2, and traces of hydrogen sulfide (H_2S), humidity, and other gases, that can be used as a source of renewable electric and thermal energy production. Digestate is rich in nutrients, but, due to a lower degradation than the one occurred during composting, it doesn't achieve suitable biological stability and requires further treatments (Gadaleta et al., 2021a).
In several studies it is pointed out how bio-plastics do not affect the composting treatment and the effect on soil of the compost produced (Gironi and Piemonte, 2011; Haider et al., 2019). On the other hand, the conditions of industrial composting and anaerobic digestion (e.g., temperature, retention time etc.) can present strong differences from the one occurred during biodegradability test. Thus, large amounts of non-degraded bioplastics remain at the end of the process, resulting in contamination of digestate and/or compost. For instance, Italian legislation admits only 0.5 % w/w of inert materials like glass, metals, and plastics without any distinction between biodegradable or conventional ones (Decreto Legislativo, 2010). In fact, plastics and even bio-plastics, are mechanically separated before the biological processes and are often disposed of in landfill (Cucina et al., 2021b).

The management of bio-plastics with mixed or residual waste implies several possible options, but the most common are biostabilization plant for a reduction of putrescible waste fractions, incineration facility for energy recovery or disposal in landfill. Research on the biostabilization treatment of bio-plastics is still unknown but the presence of bio-plastics do not represent a problem for the process due to a shorter retention time and a more heterogeneous waste processed than the organic waste treatment. Instead, waste-to-energy (WTE) incineration is not substantially affected by whether the input plastic is degradable or not. Compared to MSW incineration, combustion of bio-plastics waste does not produce more CO_2 in the biosphere. The CO_2 amount is recently captured and will be captured again when new bio-based products are produced, whereas incineration of fossil plastics emits CO_2 that had been sequestered for millions of years (van den Oever et al., 2017). On the other hand, abundance of micro-plastics has been found in the bottom ashes of incineration plants. If not properly managed, these ashes could migrate into the environment, increasing acid rain (Shen et al., 2021). Finally, landfilling management of bio-plastics is the least preferable yet the most popular option (Karan et al., 2019). In a landfill, bio-plastics degrade in semi-anaerobic conditions, producing CH_4 (a greenhouse gas with 25 times the effect of CO_2), which may then increase the greenhouse emissions if not recovered (Siddiqui and Khan, 2011).

2. AIM OF THE WORK

The amount of bio-plastics in MSW is still low if compared to other waste fractions (like conventional petrochemical plastics) therefore their influence in the current MSW management system is still not clear. However, bio-plastics could create or aggravate issues on the MSW management system (Calabrò and Grosso, 2018). In addition, the increasing spread of bio-plastics market is providing every day new bio-polymers, which effects on the MSW management system are even more unclear. To reach a sustainable system, these new materials require a coordinate and standardized waste management (Gadaleta et al., 2021b), and a wider research on the long-term role of bio-plastics on the waste management system. The spreading of the amount and type of bio-plastics in the waste streams represents a current issue, with social, technical, environmental end economics impact on the MSW management system. The suitability of waste treatment is different for each single bio-plastic and there is still ambiguity for the most suitable treatment of several bio-plastics waste.

In this context, the present study aims to evaluate the effects on the waste management system of a cellulose acetate bio-plastics, designed for active food packaging. For the scope, the collection routes of cellulose acetate waste have been hypothesized with organic, plastic and mixed waste streams (organic, plastic and mixed waste treatment route respectively). Then, in line with the current Italian waste management system, the relative waste treatment options have been identified and a comprehensive assessment has been carried out, according to three sustainability pillars as technical, environmental and economic. In particular:

- The technical assessment (seen as experimental investigation) aims to evaluate the performance of each cellulose acetate treatment process and the outputs

quality. For each waste treatment route, the technical assessment answers the following questions:

- Organic: does cellulose acetate contribute significantly to the biogas and CO_2 production during the anaerobic digestion and composting process? Does the presence of cellulose acetate influence the quality of the final compost?
- Plastic: what could be the size of cellulose acetate stream in a plastic sorting plant? Is the quality of the recycled plastics affected by the presence of cellulose acetate?
- Mixed: does cellulose acetate change the gas emissions during the process? What is the fate of cellulose acetate during the mixed waste treatment?

- Complementarily to the experimental investigation, a theoretical assessment composed of the evaluation of the environmental and economic impact of cellulose acetate waste on each waste management route has been investigated based on the technical results previously achieved:
  - For the first, the environmental impact has been estimated trough the Carbon Footprint (CF) value in order to calculate the greenhouse gas emission of each scenario;
  - For the latter, a cost-revenue analysis has been carried out to evaluate the cost of each waste route.
- Finally, the results from each pillar have been elaborated through Multi Criteria Decision Analysis (MCDA) methodology in order to identify the most preferred waste treatment route for cellulose acetate waste in the current system from technical, environmental and economic point of view.

The intention of this work was not only to point out the sustainability of each waste treatment route for cellulose acetate waste, but also to propose a replicable and reliable methodology for other bio-plastics waste useful for waste management decision-makers in dealing with a similar challenge.

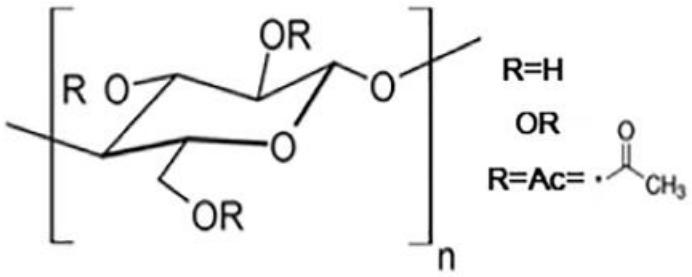
3. MATERIALS AND METHODS

3.1. Cellulose acetate

The focus of this work was a thermo-plasticised cellulose acetate, designed as an active food packaging. This bio-polymer was developed during the MultIFunctional poLymer cOmposites based on groWn matERials (MIFLOWER)" PRIN2017 project (grant number: 2017B7MMJ5_001), founded by the Italian Ministry of Education University and Research.

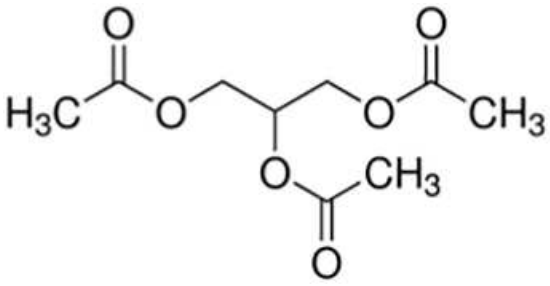
The pure cellulose acetate had a degree of substitution (DS – average number of hydrogen atoms replaced by acetylenic groups) equal to 2.45 and a specific gravity of 1.31. The melting temperature ranged between 230 and 250 °C. In order to enhance the processability of the polymer (reduced by the polar interaction network hydrogen bonding and dipole-dipole interactions) and increase its biodegradability, glycerol triacetate (triacetine, CAS number 102–76-1) was used as plasticizer. Triacetine is an eco-friendly plasticizer, commonly used with cellulose acetate since they share the same side functional groups (acetyl groups). The addition of triacetin allowed melt processing of cellulose acetate and enhanced the mechanical properties, to the point of fulfilling the packaging application requirements. Chemical structure and molecular formula of cellulose acetate and triacetine are showed in Figure 3.1.

a)



$C_{27}H_{37}O_{17}$

b)



$C_9H_{14}O_6$

Figure 3.1 - Chemical structure and molecular formula of cellulose acetate (a) and triacetine (b).

For the scope, the pure cellulose acetate was blended with 30% of triacetine. This content was proven to reduce the time to achieve a complete biodegradation from 200 to 46 days (Phuong et al., 2014). The Cellulose Acetate-Triacetine sample (CAT) was prepared through a two-steps procedure. At first, a Rheomix 600 Haake mixer was used for melt mixing the dried thermoplastic cellulose acetate pellets (supplied by GIBAPLAST – Italy) at 140 °C temperature and a 75 rpm rotational speed for 5 min. The resulting composite was then extruded with a twin-screw extruder (Thermo Scientific EuroLab 16 XL) at 40 rpm and 160 °C to form a continuous film of 150 μm thickness. A first thermo-gravimetric analysis of the CA sample was carried out with TGA (Figure 3.2). Pure cellulose acetate and triacetine have a degradation temperature of 370°C and 110-120°C respectively (Rodríguez et al., 2012; Zhu et al., 2013). The degradation of CAT samples did not occur in a specific temperature but proceeded in a wider range due to the addition of triacetine (which degrade at a lower temperature). Indeed, a first degradation of specimen areas rich in triacetine occurred at lower temperature

(between 120 and 270 °C). Then, the main degradation temperature (where the curve of Figure 3.2b reached the main peak) of CAT was 370°C.

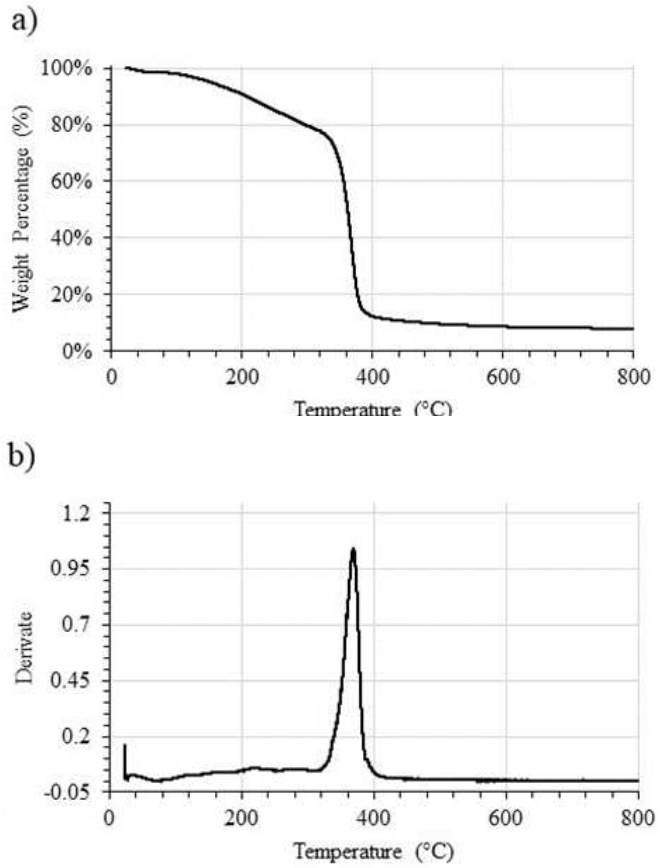


Figure 3.2 - TGA curve (a) and derivate (b) of CAT sample.

In addition, concerning DSC analysis, the same two processes were visible for the first and the second heating cycle respectively. During the first heating cycle (Fig. 3.3a), the evaporation of the moisture of the CAT samples and a first degradation of the triacetine occurred at a temperatures lower than 100°C. During this cycle, it was not possible to determine the glass transition temperature (T_g, the value that divide the glass and the rubber behaviour of the polymer matrix). The T_g value of 130°C was identified in the second heating cycle, where the curve showed an inflection (Fig. 3.3b).

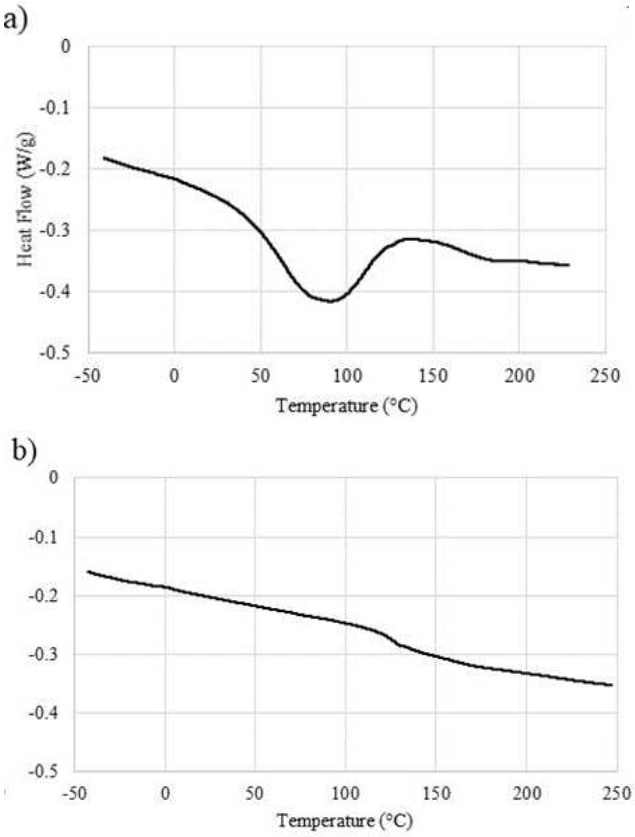


Figure 3.3 - DSC curves of CAT sample during the first (a) and the second (b) heating cycle.

Finally, from the analysis of FTIR spectrum (Figure 3.4), it is possible to investigate the structure and the chemical composition of the CAT sample.

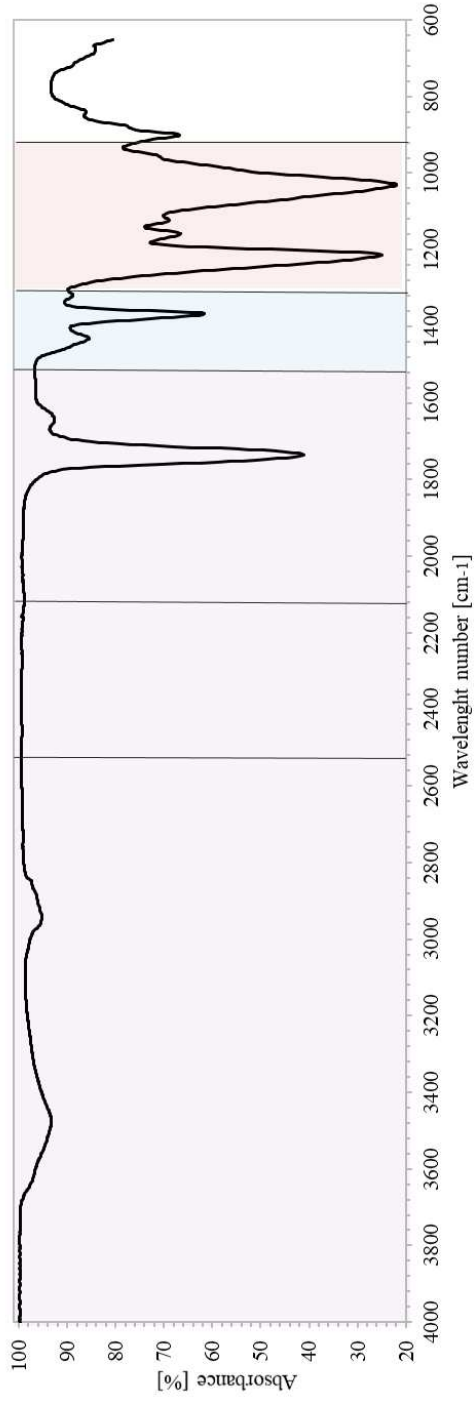


Figure 3.4 - FTIR spectrum of CAT sample.

In the spectrum of CAT, the board peaks with slight intensity at 3440 cm^{-1} and 2940 cm^{-1} are attributed to the moisture and the –OH links of cellulose and triacetine. In addition, the spectrum revealed the presence of four important peak at 1740 cm^{-1}, 1370 cm^{-1}, 1210 cm^{-1} and 1030 cm^{-1}. The first three are related to C=O, C–H and C–O links respectively. Those are the main components of acetyl group, which was substituted during the acetylation of the cellulose matrix. On the other hand, the peak at 1030 cm^{-1} represented the C–O–C chain typical of cellulose (Fei et al., 2017). Therefore, in view of the peak position, form and intensity, the absorption peaks of the acetyl group at 1740 cm^{-1} (C=O), 1370 cm^{-1} (C–H) and 1210 cm^{-1} (C–O) can be chosen as the CAT characteristic peaks, while the peak at 1050 cm^{-1} can be chosen as the reference characteristic peak of the cellulose matrix.

3.2. Waste treatment routes definition

The MCDA assessment was set according to the waste management system as described in Figure 3.5. Each single waste treatment route is described below.

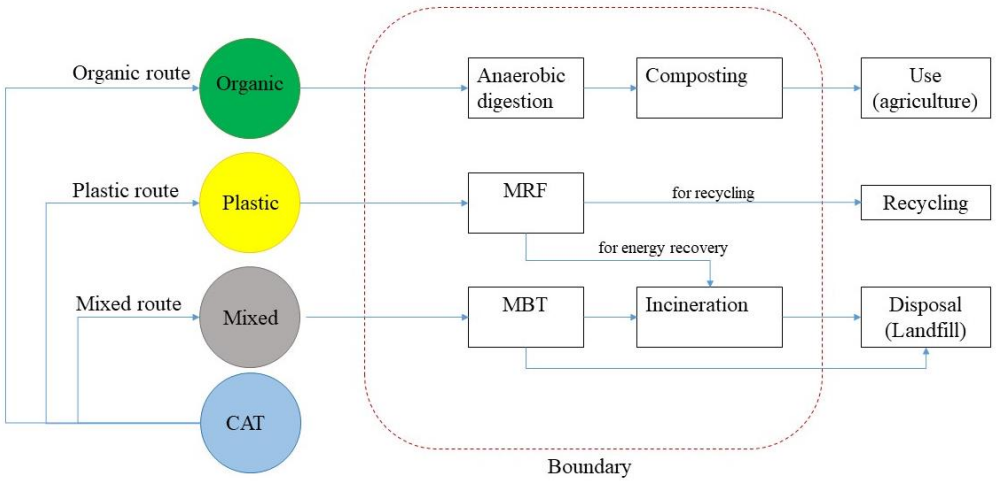


Figure 3.5 - Scheme of the waste management system and waste treatments routes hypothesized.

In this system, three different routes were identified by collecting bio-plastics (represented by CAT) with each waste fraction. Thus, organic, plastic and mixed waste treatment routes were proposed by assessing the collection of CAT in the organic, plastic and mixed waste stream respectively.

The treatment for organic waste was composed by a combined anaerobic and composting process. During anaerobic digestion, thermal and electric energy are recovered through a Combined Heat and Power (CHP) system. The output from anaerobic digestion is sent to composting in order to obtain a material rich in nutrients to be used in agriculture. The compos quality should be compliant with the Italian standards set by the (Decreto Legislativo, 2010). If the quality is not ensured because of a higher presence of contaminants (e.g., plastic residues), the compost is disposed in landfill as non-hazardous waste.

The treatment of plastic waste consisted in a first sorting in a Material Recovery Facility (MRF) where the plastic waste is selected in two main streams: one for mechanical recycling and the other, composed of a mix of non-recyclable plastics (PLASMIX), for energy recovery.

The treatment of mixed waste was in line with the Italian waste management system, where a Mechanical-Biological Treatment (MBT) is performed as a pre-treatment. The MBT aim is to increase the stability of the waste through an aerobic biostabilization. The biostabilite is then sorted into two streams: a Refuse-Derived Fuel (RDF) composed of dry waste as plastic, paper and textiles used for energy recovery; a Stabilized Organic Fraction (SOF) which is landfilled. RDF and PLASMIX are treated together in an incinerator for thermal and electric energy recovery through a CHP system.

3.3. Technical assessment

The suitability of each treatment route for CAT was first evaluated on a technical point of view. For the scope, the performance of each treatment was investigated through laboratory experiments or plant assessment, aiming to replicate each treatment. The aim of this assessment is to investigate the influence of CAT on the

treatment process and on the process output. Thus, a first analysis of the process was carried out for each treatment. Then the use of the outputs from each process was evaluated consistently to the use waste management system.

3.3.1. Organic waste treatment route

3.3.1.1. Objective

The organic waste disposal route aimed to assess the treatment of CAT with organic waste through anaerobic digestion and composting. Bio-plastics could generate more methane during anaerobic digestion but, on the other hand, they could require higher amount of oxygen for the their degradation during composting. Thus, the process was described through two parameters: 1) the methane yield during anaerobic digestion and 2) the oxygen consumption during the composting. The use of the final compost obtained at the end of the process was evaluated according to the quality required by the Italian legislation(Decreto Legislativo, 2010). The contamination of compost from plastic residues was considered as the key parameter to describe the compost quality. Indeed, if the limit for the presence of contaminants (plastics, metals and glass) with particle size > 2 mm is exceeded, the agricultural reuse of compost is not possible, and it is sent to landfill. A comprehensive assessment of this treatment route could be found in the work from Gadaleta et al. (2022a).

3.3.1.2. Materials

The organic fraction of MSW was reproduced on an experimental scale by the adoption of a Synthetic Food Waste (SFW), composed by different fractions (40.5 % fruit and vegetables, 18.1 % bakery products, 8.5 % dry products, 12.1% dairy products, 6.7% meat and fish, 14.1% ready meals) according to its current composition (Zhang et al., 2018). All the fresh fractions were manually cut to reach an average dimension of 10 mm. Then, two samples were created: a mixture of SFW and CAT (2%, Total Solid – TS - basis) in order to represent the amount of bio-plastics collected in this scenario and SFW only as a comparison. In particular, CAT specimens were used

as film squares of 25 × 25 mm, as suggested by international standard (ISO 20200, 2015).

The inoculum used for the anaerobic digestion was a stable anaerobic sludge from a local wastewater treatment plant with TS = 2.9%, Volatile Solids (VS) = 64.7%TS and pH = 7.5. Fresh compost (C/N = 14.6) and sawdust (pH = 4.9) were adopted during composting as seed and structuring, respectively.

3.3.1.3. Process description

According to current residence time (Molino et al., 2019), liquid-state anaerobic digestion was carried out for 21-days in mesophilic conditions (37 °C). 13 replicates (500 ml bottles) of SFW and SFW+CAT each were placed in a climate control room at 37±1 °C (Figure 3.6). Triplicates of microcrystalline cellulose and of inoculum only were adopted as reference (positive control) and blank samples (background biogas production from inoculum), respectively. An Inoculum-Substrate Ratio (ISR) of 2 (VS basis) was chosen to balance the buffer capacity and prevent the process inhibition for the acid formation (Holliger et al., 2016). Before starting, each test bottle was flushed with pure nitrogen for 1 min to ensure an anaerobic environment. The volume of biogas produced was measured through liquid displacement method by using a eudiometric tube filled with sodium chloride barrier solution (20% NaCl and 0.5% citric acid). Biogas volume and composition, air pressure and climate room temperature were read once per day or before every release of excess biogas from the system. After every recording, the bottles were shaken to enhance the mixture blending (Li et al., 2020). Biogas production and methane yield were calculated according to VDI (Verein Deutscher Ingenieure, 2016) without including to measure the background production from inoculum. At the end of the stage, three replicates for SFW and SFW+CAT samples were used to evaluate the digestate characterization and the CAT disintegration.

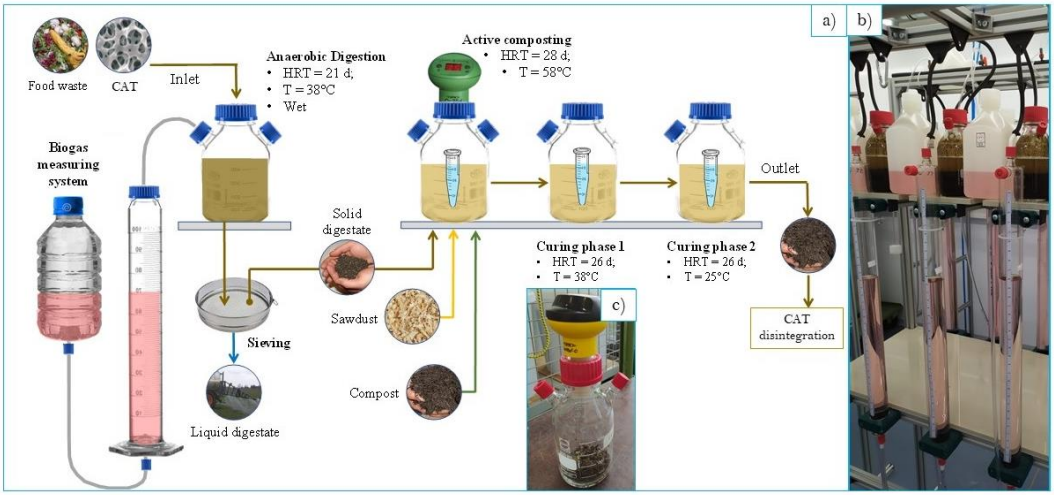


Figure 3.6 - Overall layout of the anaerobic digestion and composting experimental set-up (a), anaerobic digestion reactor and eudiometric tube system (b) and composting reactor equipped with OxiTop (c).

The transition from anaerobic digestion to composting stage required a filtering of digestate at 0.25 mm mesh to remove the liquid component from the solid digestate. Sawdust as a bulking agent and mature compost as inoculum (in a 1:2 and 1:10 w/w ratio of the solid digestate, respectively) were mixed with the solid digestate and the maximum water holding capacity of the mixture was adjusted to 50% by adding water (Fist method) (Bandini et al., 2020).

Then, three stages of composting were carried out after anaerobic digestion: one phase of active composting and two phases of curing composting, as performed by several composting plants (Evangelou et al., 2016). The active composting step consisted of a 28-days aerobic stabilisation at 58 °C with O_2 and moisture level control (Figure 3.6c). The same 500 ml bottles used for the anaerobic digestion were equipped with the pressure sensors OxiTop IS 6 system (WTW Wissenschaftlich-Technische Werkstatten GmbH, Weilheim, Germany) in order to capture the CO_2 produced during the process and measure the respective negative pressure generated (Heerenklage and Stegmann, 2005). The Oxygen Uptake Rate (OUR) [mgO_2/gTS] was determined stoichiometrically with the following equation (Binner et al., 2012) [Eq. (1)]:

$$OUR = \Delta p * \frac{M_{O_2}}{R * T} * \frac{V_{ges} - V_{abs} - V_{sample}}{m_{sample,dry}} \quad (1)$$

where Δp is the difference in pressure registered by the Oxytop [hPa], M_{02} is the molecular mass of O_2 (31,998 mg/mol), R is the ideal gas constant (83,140 l·hPa/(K·mol), T is the temperature (331.15 K), V_{ges} is the total volume (0.606 l), V_{abs} is the volume of medium CO_2 capture system (0.026 l), V_{sample} is the volume sample [l] and $m_{sample,dry}$ is the dry mass of sample [g TS].

From time to time, the negative pressure was released and the bottle was aerated for one minute (3.67 l/min). During the first week the preassure was released twice (or more) a day, instead for the rest of the experiment only once. The aerobic environment (>10% of oxygen) and a suitable moisture level were ensured by measuring gas composition and placing a tube of deionized water in the bottle.

At the end of active composting, OxyTop cups were removed and two phases of curing composting of 26-days each were carried out at 38°C and 25°C respectively. Samples were stirred and aerated (3.67 l/min flow) every 5 and 7 days for the first and second curing phases, respectively. At the end of the composting process, the final compost was weighted and characterized. In addition, at the end of each stage, 3 replicates were used to characterize the compost and to measure the CAT disintegration.

3.3.1.4. Quality of the output

The quality of the compost at the end of anaerobic and composting stages were evaluated through the amount of plastic residues in the final compost. In order to estimate the amount of CAT in the final compost, it was necessary to calculate the final disintegration of the samples. According to standard methods (ISO 20200, 2015), the degree of disintegration (D) was calculated as follows [Eq. (2)]:

$$D = \frac{m_i - m_r}{m_i} * 100 \quad (2)$$

where m_i is the initial dry mass of the test material and m_r is the dry mass of the residual test material recovered by sieving (2 mm). CAT samples were collected, washed with

deionized water, dried at 58°C for 3 days and weighed. The disintegration of the samples was evaluated at the end of every stage: anaerobic digestion, active composting, first and second curing composting.
The final compost quality (Q_c) has then calculated as follows [Eq. (3)]:

$$Q_c = \frac{p_i \times (1 - D)}{c_f} * 100 \quad (3)$$

where p_i is the initial wet mass of CAT, D is the CAT degree of disintegration at the end of the process and c_f is the wet mass of the final compost.

3.3.2. Plastic waste treatment route

3.3.2.1. Objective

If collected with municipal plastic waste, CAT are sorted in specific Material Recovery Facilities (MRFs). These plants separate the different plastic waste in homogeneous streams of specific types of polymers. Then, according to the polymer types, the streams can be recycled into new raw material or used as source for energy recovery. Polymers like PET, PE or LDPE are suitable for mechanical recycling but, other mix of plastics (PLASMIX) have not enough quality to ensure a mechanical recycling, leading in an energy recovery through incineration. In this system, it was investigated how CAT interacts with the MRFs and their outputs. The aim of this assessment was to evaluate the stream where generally bio-plastics are collected with and, for that, investigate how the presence of CAT affect its use (recycling or energy recovery). For the scope, a full-scale assessment was carried out to quantify the amount of bio-plastics in each output streams and compare their level with the technical specifications required. Then, for the one with the highest level of bio-plastics, it was investigated the effect of CAT in the further recovery step.

3.3.2.2. Process description

In this assessment, the treatment process was described through the sorting performed in the MRF and the further recovery of the output.

The sorting process was evaluated through a full-scale assessment of the amount of bio-plastics in an MRF, currently operating in Italy. The MRF scheme is described in Gadaleta et al. (2020): through a combination of automated and manual sorting stages, the plant is able to sort the municipal plastic waste from different cities in several homogeneous polymer streams. In particular, the main streams for material recovery selected by the plant are: light (CTL), light-blue (CTA) and colored (CTC) PET bottles; PE containers for liquids (CTE); PP mixed packaging (IPP); packaging film larger (FIL/M) or smaller (FIL/S) than an A3 sheet. In addition, the non-recyclable plastics are collected as PLASMIX and incinerated for energy recovery.

In this context, a sampling campaign was carried out in order to estimate the presence of bio-plastics in each plastic waste streams resulting after sorting in the MRF. A bale of each sorted plastic waste stream was opened and divided in 4 equaling weighting fractions: the sample size of each part was of about 100 kg. The amount of bio-plastics was sorted out and weighted, in order to estimate the percentage by weight of bio-plastics in each waste stream. These percentage were compared to the technical specification of each homogeneous plastic stream, imposed by the Italian authority for the plastic packaging recycling (COREPLA). These specifications point out the quality required by the plastic stream to be suitably recycle. The specification consider several plastics categories, which differ according to the plastic packaging sorted out. If some categories exceed the required specifications, the MRF is fined by COREPLA since the material is not enough pure for the recycling process and further screening are necessary. The bio-plastics detection limits present in the different technical specifications are summarized in Table 3.1.

Table 3.1 – Technical specification for each sorted plastic stream imposed by COREPLA.

Plastic stream		**Bio-plastics limit**
CTL	Light PET bottles	2.0%
CTA	Light-blue PET bottles	1.5%
CTC	Colored PET bottles	2.0%
CTE	PE containers for liquids	1.5%
IPP	PP mixed packaging	10.0%
FIL/M	Packaging film larger than an A3 sheet	5.5%
FIL/S	Packaging film smaller than an A3 sheet	7.0%

The recycling process was represented has a mechanical recovery through extrusion. For the scope, a co-rotating twin screw extruder HAAKE Rheomex CTW 100 OS (Thermo Fisher Scientific, Germany), equipped with PolyLab Monitor software system (Figure 3.7) was used. The screws (0.6 m long) were cleaned carefully before the extrusion. Then, the extruder temperatures were set on the software as follows: Z1 (feed) 155°C, Z2 160°C, Z3 165°C, Z4 170°C and Z5 (die) 175°C. When the temperatures were reached, the hopper was filled with the polymer's granules (300-500 g). The rotation of the twin screw was chosen as required to be used in the automatic mode. To prevent overheating, a cooling air flow was switched on. The extruded plastics from the die zone (5 mm diameter) was cooled directly after being discharged out in a water bath in order to maintain the thread shape.

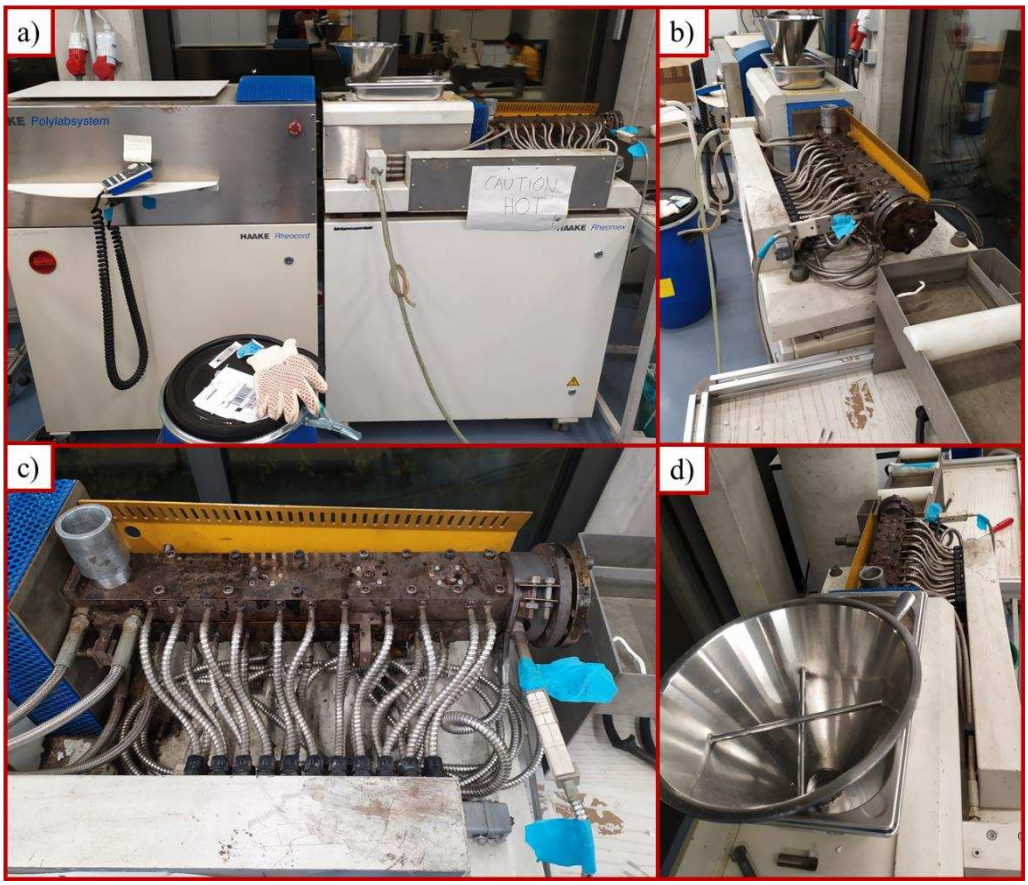


Figure 3.7 - HAAKE Rheomex extruder (a), die zone (b), extrusion channel (c) and feeding zone (d).

The influence of CAT in the recycling process was evaluated adopting five samples with different CAT content (0, 1, 5, 7.5 and 10%). CAT was used in pellet of about 50 mm dimension. The CAT was blended with a conventional LDPE (23H430) with the following properties: melt flow rate = 2.0 g/10 min; density = 923 kg/m^3; softening temperature = 95°C. Before the extrusion, CAT pellets were dried for 2 days at 58°C to remove the moisture accumulated during the storage.

3.3.2.3. Quality of the output

The recyclate quality was assessed according to different parameters. First, a visual inspection of the different extruded samples was carried out in order to evaluate

the blended LDPE processability. This parameter represents the most important parameter because it provides immediate information about the suitability of the material for mechanical recycling. The visual inspection was extended also with a Scanning Electron Microscope (SEM) analysis of the extruded specimens. An electron microscope FESEM-EDX Carl Zeiss Sigma 300 VP (Carl Zeiss Microscopy GmbH, Jena, Germany) was used, after sputtering the samples with graphite (Sputter Quorum Q150 from Quorum Technologies Ltd., East Sussex, UK).

In addition, a detailed investigation of the plastic specimens was carried out through thermo-physical analysis through Fourier Transform Infrared (FTIR) spectroscopy, Thermo-Gravimetric Analysis (TGA) and Differential Scanning Calorimetry (DSC) analysis.

The FTIR spectroscopy aimed to analyse the chemical structures of the samples. The chemical modifications induced by silsesquioxane structures on zein structures are evaluated by reflectance attenuate mode FTIR spectroscopy. FTIR spectra are collected at room temperature by using a Nicolet apparatus (Thermo Scientific, Italy) from 4000 to 600 cm^{-1} with a wavenumber resolution of 4 cm^{-1} for 64 scans.

TGA and DSC were carried out in order to investigate the thermal characteristics of the samples. TGA was carried out by using a TGA Q500 (TA Instruments, USA), submitting the samples to a heating run from 30°C up to 800°C at a heating rate of 10°C/min, under nitrogen flow. DSC analyses were performed under a nitrogen atmosphere using a differential scanning calorimeter (DSC Discovery–TA instrument, USA). Few micrograms (approximately 5–7 mg) were tested through a heating–cooling–heating procedure, with a scan rate of 50°C/min in the heating scan and 10°C in the cooling one. Temperature scan was performed from -50 to 200°C for all samples.

3.3.3. Mixed waste treatment route

3.3.3.1. Objective

This last waste treatment route was hypothesized to cover the limitations of the previous ones, as confirmed in scientific literature (De Gisi et al., 2022). In this way, the collection of bio-plastics with mixed MSW led to a mechanical and biological

treatment in specific MBT facilities. In these plants, the biodegradable fraction of mixed MSW is degraded aerobically in order to achieve a suitable stability for the landfill disposal. Before landfilling, the processed waste is screened to recover the dry fraction of waste (as plastic, paper and textiles) that, after specific treatment, is used as Refuse Derived Fuel (RDF) in incineration plant for energy recovery. In this context, this test aimed to assess how CAT influences the yield of an MBT process and how such degradation affects the use of CAT as RDF for incineration. For this, the MBT process was replicated on a lab scale where process parameters like oxygen consumption and CO_2 production were monitored (Gadaleta et al., 2022b). Then, to estimate the suitability of processed CAT as RDF, the Higher Heating Value (HHV) of CAT was measured (Gadaleta et al., 2022c).

3.3.3.2. Materials

The MSW sample used in this study was deliberately produced in the laboratory to eliminate sampling variability and seasonal fluctuations in waste composition. Using data from literature and operative MBT plants (De Gisi et al., 2018; Pantini et al., 2015; Trulli et al., 2018), a Synthetic Mixed Waste (SMW) was prepared mixing different MSW fractions as reported in Figure 3.8.

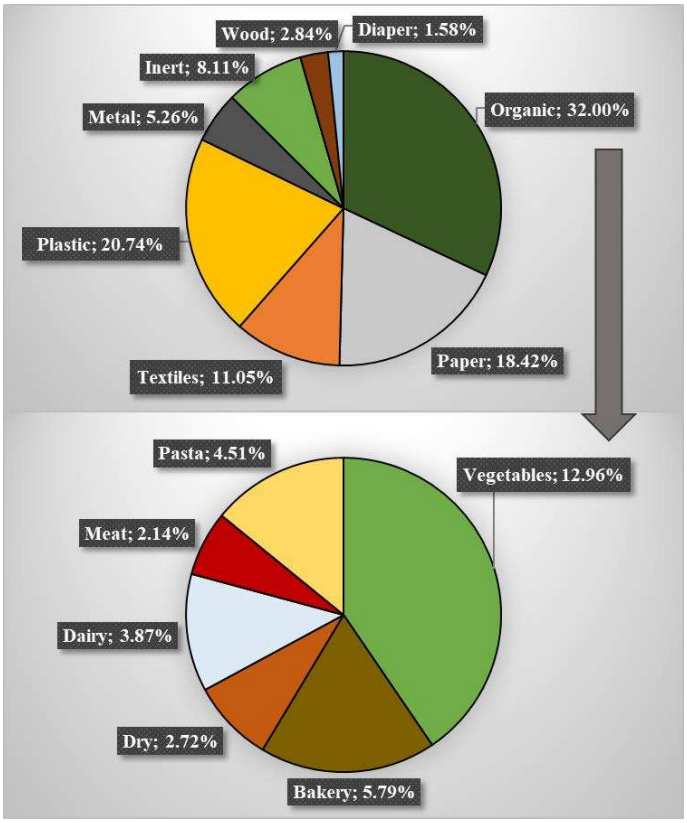


Figure 3.8 - Composition of SMW fractions.

The biodegradable fraction involved also the fine fraction of MSW, which is usually composed of organic materials (Gadaleta et al., 2021c; Grosso et al., 2016). Each fraction was manually cut or shredded with Retsch SM 300 cutting mill (Retsch GmbH, Haan, Germany) to reach a final dimension of about 20 mm. Then, all the fractions were mixed and, to enhance the biostabilization process, the initial SFW moisture content of 22% was increased to 56.5% by adding a specific amount of demineralized water (Fist method). The wet SFW sample had a weight of about 615 g, VS = 80.99%TS and density = 0.393 kg/l. According to the waste management scenario considered, 2% (wet basis) of CAT film (squares of 25 × 25 mm) was added to the SMW in order to have two set of samples: SMW only and SMW+CAT. Thereafter, samples were stored at 4°C in an airtight plastic bag for 12 hours before the beginning of the experiment.

3.3.3.3. Process description

To reproduce the MBT process, the self-induced bio-stabilization (temperature evolution that follows the natural trend of biomass degradation) was chosen, in order to evaluate the worst case of treatment. The process took place in a thermally isolated reactor with a controlled aeration and moisture level for 14 days, in line with the maximum residence time of current MBT plants (Gadaleta et al., 2022d). The reactor consisted in a cylindrical Dewar flask (KGW Isotherm) of 1.5 L capacity (D= 100 mm; h= 240 mm) covered by an aluminum coating, which isolated the samples from the outside and allowed the self-heating biostabilization (Serna-García et al., 2021). At the bottom of the reactors, an iron mesh was installed to separate leachate from the sample. On the top, the reactor was sealed with a three-ports cup for the waste aeration, the temperature monitoring and the gas outlet. An air flow of 64 l/d and 37 l/d (for the first 4 days and the rest of the test respectively) was ensured by regulating an air pump (Wisa DBGM, 5600 Wuppertal 12, FRG) of 220 l/h through a flowmeter manufactured by Krohne (Tom et al., 2016). The air flow was moistened by bubbling into demineralized water and connected on the iron mesh at the bottom of the Dewar flask, to aerate the waste from the bottom. The off-gases were measured volumetrically with a milli-gas counters (Ritter, MGC-1) and their composition was evaluated from a sampling port placed between the milli-gas counter and the Dewar flask outlet port. The design of the reactor is showed in Figure 3.9.

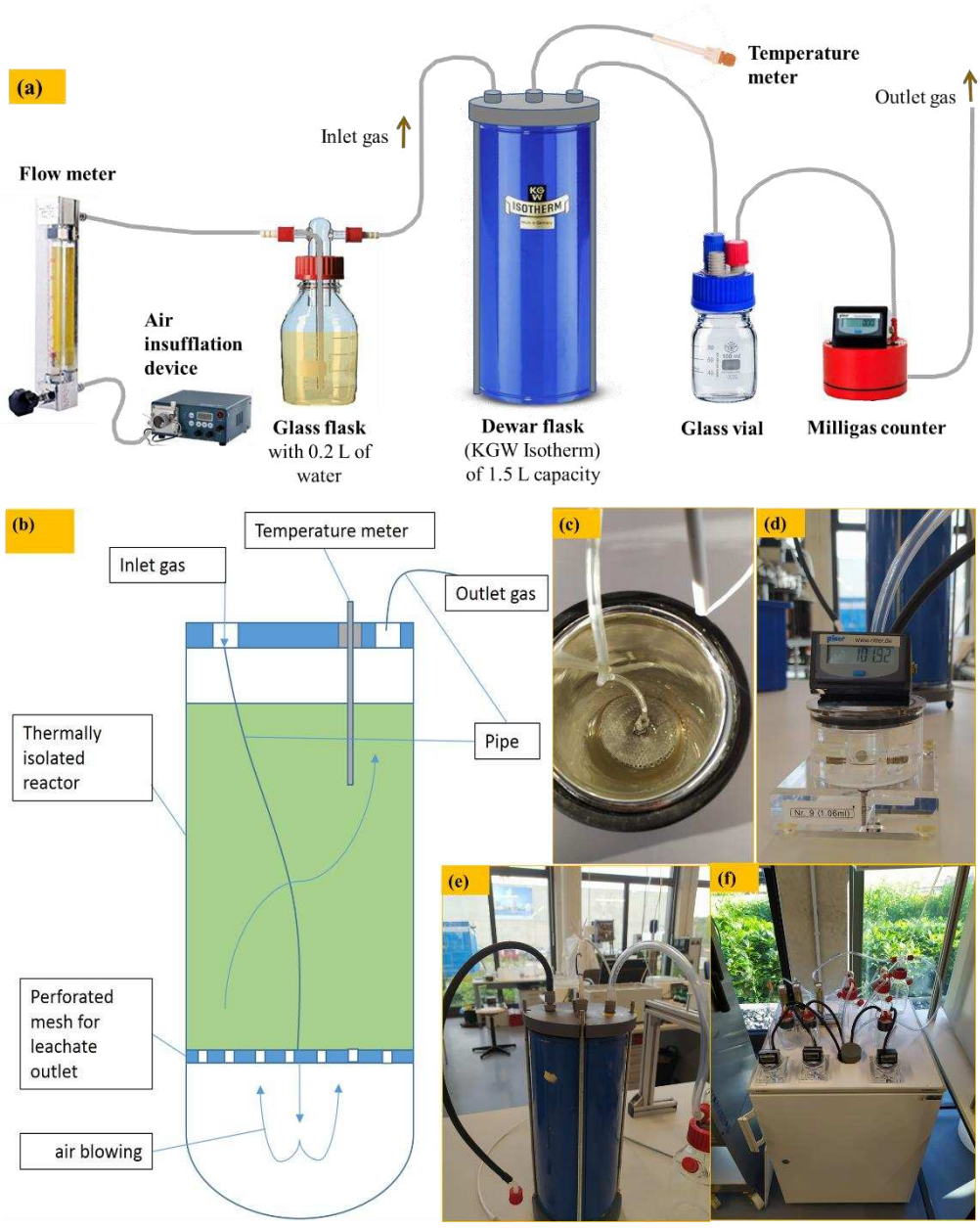


Figure 3.9 - Layout of the experimental set-up (a), Dewar reactor scheme (b), view of the waste/leachate separation grid (c), milligas counter (d), reactor head with three openings (e) and overall view of the Dewar reactor system in a temperature controlled environment (f).

Two samples were created for this test: SMW and the mixture of SMW and CAT processed for 14 days (SMW and SMW+CAT respectively).
The biostabilization process was monitored 3 times per day with the key parameters of the process: temperature (°C), airflow rate (l/d), oxygen consumption (g O_2/kg DM * d), carbon dioxide production (g CO_2/kg DM * d) and Respiratory Quotient (RQ).
Oxygen consumption S was calculated as suggested in Sohoo et al. (2021b) [Eq. (4)]:

$$S = 1.43 * P * \frac{(20.9 - O_{2,out})}{100} \quad (4)$$

where S = average rate of oxygen consumption [g O_2/kg DM/day]; P = airflow rate [l/kg DM/day]; O_{2out} = oxygen concentration in the off-gas [%, v/v] and 1.43 = conversion factor from O_2 volume to O_2 weight.
Carbon dioxide production C during the biostabilization was calculated as follow [Eq. (5)]:

$$C = 1.77 * P * \frac{CO_{2,out}}{100} \quad (5)$$

where C = average rate of carbon dioxide production [g CO_2/kg DM/day]; P = airflow rate [l/kg DM/day]; CO_{2out} = carbon dioxide concentration in the off-gas [%, v/v] and 1.77 = conversion factor from CO_2 volume to CO_2 weight (Di Lonardo et al., 2015).
The respiratory quotient (RQ) is a dimensionless number calculated from the ratio of carbon dioxide produced to oxygen consumed. RQ was calculated through the following formula [Eq. (6)]:

$$RQ = \frac{CO_{2,out}}{20.9 - O_{2,out}} \quad (6)$$

where CO_{2out} and O_{2out} are the carbon dioxide and oxygen concentration in the off-gas [%, v/v] respectively (CO_2 percentage in inlet air was neglected) instead 20.9 is the usual oxygen concentration of the airflow in input. RQ is rarely measured in MSW biostabilization processes, but it can be useful to understand how the process is

developing. Under aerobic conditions, RQ is approximately equal to 1, however its variability is still not clear because it depends on which macronutrients are being consumed during the process (Gea et al., 2004).

3.3.3.4. Quality of the output

In line with the waste management treatment of this scenario, the output from the biostabilization process is sent to an incineration plant, after specific pre-treatments. For the scope, the CAT specimens were investigated in order to assess their disintegration and their HHV. The first parameter was calculated as described above [Eq. (2)]. The HHV was measured in triplicate by a bomb calorimeter (model IKA C5000) using a recognized National Standard procedure (DIN EN 51900). Samples were dried (105°C for 3 days), screened to remove hard materials (glass and metals) and then pulverized by a mill (Retsch SM 300, Retsch GmbH, Haan, Germany). The values were adjusted considering a null HHV for glass and metals fractions.

3.4. Economic assessment

After the technical assessment, the treatment of CAT in the whole waste management system was evaluated from an economic point of view. With the economic assessment, each CAT treatment scenario was included in the waste management system and, through an estimation of expenses and revenues, the specific cost of each option was calculated. The economic assessment used the primary results from the technical evaluation and the detailed assessment is described in Gadaleta et al. (2022e).

3.4.1. Objective

The economic assessment aimed to estimate the cost per ton of waste processed in the waste management system for each treatment scenario. For the scope, a functional unit of 1 ton of waste, composed by organic, plastic, mixed and cellulose acetate waste, was considered. In line with the current amount of waste

produced in Italy, the organic, plastic and residual fraction of waste was 35.38%, 7.98% and 54.76% of the functional unit (ISPRA, 2021), respectively. To simulate the use of CAT as a reference for the bio-plastics market, the CAT content in the functional unit was considered equal to 1.88% (Figure 3.10). This amount was calculated by summing the amount of bio-plastics in each waste stream assessed. In particular, 4.0% and 1.1% of organic and plastic waste respectively is made of bio-plastics (Alaerts et al., 2018; Cucina et al., 2021a; Dolci et al., 2022). In addition, for the quantification of bio-plastics in the mixed waste stream, specific analyses performed in Italy were considered (IPLA, 2015), which revealed how bio-plastics are 0.58% of mixed waste.

This analysis was carried out under the waste management administration point of view, which means that all the waste treatment plants (except the landfill) were owned by the waste manager.

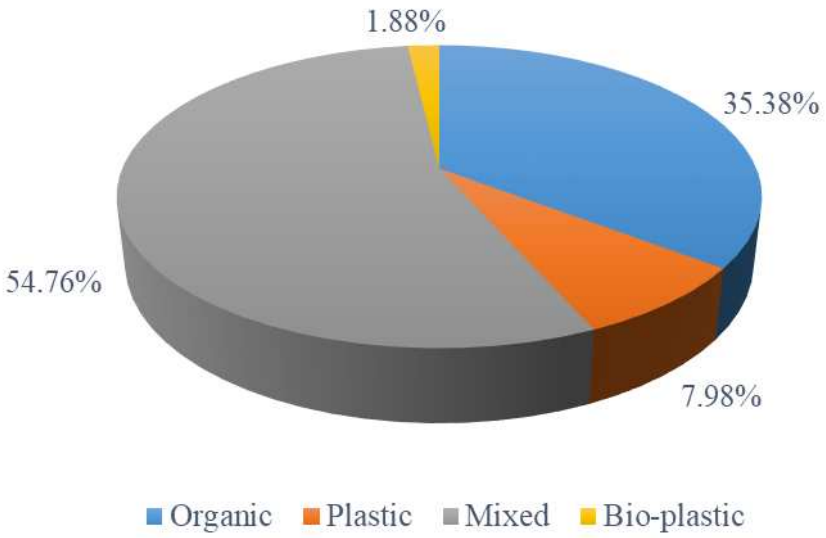


Figure 3.10 - Composition of economic analysis functional unit in terms of organic, plastic, mixed and bio-plastic waste fractions.

3.4.2. Expense & Revenues

The economic analysis was carried out estimating the expenses and revenues of the waste management system in each scenario (Table 3.2). Then, by subtracting the revenues from the expenses, the specific cost of each scenario was calculated.

Table 3.2 - Specific expenses and revenues in terms of €/ton used in the economic analysis.

Waste treatment	Expense/revenue	Unit	Ref.
Organic	Anaerobic digestion plant operational cost	105	(Gadaleta et al., 2021a)
	Composting plant operational cost	13	(Palese et al., 2020)
	Organic waste transportation cost	183	(D'Onza et al., 2016)
Plastic	Waste sorting revenue (recycling)	210	(Gadaleta et al., 2020)
	Waste sorting revenue (energy recovery)	75	(Gadaleta et al., 2020)
	Fines for waste contamination	4.25	(Gadaleta et al., 2020)
	MRF operational cost	159	(Gadaleta et al., 2020)
	Plastic waste transportation cost	224	(D'Onza et al., 2016)
Mixed	MBT operational cost	31	(Rigamonti et al., 2019)
	Incineration plant operational cost	31	(Bozorgirad et al., 2013)
	Mixed waste transportation cost	79	(D'Onza et al., 2016)
	Disposal in landfill tariff	120	(Ghosh and Di Maria, 2018)

Concerning expenses, only those related to plant operation, waste transportation and disposal were considered. The waste treatment tariff was not considered because of the assumption that the waste treatment facilities are owned by the waste management administration.

Regarding revenues, in addition to the ones presented in Table 3.2, those related with the waste management system were considered, as presented in section 3.2. For organic waste treatment, revenues came from the energy production during anaerobic digestion. For plastic waste treatment, the revenues from plastic waste sorting (for recycling and energy recovery) were considered. The revenues related to mixed waste treatment are made from the energy from incineration of processed mixed waste and sorted plastic waste for energy recovery. The prices for the selling of electric and thermal energy was consider equal to 0.2 and 0.045 €/kWh respectively (Gadaleta et al., 2021a).

3.4.3. Waste treatment description

The main waste treatment facilities were described and modelled according to different key parameters. Methane production during anaerobic digestion was chosen from laboratory test of technical assessment by an average of organic waste and CAT values. The energy from anaerobic digestion was calculated considering a High Heating Value (HHV) of biogas of 35.2 MJ/m^3 and an electrical and thermal efficiency of CHP system of 35% and 50% respectively (Gadaleta et al., 2021a). The sorting performance of MRF was modelled considering 47.26% and 52.74% of the input waste for recycling and energy recovery respectively (Gadaleta et al., 2020). The MBT plant was modelled as follow: 32.6% and 42.6% of the input were send to incineration and landfill respectively (Gadaleta et al., 2021c). For incineration, operating in CHP mode, the energy produced was calculated from the HHV of the waste stream in input. If it was adopted the HHV values of RDF, virgin and processed CAT from experimental tests, the HHV of PLASMIX was considered equal to 38,416 kJ/kg (Gala et al., 2020). The average incineration electrical and heat recovery efficiencies of 15% and 37.1%, respectively, were chosen (Di Maria and Sisani, 2018). Finally, the ashes post incineration process were set to 3% of the input (Xin-Gang et al., 2016). For the CAT degradation, it was adopted experimental results from the different laboratory tests.

3.5. Environmental assessment

Finally, the CAT treatment scenarios were investigated from an environmental point of view. For the scope, the Carbon Footprint (CF) was assumed as a decision support value. CF is defined as ‘‘the total set of greenhouse gas emissions caused directly and indirectly by an [individual, event, organisation, product] expressed as carbon dioxide equivalent (CO_2-eq)” (Marrucci et al., 2020). In this sense, the CF value can be used to estimate the emissions of greenhouse gases (GHG) associated with MSW treatments, evaluating the environmental impact in terms of climate change for each solutions, that is the calculation of Carbon Footprint (CF) (Pérez et al., 2018). In this work, the CF of each waste treatment scenario were calculated through the WRATE (Waste and Resource Assessment Tool for Environment) software, provided by Golder

(Gadaleta et al., 2022e). Thanks to its user-friendly view and its wide database, WRATE is the software suggested by the Italian Ministry of Ecological Transition for the regional waste management planning. Thus, the use of WRATE in this study strengthened the application of the results for the waste management planning in Italy.

3.5.1. Objective

The goal of this assessment was to evaluate the CF of the waste management system by varying the collection of bio-plastics (represented by CAT) with organic, plastic and mixed waste streams. Concerning scope, the assessment boundaries comprised the treatment and disposal of waste (collection and transportation of waste were neglected). The functional unit was set to 1 ton of waste, with the same composition of the one adopted in the economic assessment. Three scenarios were compared, referring to the one explained in the "Waste treatment routes definition" section. Any environmental burdens for energy and material costs arising during the manufacture or use of the waste were excluded in this study (zero burdens approach) (Oldfield et al., 2018).

3.5.2. Inventory

The inventory was based on the WRATE database Ecoinvent 2.1 and background data. The inputs were raw materials used in facilities, waste streams and energy for the MSW treatment. The outputs consisted of emissions into water and soil, residual waste amounts and recovered materials.

In line with the economic assessment, the functional unit was composed by 35.38% of organic waste, 7.98% of plastic waste, 54.76% of mixed waste and 1.88% of bio-plastics. The latter was modelled as a cellulose-based packaging, as provided by the WRATE database. WRATE database also provided waste streams sub-composition and properties.

To be consistent with the current Italian situation, the electricity mix was assumed equal to the "Medium carbon mix" available in WRATE, in which fossil fuels and natural gas represented the main primary energy sources.

The treatment technologies adopted in this study, briefly described below, are referred to the WRATE database.

The biological treatment was modelled as a combined anaerobic digestion and composting process. After a pre-sorting of extraneous waste with a rotary drum, the waste is anaerobically digested at mesophilic temperature for 3 weeks. At the end of the anaerobic digestion stage, the waste is dewatered through a press and the pressed digestate is composted for 3 weeks with intensive aeration and for two months in piles at ambient temperature (curing). It was assumed that the plant has an air emission treatment process and the wastewater produced is treated before release.

The Materials Recovery Facility (MRF) was modelled according to the current sorting plant for plastic waste in Italy. The sorting process adopted consists in a semi-automatic procedure including optical sensors, magnetic separators, eddy separators and manual sorting (Gadaleta et al., 2020). After sorting, valuable materials recovered are recycled and non-recyclable waste are sent to incineration for energy recovery.

Aerobic Mechanical Biological Treatment (MBT) consists in a first metals separation by magnetic sorting and a subsequent aerobic biostabilization of 10-14 days. Exhausted hot air from active waste is re-circulated into the waste to ensure high temperature. Finally, a screening allowed to separate dry high calorific fractions used as RDF in incineration and low calorific fractions, sent to landfill.

Concerning incineration, the grate furnaces technology was considered in this study. Energy is recovered from flue gases leaving the furnaces using a water-tube boiler and finned tube economiser. Fly and bottom ashes are recovered and disposed in landfill instead CO and NO_x abatment is performed with Ecotube system.

A sanitary landfill was chosen as waste final disposal. The surface of the landfill area is covered progressively with inert materials. No gas recovery was selected.

3.5.3. Impact assessment

The adopted impact assessment was IPCC-Fifth Assessment Report, using the Global Warming Potential (GWP) with a 100-year horizon (IPCC, 2013) as characterization factor. The impact category indicator used is CO_2 equivalent emissions

(kg CO_2-Eq), estimated as the weighted average of the emissions of each GHG and their corresponding GWP. The levels of carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) was evaluated, as these are the main direct GHGs regulated by the Kyoto Protocol and produced by various waste types through treatment plants (Marrucci et al., 2020).

3.6. Multi Criteria Decision Assessment

Through the MCDA approach, a comprehensive comparison of the three waste treatment routes for bio-plastics waste under investigation was carried out. Each result from technical, economic and environmental assessment was used as the key pillars in the MCDA. Indeed, as suggested by the EU research Centre (OECD-JRC, 2008), the MCDA objective is to select the best solution among a wide range of options or to provide a classification of alternatives for a specific problem, especially in the environmental field (De Gisi et al., 2015; Gadaleta et al., 2021b). The MCDA methodology consists in four different steps: (i) evaluation criteria; (ii) alternative matrix definition; (iii) relative weight adoption; (iv) preference index calculation (De Gisi et al., 2014).

3.6.1. Evaluation criteria

The different bio-plastics waste treatment routes were compared from a sustainable point of view through criteria related to technical, economic and environmental aspects. For the scope, the methodology adopts the following indices: performance (P) of treatment as technical criteria; avoided GHG emission (GHG_a) as environmental criteria (kgCO_2-Eq./ton); specific waste management system costs as economic criteria (C).

The performance P considered the suitability of each waste technology for the CAT treatment. It was composed by the sum of two parameters: performance of the process (P_p), which was referred on the influence that CAT has on the treatment process; performance of the output (P_o), which estimate the possible utilization of the process

output from each treatment. For both P_p and P_o, 7 qualitative classes were assigned to each waste treatment route, according to the results from the technical assessment: High Worsening (HW); Medium Worsening (MW); Slight Worsening (SW); No Effect (NE); Slight Improvement (SI); Medium Improvement (MI); High Improvement (HI). The qualitative values were converted in a quantitative form, giving the following values: HW = 0.071; MW = 0.214; SW = 0.357; NE = 0.500; SI = 0.643; MI = 0.786; HI = 0.929.

C represented the cost of 1 ton of waste in each waste management system, by varying the CAT collection, as calculated in the economic assessment.

The environmental index was represented by the GHG_a. It expressed the avoided $kgCO_2$-Eq. per ton of waste of each scenario compared to the worst scenario (the one with the highest CF value).

Finally, it has to be clarified whether a criteria has to be maximized (in the case it is a benefit) or minimized (in the case this is a cost) in order to provide the normalization of the alternative matrix. In the specific case, C has to be minimized, instead P and GHG_a have to be maximized.

3.6.2. Multi-criteria assessment

This section involves the alternative matrix definition, weight adoption and preference index calculation stages.

The alternative matrix was composed with the waste treatment routes as rows and the evaluation criteria as column. In order to compare the values of the different criteria, the alternative matrix has to be normalized recalculating the value in the range [0, 1]. Assumed the alternative matrix (n, m) with n number of rows and m number of columns, where x_{ij} is the value of the i-alternative toward the j-criteria, the x_{ij} normalized value ($\bar{x}_{ij}$) was calculated with the relations [Eq. 7] or [Eq. 8], if the criteria has to be maximized or minimized, respectively.

$$\bar{x}_{ij} = x_{ij} / Max\,(x_j) \tag{7}$$

$$\bar{x}_{ij} = Min\,(x_j) / x_{ij} \tag{8}$$

The adoption of weight were determined by means of the Paired Comparison technique (PCT) as defined by Mondy and Noe (2008) and then the Simple Additive Weighting (SAW) method (Hwang and Yoon, 1981). In order to verify the robustness of the obtained solution, a sensitivity analysis was carried out by varying the weights vector of the evaluation criteria (Soltani et al., 2015). For the scope, 4 types of decision makers were identified: environmentalist (Env), waste facilities manager (Man), policy maker (Pol); balanced (Bal). The decision-makers were first made aware of the purpose of the study and then interviewed, returning the vector of weights for each of them. The environmental decision-maker (Env) was the one who attached the greatest importance to pro-environmental criteria; the following condition was defined: $GHG_a > P > C$. On the other hand, the decision-maker in the waste treatment facilities (Man) was the one who attached the greatest importance to the technical criteria, the following condition was therefore defined: $P > C > GHG_a$. Similarly, the policy maker (Pol) was the one who attached the greatest importance to pro-management criteria; the following condition was defined: $C > P > GHG_a$. Finally, the balanced decision-maker (Bal) was what he considered the 3 indicators to be equally important.

In order to compare the three waste treatments routes, the composite indicator Preference Index (PI) is constructed. PI is an aggregator index, itself constructed from the GHG_a, C and P indices, calculated with [Eq. 9].

$$PI_i = \sum_{j=1}^{3} \bar{x}_{ij} * w_j \tag{9}$$

where, $\bar{x}_{ij}$ is the normalized value of the alternative *i*-th (each waste treatment route) with respect to the *j*-th evaluation criterion (GHG_a, C, P) and w_j is the weight for the *j*-th evaluation criteria. The best scenario is the one with the highest PI value.

4. RESULTS

In this section, the results obtained during the three assessments and the following MCDA application are presented. Firstly, the technical results from the organic, plastic and mixed waste treatment routes will be discussed; then, on the basis of their results, the economic and environmental assessment will be presented. Lastly, the results of these three assessments will be condensed in the MCDA in order to determine the best waste treatment route to adopt for cellulose-based bio-plastics waste.

4.1. Technical assessment

4.1.1. Organic waste treatment route

Figure 4.1 shows the cumulative average methane production of the two samples: the mixture of CAT and SFW (SFW+CAT) and the SFW only (SFW). These are net values, whereby the background methane production from the inoculum was subtracted. After 21 days of anaerobic digestion, SFW+CAT and SFW generated 2,184.68 and 2,069.40 ml CH_4, respectively. Most of the methane was produced after almost 3 days, confirming a fast degradation of the food waste (Elbeshbishy et al., 2012). The SFW was completely digested after 8 days for the SFW+CAT sample, as suggested by the plateau in the curve. A small surplus of methane production (115.28 ml CH_4) was noticed for SFW+CAT sample attributed to the bio-plastics degradation.

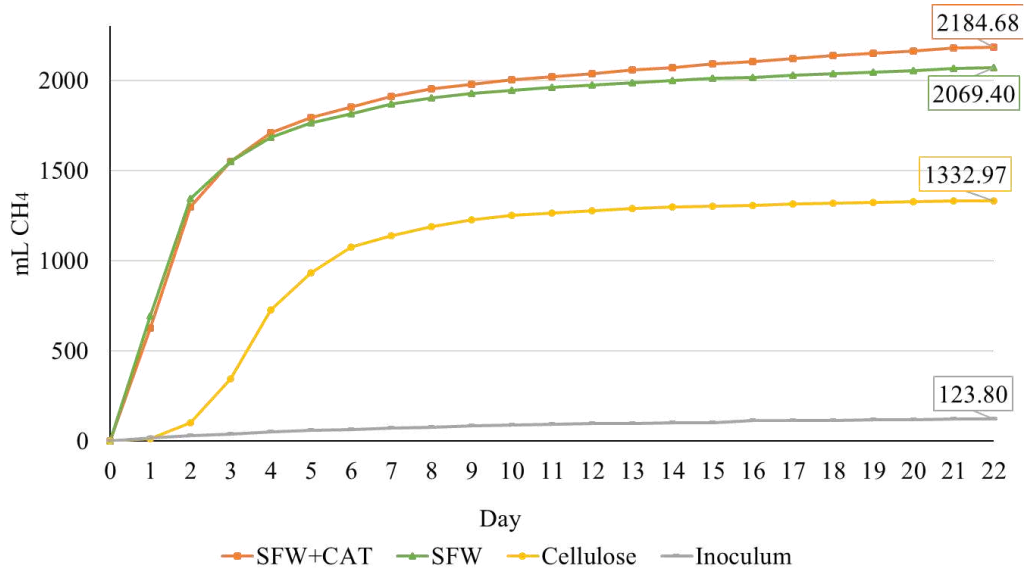


Figure 4.1 - Methane production during 21 days of anaerobic digestion of SFW, SFW+CAT, cellulose and inoculum in terms of ml CH_4.

The specific methane production was 557.96 and 574.83 ml CH_4/g VS for SFW+CAT and SFW respectively. Indeed, anaerobic digestion of food waste can generate from 500 to 600 ml CH_4/g VS (Bong et al., 2018). Used as reference, cellulose reached a final methane yield of 370.27 ml CH_4/g VS (between the value of 345 and 390 ml CH4/g VS), which value ensured compliance with the process (Holliger et al., 2021). The reliability of the process was also confirmed by the final pH values, which were about 7.5.

During the active composting step, the O_2 consumption from the mixture of solid digestate, sawdust, and compost (mixed or not with CAT) is reported in Figure 4.2. O_2 uptake of SFW was almost constant during the first 16 days of the process (except for the first day), showing a slight reduction after that moment. Instead, the SFW+CAT sample, maintained the constancy till the 28th day of the test, also due to the growth of a fungus, which required a higher O_2 consumption. Despite the different trends, the final O_2 uptake achieved by SFW+CA and SFW was 74.79 and 74.51 mg O_2 /g DM, respectively.

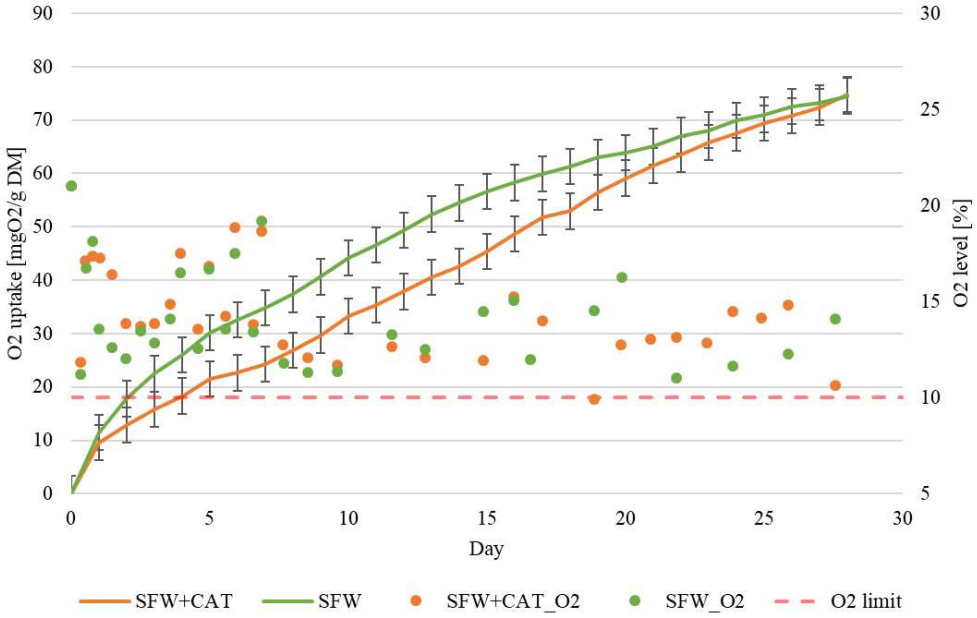


Figure 4.2 - Oxygen uptake and O_2 level of SFW SFW+CAT samples during 28 days of active composting.

After active composting, two stages of curing composting (each of 26 days) were carried out where, an overall VS reduction of 34.3% and 38.0% for SFW+CA and SFW respectively were reached. All the values were higher than 30%, as required by ISO 20200 and comparable with other composting tests in the literature (Cucina et al., 2018).

Concerning the whole composting process, the increase of TS from about 35% (start of active composting) to 75% (end of curing composting) along the whole process revealed a progressive bio-drying of the mixture. At the end of the curing composting stage, the pH level was around 5.5. The acidity was due to the gradual degradation of the solid digestate, leaving only the materials with a lower bio-degradability rate, like the sawdust (characterised by a low pH).

The degree of disintegration was evaluated for CAT samples in each step of the tests and the results are shown in Figure 4.3. Overall, the combined anaerobic digestion and composting process ensured a significant CAT disintegration (73.82 %). The main

contribution of disintegration was achieved during anaerobic digestion, where 66.76 % of CAT samples was fragmented. The further aerobic steps, although carried out for a longer time, did not affect substantially the disintegration value.

From a visual inspection (Fig. 4.3a), at the end of the entire process, CAT was completely fragmented, brittle, and with a reduced thickness than the original one. On the surface, a significant number of cracks and holes was evident. The highest disintegration under anaerobic digestion was possible also because of the high water content during the process. Especially if blended with triacetine, cellulose acetate results susceptible to water degradation (Olaru et al., 2001).

Despite the high degree of disintegration, the final compost quality (Q_c) reached an average value of 1.10%, not in compliance with the legal limits of 0.5% of the Italian legislation (Decreto Legislativo, 2010). This means that the compost could not be re-used in agriculture as fertilizer and should be disposed in landfill as surface cover, accounting the respective costs. On the other hand, a potential phytotoxicity of the compost was averted even if the compost quality is not ensured from the high presence of bio-plastics residues (Gadaleta et al., 2022a).

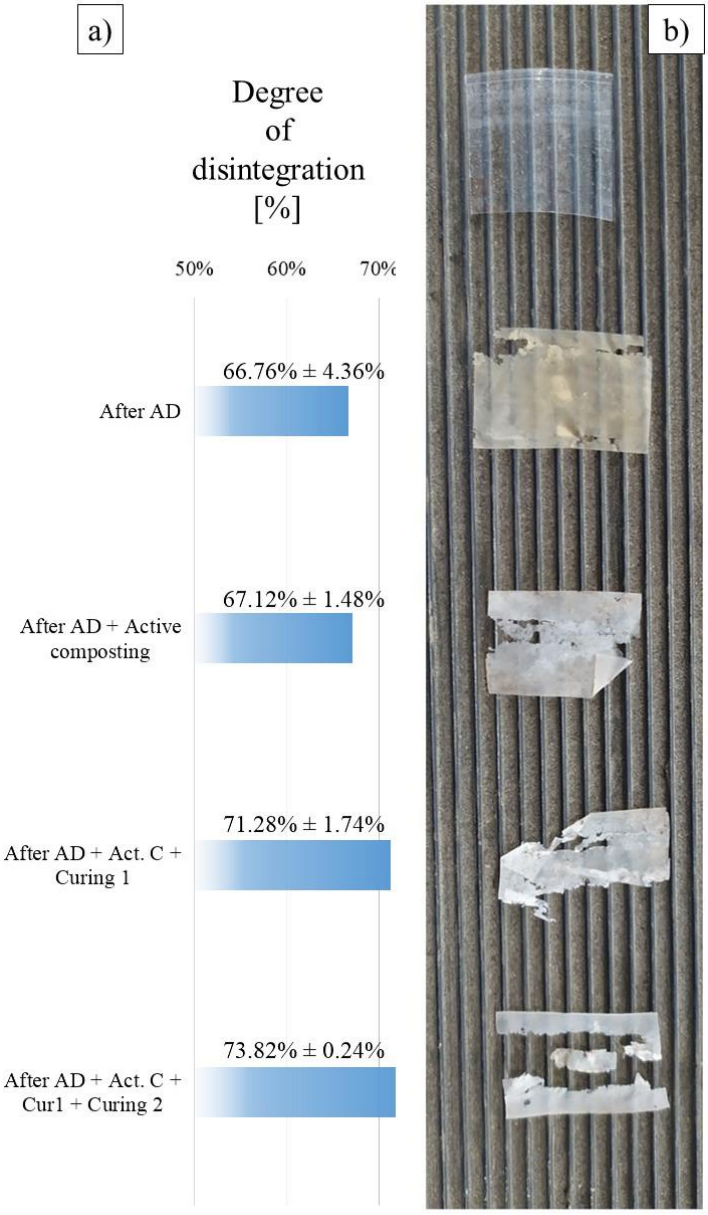


Figure 4.3 - Cumulative value of the degree of disintegration (D) (a) and visual inspection of CAT (b) during each stage of the experiment (from left to right: virgin CAT, end of anaerobic digestion, active composting, first and second curing composting).

Overall, the technical assessment of organic waste treatment route pointed out that the current amount of bio-plastics in the organic fraction of municipal solid waste did not affect the biological treatment processes such as anaerobic digestion and composting. Actually, the surplus of methane from CAT during the anaerobic digestion resulted in slight technical benefit. At the same time, a significant presence of plastics in the compost was observed with an increase in compost impurity.

4.1.2. Plastic waste treatment route

When a stream of mixed plastic waste enters the MRF, it is sent through several sorting stages. Each stage is responsible to positively sort target materials. Sorting analyses provide more insights as to what materials are difficult to sort in the different parts of the MRF process and where contamination may be prevalent (Damgacioglu et al., 2020). This understanding allows to evaluate the contamination of bio-plastics in different homogeneous plastic waste streams. The bio-plastics presence for each material and process are provided in Figure 4.4.

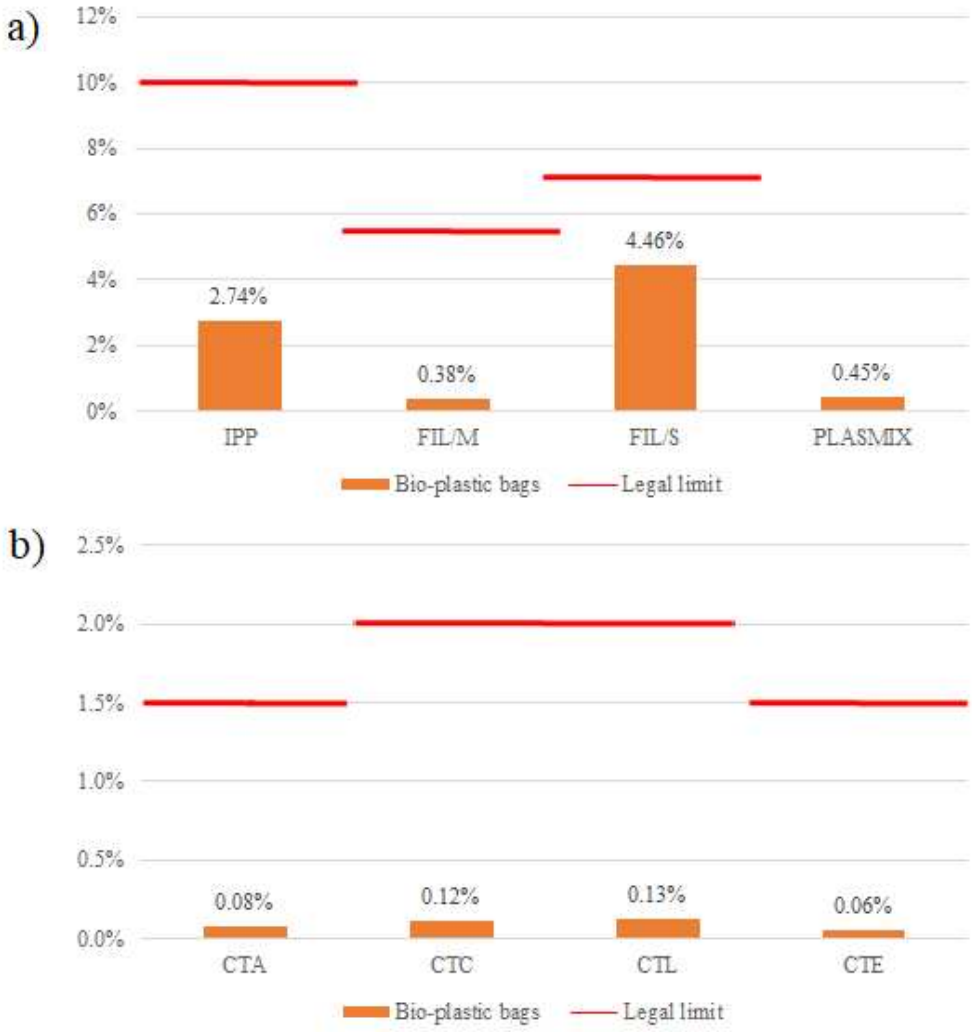


Figure 4.4 - Amount of bio-plastics (%) in PP mixed packaging (IPP), packaging film larger (FIL/M) or smaller (FIL/S) than an A3 sheet, PLASMIX (a) and PET-PE bottles (CTL= light PET; CTA= light-blue PET; CTC= colored PET; CTE= PE) (b) streams detected in the investigated MRF.

More specifically, Figure 4.4a shows the bio-plastics contamination for flexible or mixed plastic waste, instead Figure 4.4b the one for rigid plastic waste of PET and PE.
Concerning flexible plastic waste, bio-plastics are mainly found in LDPE packaging films smaller than an A3 sheet (FIL/S) and mixed PP packaging(IPP), counting 4.46% and 2.74% of these streams respectively (Figure 4.4a). In non-recyclable plastic waste

mix (PLASMIX) and LDPE packaging films larger than an A3 sheet (FIL/M) the presence accounts barely 1%. In the rigid plastics streams, bio-plastics are also less than 1%. In all these streams, bio-plastics are definitively lower than the technical specification limit (red line). The negligible presence of bio-plastics in the rigid plastic waste streams is due to the positive sorting process performed. The differences in shape and polymer type reduced the presence of bio-plastics in these streams, avoiding any possible contamination (as shown by the technical specification limit – red line in Figure 4.4b). On the other hand, the sorting process through ballistic screen does not allow to separate conventional plastic films from bio-plastics ones, which remain in FIL/S streams. It is important to note that, the majority of Italian MRF does not provide a recycling of FIL/S streams, which are sent to incineration with PLASMIX streams (Gadaleta et al., 2020).

Then, in the MRF process, CAT (represented as the current amount of general bio-plastics waste) would not affect the sorting process since the amount of bio-plastics bags are significantly lower than the technical specification limit but also because FIL/S is not further recycled but used in incineration with PLASMIX for energy recovery.

On the other hand, since the LDPE streams is the one mostly subjected to a potential CAT contamination, the further mechanical recycling of LDPE by varying the CAT content was investigated.

TGA analysis of neat LDPE and LDPE-CAT blends extruded samples are shown in Figure 4.5.

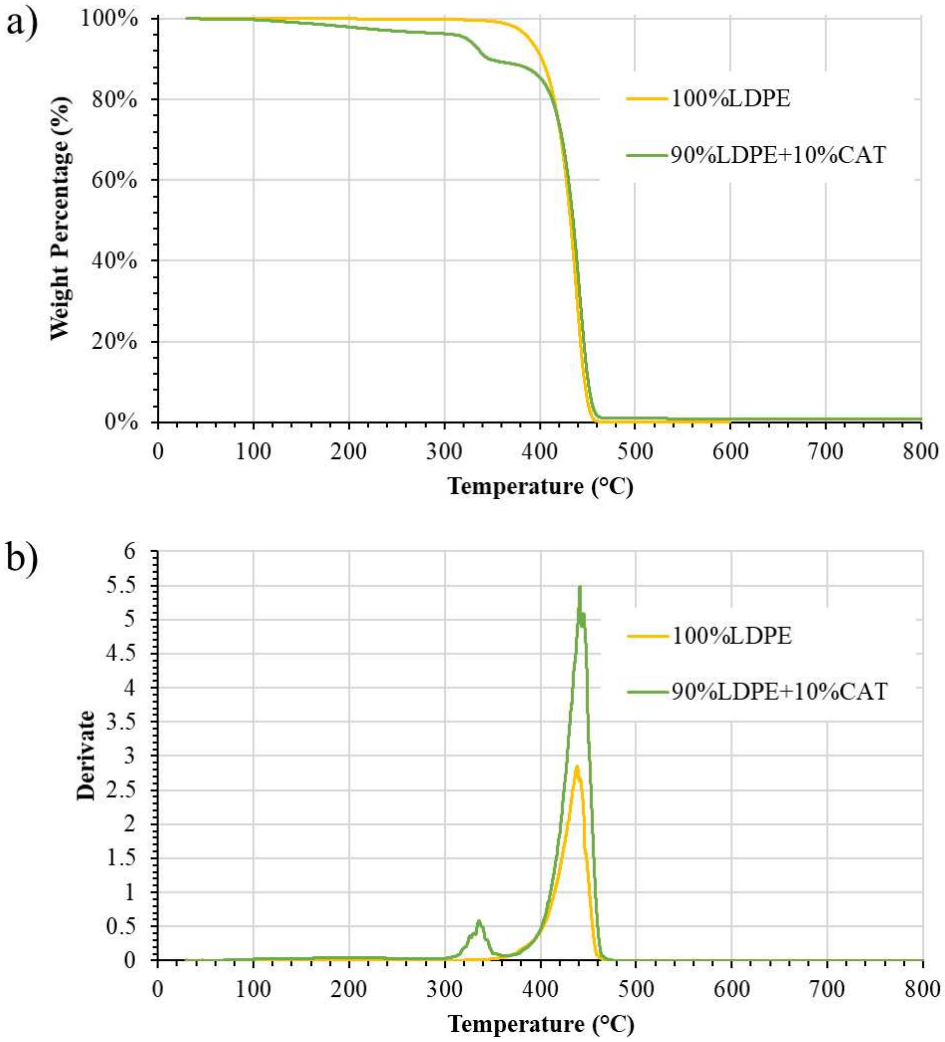


Figure 4.5 - TGA thermograms (a) and its derivate (b) for LDPE-CAT blends.

Neat LDPE undergoes single stage degradation in the range of 370-480°C, achieving the maximum degradation at 440°C, as reported in scientific literature (Chaudhary et al., 2021). The LDPE-CAT blends were characterized by a two-stage degradation at 340°C and 440°C (Fig. 4.5a). The first peak is due to the degradation of CAT, which has a lower degradation temperature than LDPE (Cindradewi et al., 2021), while the second one remained the main LDPE degradation peak. Similar trends were already

reported in Sailaja and Seetharamu (2009). TGA do not suggest a significant difference in degradation between neat LDPE and LDPE-CAT blend (10%), especially from the fact that the maximum CAT blend was analyzed.
The DSC plots for the different LDPE-CAT blends are shown in Figure 4.6.

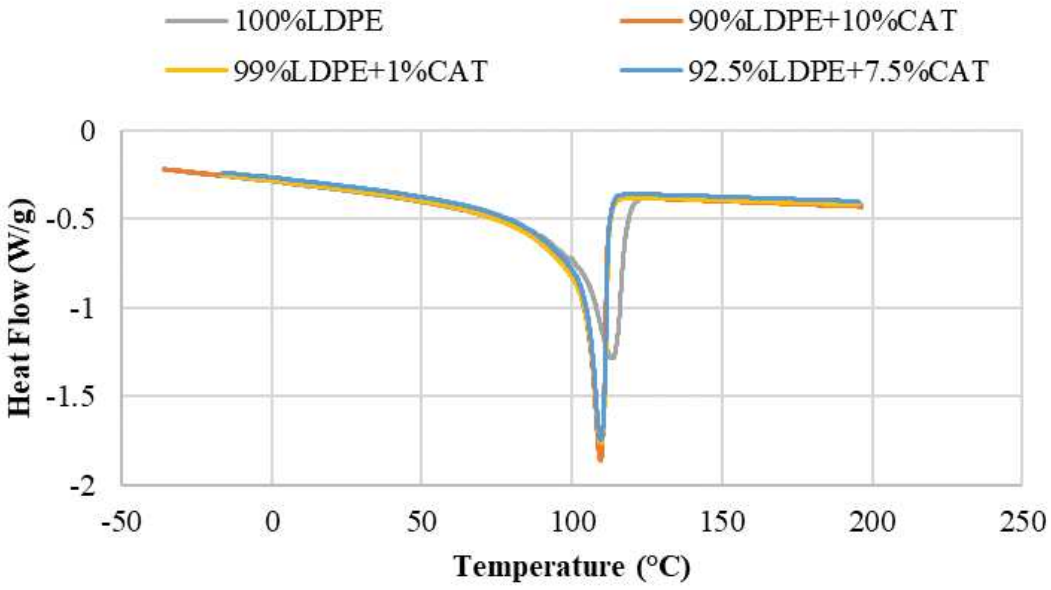


Figure 4.6 - DSC thermograms for LDPE/CAT blends in the second cycle of heating.

The peak temperature of neat LDPE revealed a melting point of 116°C. The addition of CAT in the LDPE matrix decreased the melting point to 109°C, independently from the amount of CAT in the blend. Indeed, all the peaks of LDPE-CAT blends achieved the same heat flow value. The shifting of melting point was followed by an increase in the enthalpy of fusion since the area under the curves was grown. The overall similarity of neat LDPE and LDPE-CAT blends, except from the slight decrease of melting point, was confirmed by Kosaka et al. (2006).
Structural changes in LDPE extruded samples before and after each CAT blending were analysed by using FTIR (Figure 4.7).

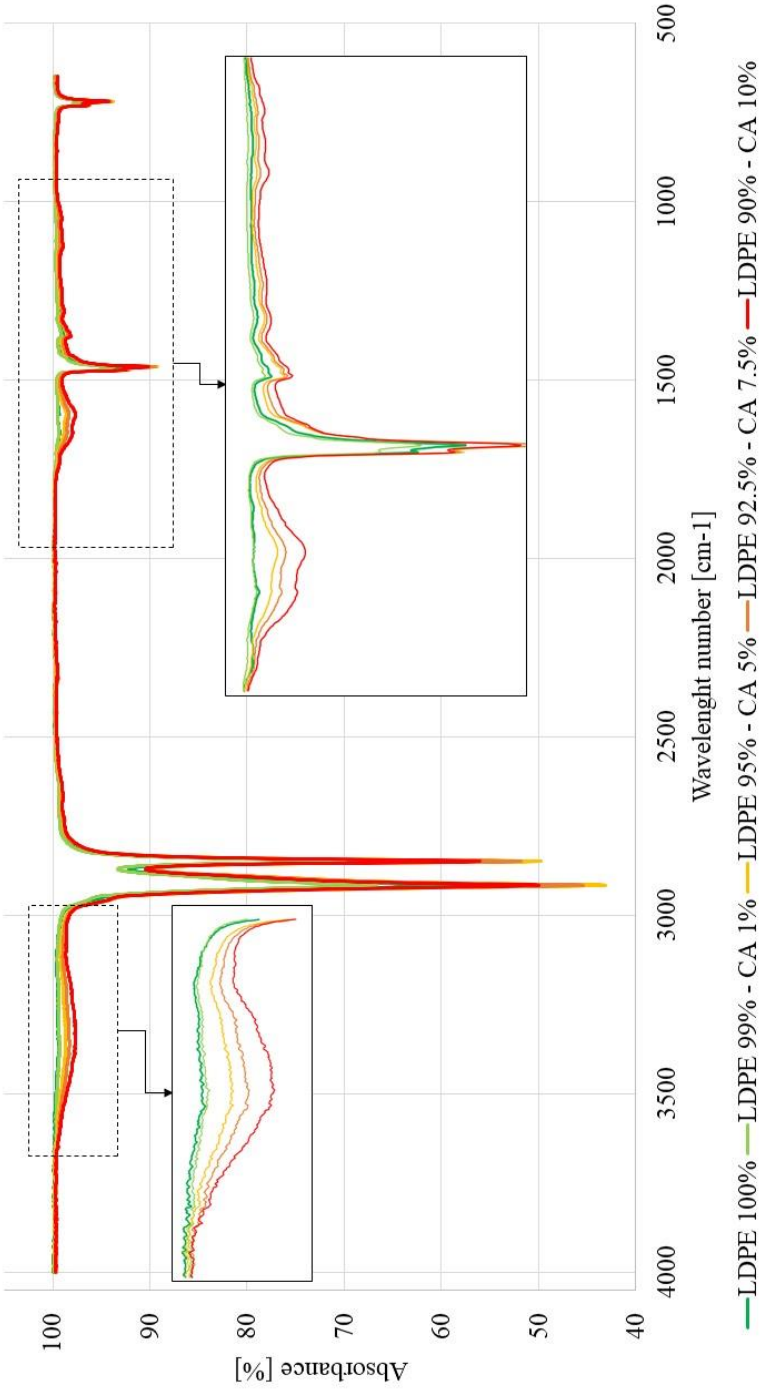


Figure 4.7 - FTIR spectra for LDPE/CAT blends.

In the neat LDPE sample, significant characteristic peaks at wave numbers of 2915 cm^{-1}, 2850 cm^{-1}, 1460 cm^{-1} and 720 cm^{-1} are visible. These peaks correspond to CH_2 asymmetric stretching, CH_2 symmetric stretching, bending deformation and rocking deformation respectively (Chaudhary et al., 2021). The addition of CAT in different proportions does not significantly alter the shape of the FTIR spectrum. However, two weak peaks centered at 3340 cm^{-1} and 1595 cm^{-1} appear. The first peak is attributed to the O-H bond stretching of CAT, triacetine or water absorbed. The second is not related to the CAT since no characteristic peaks are associated to that wavenumber. Therefore, other phenomena are involved, such the excitation of carbonyl compounds know as Norrish reactions of type I, II and III (Olajire and Mohammed, 2019). In addition, the area between 1465-1000 cm^{-1} was shifted to lower absorbance, mainly when the CAT was more abundant, since this range is the one where CAT has its characteristic peaks.

Finally, a visual inspection of the extruded samples was carried out in order to investigate the changes in processability of LDPE with the variation of CAT. Images of the different LDPE-CAT blends are shown in Figure 4.8.


b)


c)

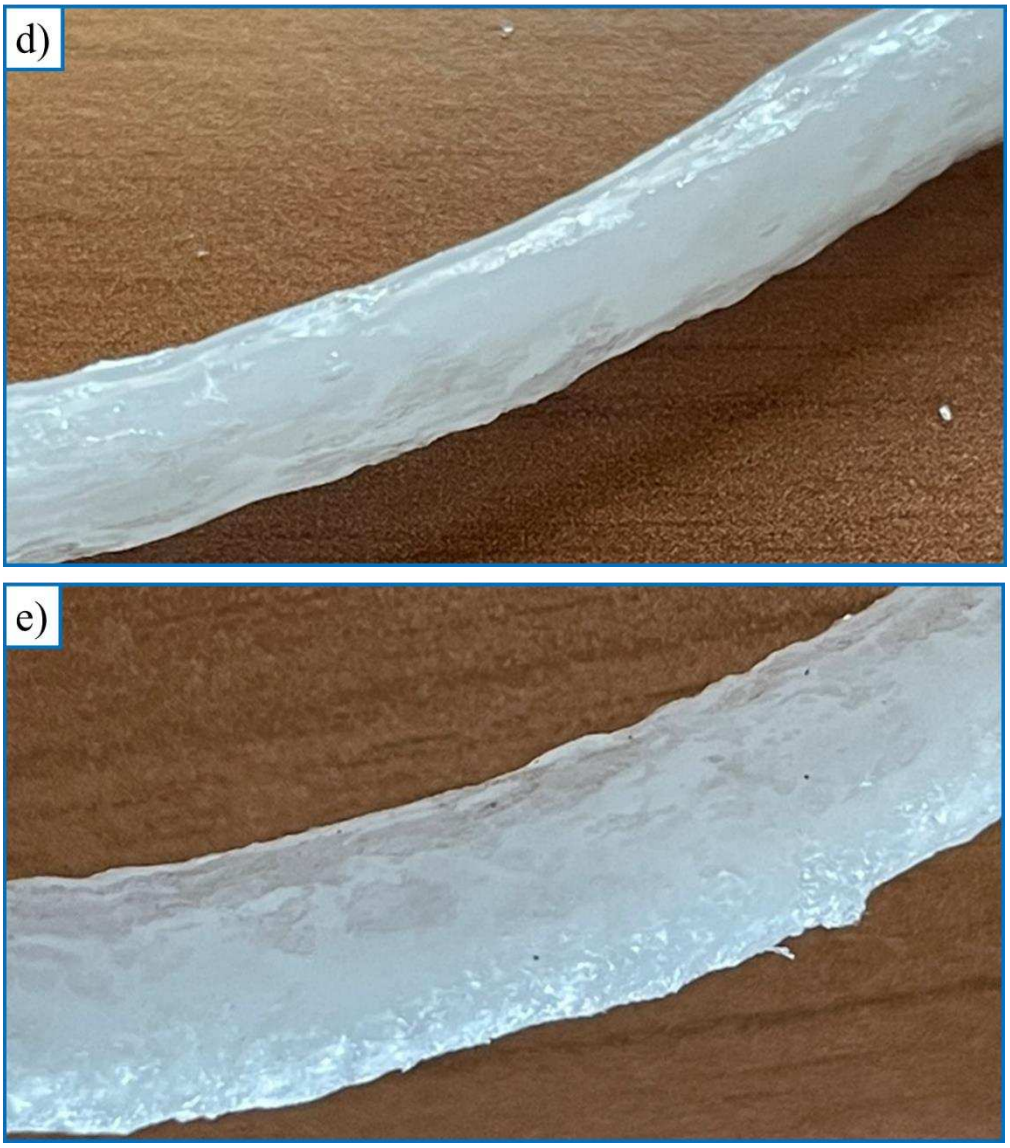
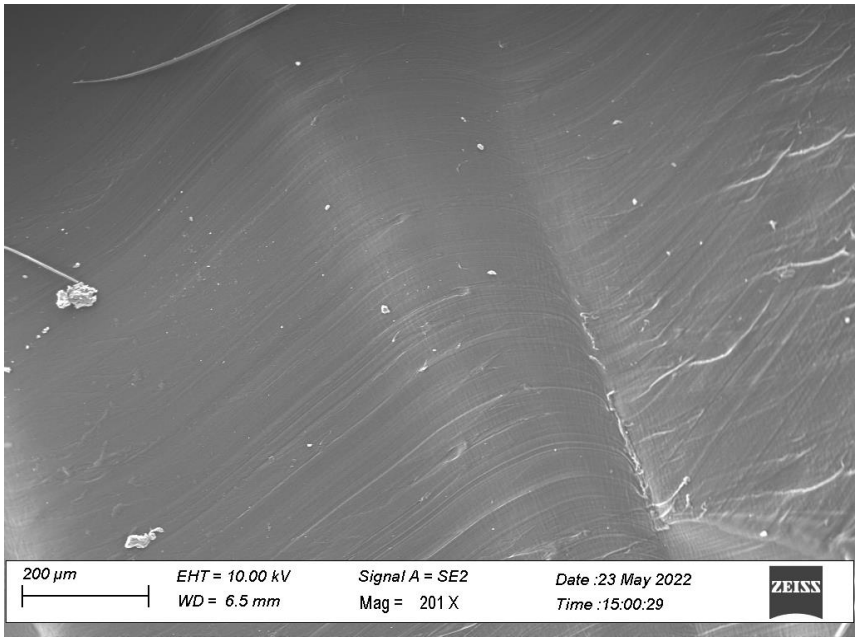


Figure 4.8 - Visual inspection of extruded samples by varying the CAT content to (a) 0%, (b) 1%, (c) 5%, (d) 7.5% and (e) 10%.

Initially, the neat LDPE was smooth and clear without any significant irregularities (Fig. 4.8a). The addition of CAT induced surface roughness and exfoliation, becoming more evident with the rise of CAT content (Fig. 4.8b – 4.8e). In addition, a focus of the section of the samples was carried out through SEM images (Figure 4.9).

a)



b)

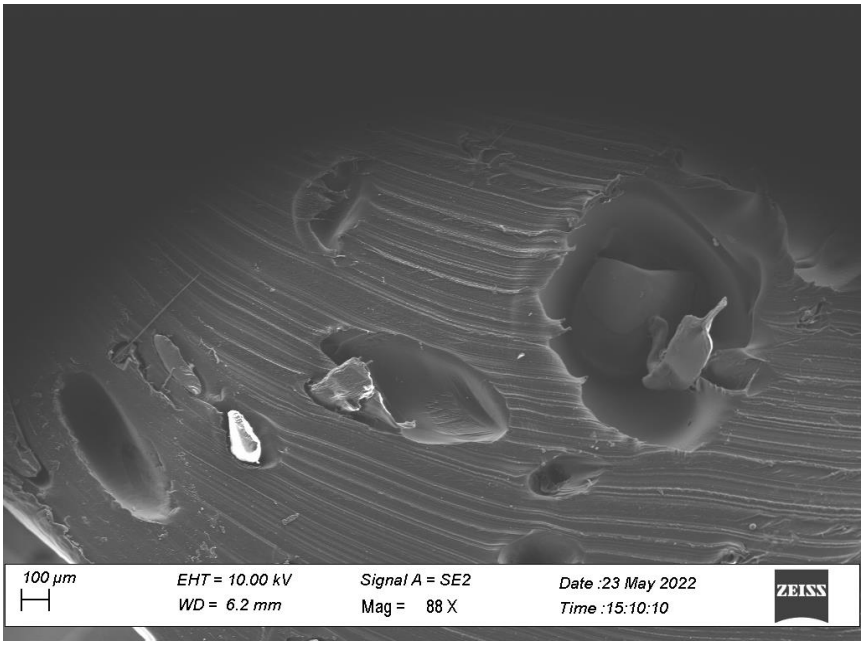


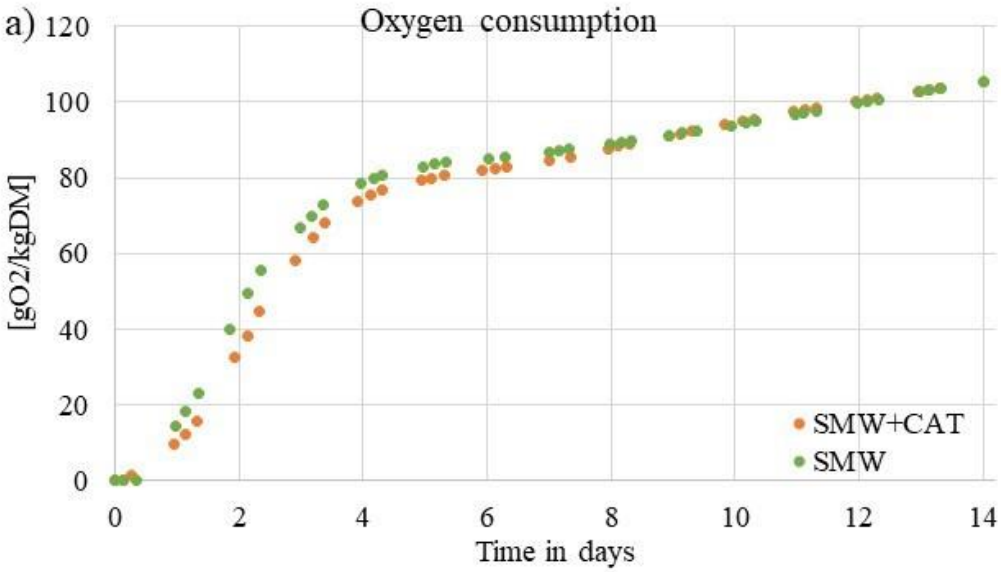
Figure 4.9 – SEM images of (a) neat LDPE and (b) LDPE blended with 10% of CAT.

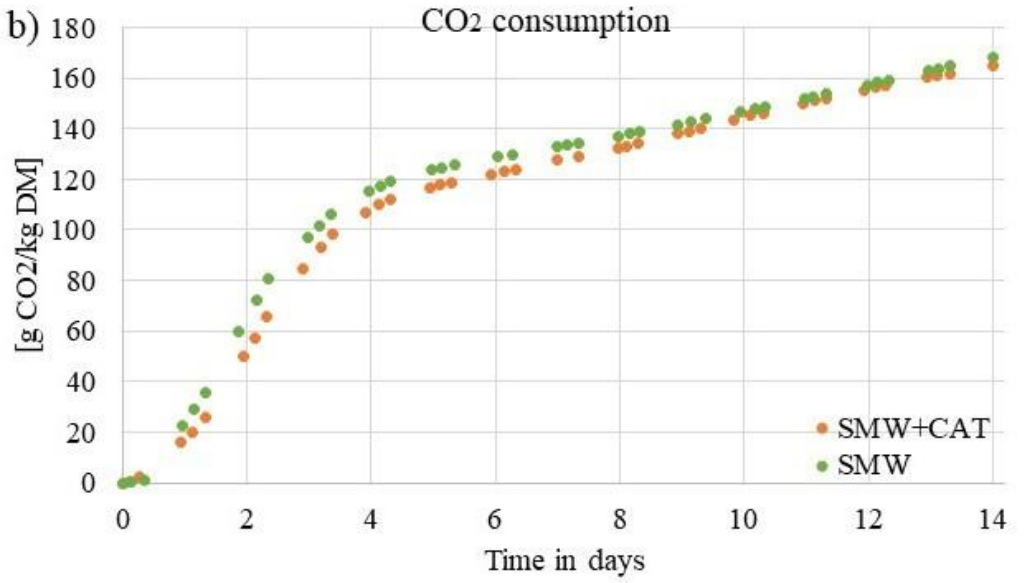
The section of LDPE-CAT blends contained cavities and holes (Fig 4.9b) than the one of neat LDPE (Fig. 4.9a), which appeared smooth. These cavities could be found also in a magnitude two times smaller than the one of neat LDPE.

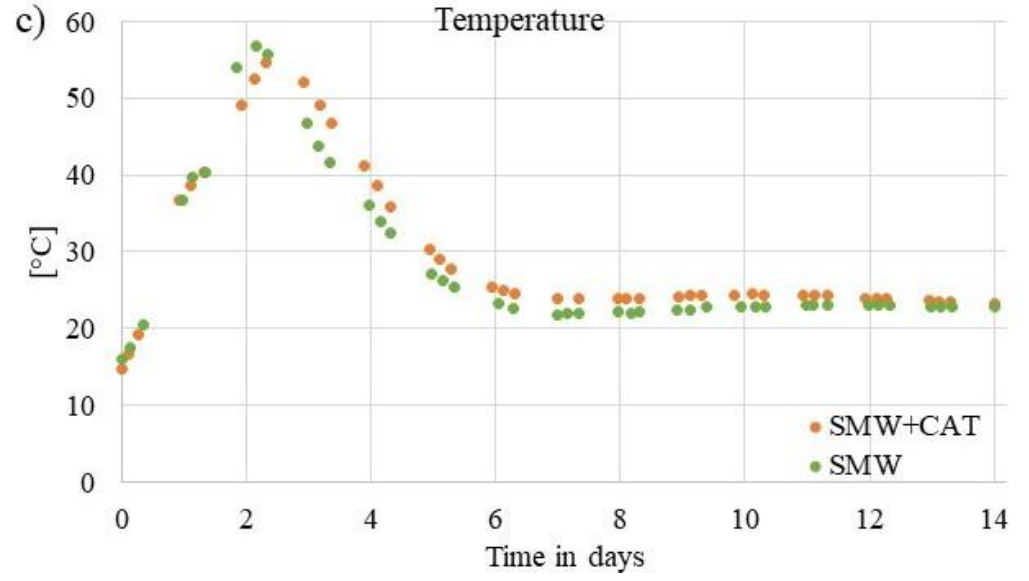
Overall, despite the thermogravimetric and FTIR analysis did not show significant difference of the LDPE-CAT blends, the inspection of extruded samples suggested a reduction in LDPE recyclability, also at low CAT content. These findings resulted in a critical output contamination, since the addition of CAT would strongly affect the LDPE recycling.

4.1.3. Mixed waste treatment route

The development of a lab-scale biostabilization for treating mixed MSW with and without CAT is very significant in the field of waste management system since it opens to a possible future management of bio-plastics. Process parameters of SMW+CAT and SMW samples are shown in Figure 4.10.




b)
CO2 consumption
[g CO2/kg DM]
180
160
140
120
100
80
60
40
20
0
0
2
4
6
8
10
12
14
Time in days
SMW+CAT
SMW


c)
Temperature
[°C]
60
50
40
30
20
10
0
0
2
4
6
8
10
12
14
Time in days
SMW+CAT
SMW

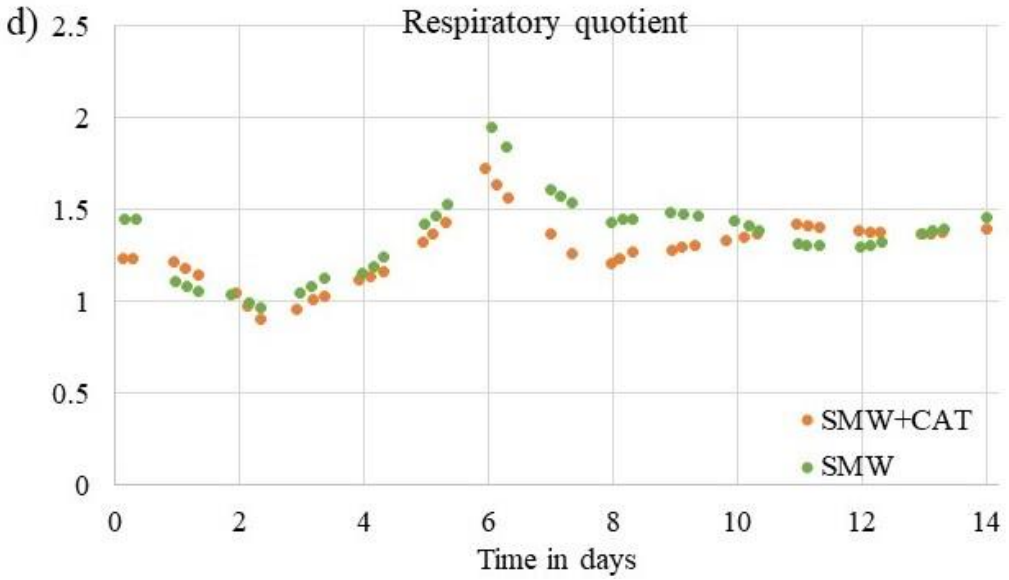


Figure 4.10 - (a) O_2 consumption, (b) CO_2 production, (c) temperature trend and (d) Respiratory Quotient of SMW+CAT and SMW during 14-days of biostabilization.

The biostabilization processes inside the reactors can be explained by studying the O_2 consumption as well as the CO_2 production. During the aerobic biostabilization, both the samples consumed comparable amounts of oxygen (Figure 4.10a): SMW+CAT and SMW achieved an oxygen consumption of 105.38 and 105.47 g O_2/kg DM respectively. The main consumption occurred in the first 4 days, where a daily O2 consumption of 16 g O_2/kg DM*d was maintained. Then, the biostabilization continued slower, with a rate of about 3 g O_2/kg DM*d. A similar trend could be noticed also for the CO_2 production (Figure 4.10b): SMW+CAT and SMW produced 164.94 and 168.32 g CO_2/kg DM respectively. The maximum rate of 24 g CO_2/kg DM*d was reached in the first 4 days, remaining constant (5.7 g CO_2/kg DM*d) till the end of the experiment. The O_2 consumption and CO_2 production trends resulted comparable and in line with the one in literature (Slezak et al., 2015; van Praagh et al., 2009).

Starting from an average temperature of 16.3°C, SMW+CAT and SMW samples reached an average peak of 55°C at day 2. Then, the temperature decreased until 22°C, remaining constant for the last 8 days (Figure 4.10c). As reported in (Pecorini et al.,

2020), a similar value (59°C) was reached by a fresh compost, resulting higher than the one obtained by the SMW biostabilization for the greater presence of putrescible fraction than the SMW adopted in this assessment. This high temperature heating ensured that the resulting product is free from pathogens, since one hour heating in the range of 50–55°C would kill most of the common parasites and pathogens (Tom et al., 2016).

The trend of RQ is visible in Figure 4.10d. RQ started from 1.35, decreasing to 0.99 at day 2 (where the maximum O_2 consumption and CO_2 production rates were achieved) and increasing again to 1.75 until day 6. Then, it started to reach a constancy around the value of 1.42 from day 8, which showed the end of the main biostabilization process and the beginning of a curing phase. The preservation of the aerobic environment during the experiment was ensured by the RQ value, which never exceed the aerobic limit of 1.

At the end of 14 days of biostabilization, the CAT specimens reduced their weight 22.57%, mainly due to the melting of triacetine (used as plasticizer). Even if a not negligible weight loss was achieved at the end of the process, no signs of degradations (cracking, holes etc.) was found. Indeed, the specimens maintained their shape, resulting in a slight yellowness of the surface due to the high degree of substitution of cellulose acetates, which lead to aerobic degradation (Leppänen et al., 2020). Even if the degradation of CAT specimens is not ensured after biostabilization, the output is not contaminated by their presence (as occurred at the end of composting). Hence, this opens to the use of processed CAT in incineration as waste-to-energy process. The average HHV from triplicates of non-processed and 14-days biostabilite SMW+CAT, SMW and CAT specimens are presented in Figure 4.11.

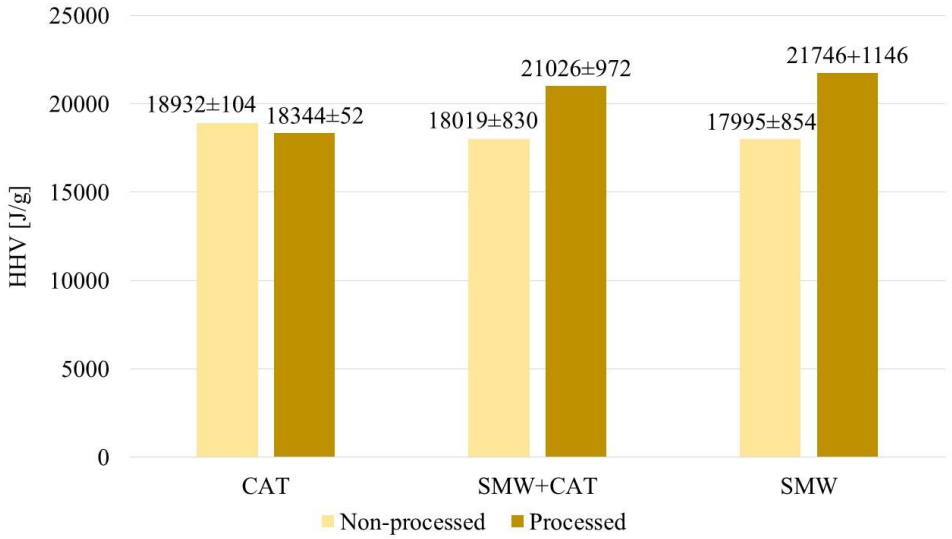


Figure 4.11 - HHV of CAT specimens, SMW+CAT and SMW samples before and after the 14-days biostabilization.

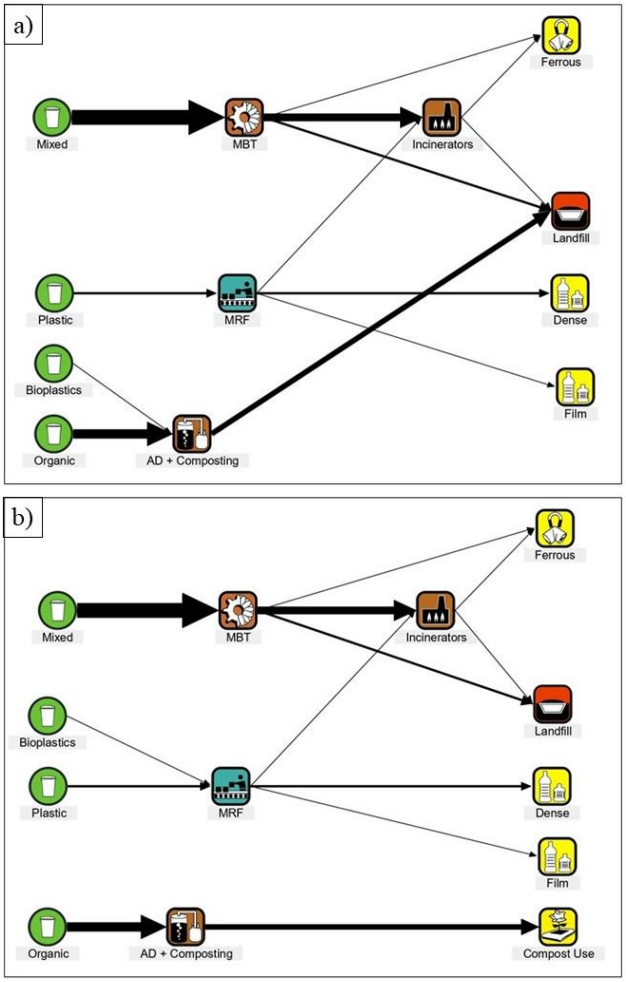
Focusing on CAT specimens, fresh and processed untreated specimens were characterized by an HHV of 18,932 and 18,344 J/g respectively. The HHV decreased of 3.1% compared to the fresh one, since the degradation of cellulose acetate is very slow in aerobic environment (Phuong et al., 2014).

Concerning the sample of waste and bio-plastics (SMW+CAT and SMW), fresh samples achieved a HHV of 18,019 and 17,995 J/g for SMW+CAT and SMW respectively. After the biostabilization, the reduction of organic fraction in the mixture increased the HHV of processed SMW+CAT and SMW of 17.1% (21,026 J/g) and 20.8% (21,746 J/g) respectively than the one of fresh samples. The obtained results were in line with other heating value of fresh and processed mixed MSW, which can range from 12,000 to 27,600 J/g (Amen et al., 2021; Dashti et al., 2021).

From a technical point of view, the analysis of the biostabilization parameters confirms that the addition of CAT is not a limiting factor on the aerobic biostabilization of mixed MSW. In addition, the treatment of CAT with MSW did not influence also the final HHV. On the other hand, processed CAT maintained a high HHV and, which can generate an additional slight amount of energy if treated in incineration for energy recovery.

4.2. Economic assessment

From the technical assessment, it was possible to determine the mass streams of the whole waste management system for each waste treatment route (Figure 4.12).



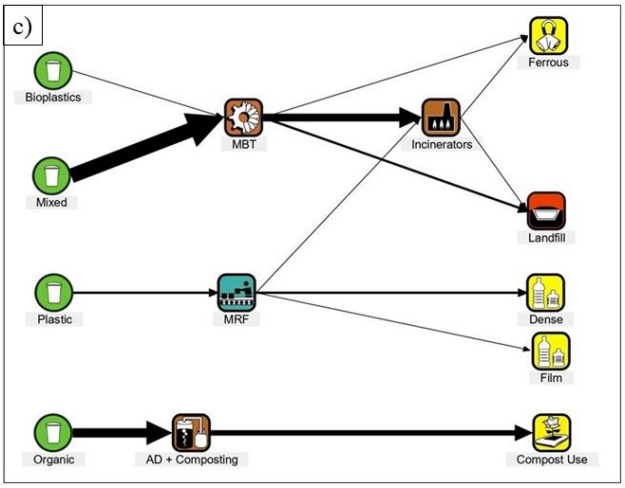


Figure 4.12 - Mass balances of organic (a), plastic (b) and mixed (c) waste treatment route.

It is possible to note that in the organic waste treatment route (Fig. 4.12a) the CAT disintegration (equal to 73.82%, as reported in the technical assessment section) did not ensure a suitable compost quality. For this reason, it was assumed that the compost would be landfilled, and used as surface cover. Instead, in the plastic and mixed waste treatment route (Fig. 4.12b and 4.12c), the use of pure organic waste in the biological process ensured to achieve a suitable quality for compost recycling, since the CAT was collected with other waste streams.

From a quantitative point of view, the mass and energy flows involved are summarized in Table 4.1.

In the organic waste treatment route, the conversion of CAT during the anaerobic digestion stage increased the amount of methane of about 18% compared to the one generated in the other two scenarios. Due to the disposal in landfill of the produced compost, the waste management system showed the highest amount of waste disposed, whereas the incineration generated the lowest amount of ashes, compared to the other two treatment routes.

In the plastic waste treatment route, the input of MRF increased for the addition of CAT. This led to a higher amount of PLASMIX, in line with the technical assessment findings and with the absence of FIL/S sorting in mostly of Italian MRF. Compared to organic

route, the high amount of PLASMIX input to incineration increased the energy recovered during this stage, generating a 7.2 kg of ashes. The use of high-quality compost in agriculture as fertilizer resulted in a significant reduction (about 28%) of waste landfilled: from 333.50 kg in the organic scenario to about 240 kg in the other ones.
Finally, the mixed waste treatment route showed a rise in RDF produced during the MBT. The slight degradation (22.57% as resulted in the technical assessment) occurring during this stage reduced the amount of CAT in the incineration process. Since the HHV was comparable to the non-processed one, the final amount of the produced energy during this step was comparable to the one obtained in the plastic waste treatment route. However, compost recovery maintained the amount of waste landfilled below 300 kg. Importantly, this system generated the least amount of energy compared to the previous two scenarios.

Table 4.1 - Mass and energy balances of organic, plastic and mixed waste treatment routes for every treatment.

Treatment	Stream	Organic WTR	Plastic WTR	Mixed WTR
AD+composting	Input [kg]	372.66	353.82	353.82
	Compost [kg]	98.71	93.78	93.78
	% BPW in compost [%]	5.00	0.00	0.00
	Biogas produced [m3CH4]	54.87	46.61	46.61
	Electric energy [kWh]	187.78	159.51	159.51
	Thermal energy [kWh]	268.25	222.87	222.87
MRF	Input [kg]	79.77	98.61	79.77
	Recycled [kg]	37.77	37.77	37.77
	PLASMIX [kg]	42.07	60.91	42.07
MBT	Input [kg]	547.57	547.57	566.41
	RDF [kg]	178.51	178.51	193.10
	SOF [kg]	233.10	233.10	233.10
Incineration	Input [kg]	220.58	239.42	235.17
	Electric energy [kWh]	224.45	239.31	235.59
	Thermal energy [kWh]	555.13	591.90	582.69
	Ashes [kg]	6.62	7.18	7.06
Tot.	Waste landfilled [kg]	333.50	240.28	240.16
	Electric energy [kWh]	412.23	398.82	395.09
	Thermal energy [kWh]	823.39	819.77	810.55

On the basis of the mass balance, Table 4.2 provides the expenses, revenues and total costs of the waste management system for each treatment route. First, the combined anaerobic digestion and composting process achieved the highest cost in each scenario, accounting between 49.4% and 44.1% of the total expenses. Most of them are achieved by the transportation (around 27%), while the whole treatment did not exceed 17%. Instead, landfilling costs for biological process were present in the organic waste treatment route only, since the low-quality of compost required its disposal. Due to its great amount of waste processed, the second process with the highest expenses was the mixed waste one, composed by the MBT and incineration. However, transportation revealed a strong influence on expenses, being between 17.3% to 19.1% but lower than biological treatment. Concerning the treatment process, the MBT achieved higher expenses (17.14-17.73 €) than the incineration one (6.73-7.31 €). Due to the high amount of Stabilized Organic Fraction (SOF) and of ashes after incineration, the cost of landfill was up to 28.83 €. Plastic sorting and recycling processes achieved a relatively low cost in the waste management system: 38.23 (12.3%) and 30. 93 € (15.9%), respectively. The reason could be not only the the lower amount of waste in input but also the well-established sorting process. Indeed, the existence of subsidies through the extended producer responsibility fees, which producers pay when their (plastic) packaging enters the market, allow to cover sorting and recycling costs. On the other hand, the MRF process revealed a cost comparable that of the MBT, although the amount of waste processed was nearly 15% of that treated by MBT. Overall, can be seen that the collection of CAT in each waste stream has obviously increased the expenses of the specific treatment process.

The highest revenues were obtained by the organic waste treatment route, since more energy was achieved during anaerobic digestion (the process with the highest efficiency). Comparing anaerobic digestion and incineration as source of revenues, the latter resulted higher in each scenario. Although the efficiency of energy production is higher in the treatment of organic waste during biogas combustion, the highest waste input into the mixed waste system allowed for more energy and thus more of revenues.

In fact, more than 90% of revenues were from the sale of energy; the remaining part came from the sorting of plastic waste.

Finally, all the scenarios achieved a positive cost, revealing how the expenses calculated exceeded the revenues. It is important to note that revenues from the tariff paid by users of the waste management system were not considered, as this assessment focused only on treatment. The CAT treatment cost revealed how the highest one was achieved by the organic scenario (120.35 €), followed by the plastic (112.21 €) and mixed one (109.43 €).

Table 4.2 - Waste management system expenses, revenues and costs of organic, plastic and mixed waste treatment routes values and contribution (%).

	Organic WTR		**Plastic WTR**		**Mixed WTR**	
	€	**%**	**€**	**%**	**€**	**%**
EXPENSES	**250.93**		**241.36**		**236.01**	
Organic	**123.92**	**49.4%**	**106.41**	**44.1%**	**106.41**	**45.1%**
Treatment	43.97	17.5%	41.75	17.3%	41.75	17.7%
Landfill	11.85	4.7%	0.00	0.0%	0.00	0.0%
Transportation	68.10	27.2%	64.66	26.8%	64.66	27.4%
Plastic	**30.93**	**12.3%**	**38.23**	**15.9%**	**30.93**	**13.1%**
Fines	0.34	0.1%	0.42	0.2%	0.34	0.1%
Treatment	12.69	5.1%	15.68	6.5%	12.69	5.4%
Transportation	17.90	7.1%	22.13	9.2%	17.90	7.6%
Mixed	**96.08**	**38.3%**	**96.72**	**40.0%**	**98.67**	**41.8%**
Treatment (MBT)	17.14	6.8%	17.14	7.1%	17.73	7.5%
Treatment (Inc.)	6.73	2.7%	7.31	3.0%	7.18	3.0%
Landfill	28.77	11.5%	28.83	11.9%	28.82	12.2%
Transportation	43.44	17.3%	43.44	18.0%	44.94	19.1%
REVENUES	**130.58**		**129.15**		**126.58**	
Organic	**49.63**	**38.0%**	**42.16**	**32.6%**	**42.16**	**33.3%**
Energy	49.63	38.0%	42.16	32.6%	42.16	33.3%
Plastic	**11.08**	**8.5%**	**12.49**	**9.7%**	**11.08**	**8.7%**
Recycling	7.92	6.1%	7.92	6.1%	7.92	6.3%
PLASMIX	3.16	2.4%	4.57	3.6%	3.16	2.5%
Mixed	**69.87**	**53.5%**	**74.50**	**57.7%**	**73.34**	**58.0%**
Energy	69.87	53.5%	74.50	57.7%	73.34	58.0%
Total Cost	***120.35***		***112.21***		***109.43***	

The contribution of each process in the calculation of the total cost is shown in Figure 4.13. Except for landfilling, all the process achieved a similar contribution. The overall treatment (anaerobic digestion and composting, MBT, MRF and incineration) was slightly over 20 %. These values indicate how focusing on one treatment alone would have led to an incorrect result. Waste transport accounted for the highest contribution in the economic evaluation, as this stage has a great influence in the waste management system (Gadaleta et al., 2021b). Regarding energy generation, the results revealed that the collection of CAT in different waste streams did not significantly vary the system. The same considerations can be highlighted for sorting revenues obtained in MRFs. The significant difference was found in the landfill. Considering that the amount of ash at the end of incineration is very similar in all scenarios, the key difference is based on the use of compost: the incomplete degradation of CAT reduces the quality of compost which, as a consequence, has to be disposed in the landfill. Thus, the contribution of landfill in the organic waste treatment route increased from about 7% to 11%. Although CAT showed no significant effect on each waste treatment, their presence may strongly influence the quality of the outputs and thus their further use.

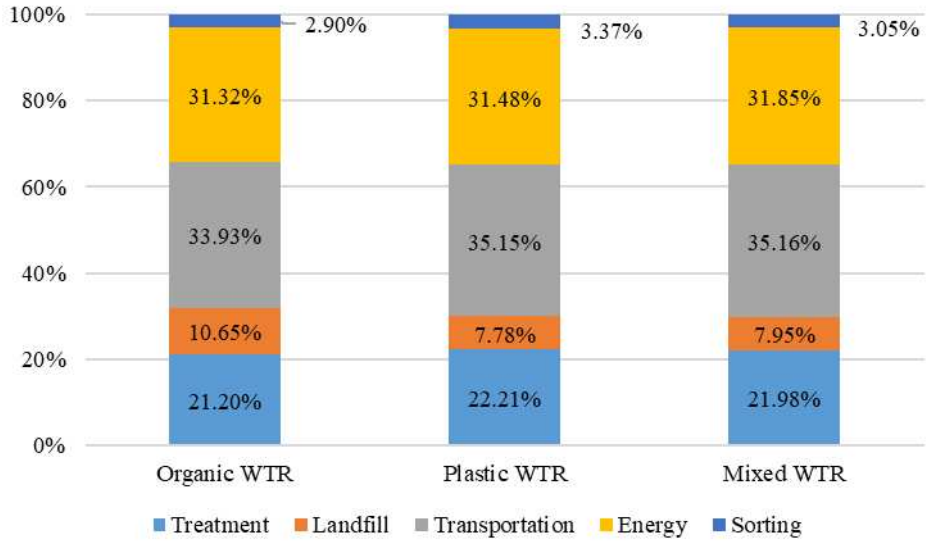


Figure 4.13 - Contribution of each process in the economic assessment of organic, plastic and mixed waste treatment routes.

4.3. Environmental assessment

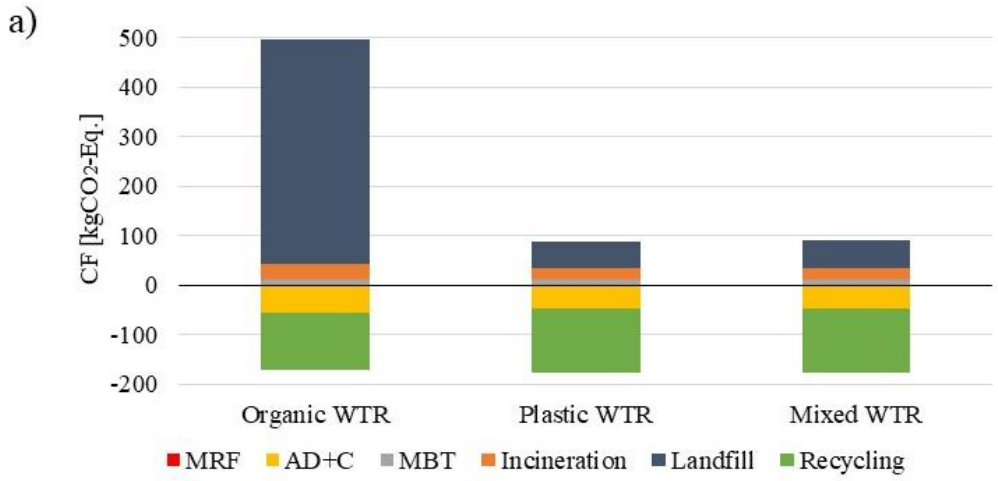
The Carbon Footprint (CF) in terms of $kgCO_2$-eq. was used in order to quantify the environmental impact of each scenario: CF results are shown in Table 4.3.

Table 4.3 - Carbon Footprint (CF) in terms of $kgCO_2$-eq. Of the whole waste management and each treatment for organic, plastic, and mixed waste treatment routes.

Treatment	Organic WTR	Plastic WTR	Mixed WTR
MRF	1.24	1.53	1.24
AD + composting	-54.60	-47.20	-47.20
MBT	11.40	11.40	11.80
Incineration	30.60	21.80	22.00
Landfill	454.00	54.3.00	56.00
Recycling	-118.00	-131.00	-131.00
Total	*324.64*	*-89.17*	*-87.16*

Organic waste treatment route achieved the highest CF (324.64 $kgCO_2$-eq.) among the three scenarios, followed by mixed and plastic waste treatment routes (-87.16 and -89.17 $kgCO_2$-Eq. respectively). The first main finding is that only two scenarios (plastic and mixed) showed negative values, revealing how these systems generated an environmental benefit (Figure 4.14a). Instead, the organic one achieved positive CF, which means an environmental burden of the system (Goulart Coelho and Lange, 2018). With this concept, it is easy to note that the environmental credit in every scenario was given from the combined anaerobic digestion and composting process and from the recycling of waste (plastics, compost etc.). In fact, energy production during anaerobic digestion and reuse of materials instead of virgin ones have reduced the GHG emissions in the systems. On the other hand, MRF, MBT, incineration and especially landfill achieved positive CF value. MRF and MBT are processes that require energy and generate GHG. In each of them, the introduction of CAT in plastic and mixed waste increased the CF values of about 0.3 $kgCO_2$-Eq. Processing through MRF had a

negligible effect from an environmental point of view, since the contribution in the CF calculation was lower than 1% (Figure 4.14b). Also, incineration resulted in a positive value: the highest impact was the one obtained in the Organic waste treatment route, where the energy production was the lowest. Indeed, despite incineration allowed to recover energy, it is known as this process is a source of GHG as CO_2, CO and N_2O (Yang et al., 2012). Finally, the highest environmental burden was achieved in every scenario by the landfill. If in plastic and mixed waste treatment route the CF value was 54.3 and 56 $kgCO_2$-Eq., the value was almost 10 times higher (454 $kgCO_2$-Eq.) in the organic route. Indeed, the highest amount of waste landfilled, composed mainly by organic materials as compost, increased the GHG emission produced during this stage. The degradation of compost in semi-aerobic environment of the landfill generated also CH_4, with a GWP 21–23 times higher than CO_2 (Sohoo et al., 2021a). This amount was not recovered by the plant and increased the CF value, resulting in 67.78% of the total CF value (Figure 4.14b).



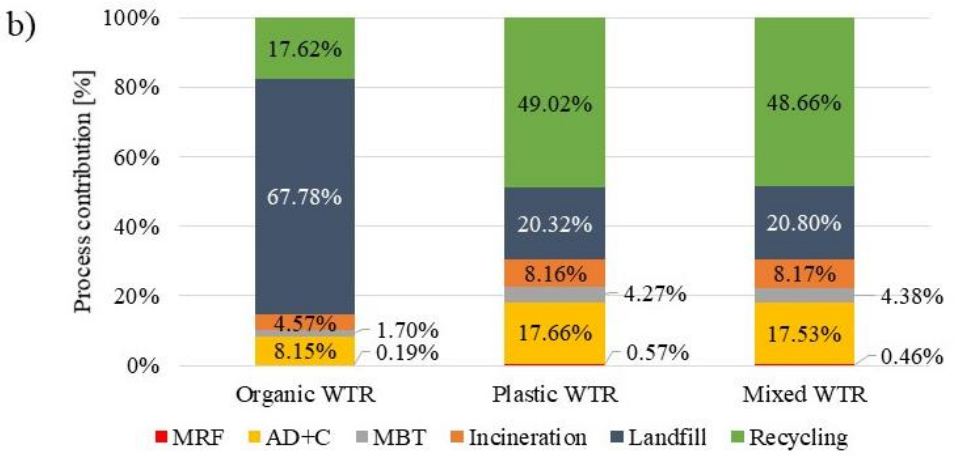


Figure 4.14 - Carbon footprint results (a) and process contribution in CF value (b) of organic, plastic and mixed waste treatment route.

4.4. Multi criteria decision assessment

The waste management decision-making cycle is incomplete without assessing the performance of each option against indicators other than those that directly relate to the goals and aspects of the study. Thus, each assessment was used in a Multi Criteria Decision Assessment (MCDA) in order to select the waste treatment route that will ensure the optimal improvement on the effectiveness of the CAT management.

Concerning the technical assessment, the waste treatment route performance (P) was calculated by summing up the performances of the process (P_p) and the outputs (P_o). The process performance (P_p) achieved the following values for each waste treatment route (Table 4.4):

- Organic: the addition of CAT with organic waste increased the methane production during anaerobic digestion of 5.6%, while the CO_2 production during composting did not change. Then, for this route, a slight improvement value (SI = 0.643) was obtained;

- Plastic: CAT would have a negligible amount in the MRF sorting process, without any excess in the technical specification of the sorted waste streams mainly. For this reason, no effect value (NE = 0.500) was considered for this route;
- Mixed: no differences were founded in all the aerobic biostabilization parameters (temperature, O_2 consumption, CO_2 production, respiratory quotient) between samples with and without CAT. Therefore, no effect value (NE = 0.500) was considered for this route.

On the other hand, the output process (P_o) achieved the following values for each waste treatment route (Table 4.4):

- Organic: the final degradation of CAT at the end of combined anaerobic digestion and composting was not compliant to fulfil the compost quality requirement, resulting in a high worsening value (HW = 0.071);
- Plastic: despite the thermo-chemical properties of recycled LDPE with different amount of CAT were almost unchanged, the processability of LDPE was strongly affected even at small CAT contamination. For this reason, high worsening value (HW = 0.071) was considered for this route;
- Mixed: the heating value of CAT decreased of only 3% after the biostabilization process and, despite a weight loss of about 25%, CAT specimens maintained their shape, confirming a suitability in further incineration. Since the overall heating value of the mixture could be increased, a slight improvement value (SI = 0.643) was considered for this route.

The technical performance P was 0.714, 0.571 and 1.143 for organic, plastic and mixed waste treatment route respectively (Table 4.4). From these results, it is possible to note that plastic waste treatment route achieved the worst performance, revealing how, from the technical point of view, this is not a viable option for CAT treatment. Organic waste treatment route resulted slightly higher, despite the value was not sufficient. Mixed waste treatment route instead resulted the scenario with the highest value (Fig. 4.15a).

The total cost indicator C resulting from the economic assessment was considered as the economic criteria in the MCDA (Fig 4.15b). As discussed in the economic assessment section, the total cost indicator C did not presented a big variability, resulting higher for organic waste treatment route. The main difference relied basically on the results from the mass balance, where the compost disposal increased the costs. Finally, the index used as environmental criteria was the avoided $kgCO_2$-Eq. per ton of waste (GHG_a) of each scenario compared to the worst scenario (the one with the highest CF value). The environmental assessment revealed how the worst result was achieved by the organic waste treatment route (324.64 $kgCO_2$-Eq./t), which was chosen as the reference (zero). The GHG_a of plastic and mixed waste treatment route were calculated by summing the CF of organic scenario to the one achieved by the selected treatment route, as described in Table 4.4. In order to evaluate the avoided GHG, all the results were considered as positive value (Table 4.4). Then, the GHG_a indicator achieved a value of 0, 413.81 and 411.80 (Fig. 4.15c).

Table 4.4 – Technical performance (P), total cost (C) and avoided $kgCO_2$-Eq. (GHG_a) for each waste treatment route used in MCDA.

Indicator	Unit	Organic WTR	Plastic WTR	Mixed WTR
P_p	[-]	0.643 (SI)	0.500 (NE)	0.500 (NE)
P_o	[-]	0.071 (HW)	0.071 (HW)	0.643 (SI)
P	[-]	0.714	0.571	1.143
C	[€/t]	120.35	112.21	109.43
GHG_a	[kgCO2-Eq./t]	-[324.64-324.64]= 0	-[-89.17-324.64]= 413.81	-[-87.16-324.64]= 411.80

From Table 4.4, the alternative matrix and the normalized one (Table 4.5) were created, as described in the section 3.6.2. In particular, the normalized matrix was created minimizing C and maximizing P and GHG_a.

Table 4.5 – Alternative matrix (quantitative and normalized) used for MCDA analysis: waste treatment routes as rows and evaluation criteria as column.

WTR	P [-]	C [€/t]	GHG_a [$kgCO_2$-Eq./t]

WTR	P [-]	C [-]	GHG_a [-]

Organic	0.714	120.35	0.00
Plastic	0.571	112.21	413.81
Mixed	1.143	109.43	411.80

Organic	0.625	0.909	0.000
Plastic	0.500	0.975	1.000
Mixed	1.000	1.000	0.995

The robustness of the obtained solution was verified through a sensitivity analysis. As described in section 3.6.2, 4 stakeholders were assumed in order to vary the weights vector of the evaluation criteria. The application of the SAW-PCT method made it possible to determine, with reference to criteria GHG_a, C and P, respectively, the following weights: W_{Env}= (0.333; 0.500; 0.167); W_{Man}= (0.333; 0.167;0.500); W_{Pol}= (0.500; 0.167;0.333); W_{Bal}= (0. 333; 0. 333; 0. 333).

Finally, the composite indicator Preference Index (PI) was constructed aggregating the indices (P, C and GHG_a) with the different weights obtained by the sensitivity analysis, as explained by [Eq. 9]. Using the normalized alternative matrix, the best option was deemed the alternative with the highest PI. MCDA results of PI referred to each stakeholder considered is shown in Figure 4.15d.

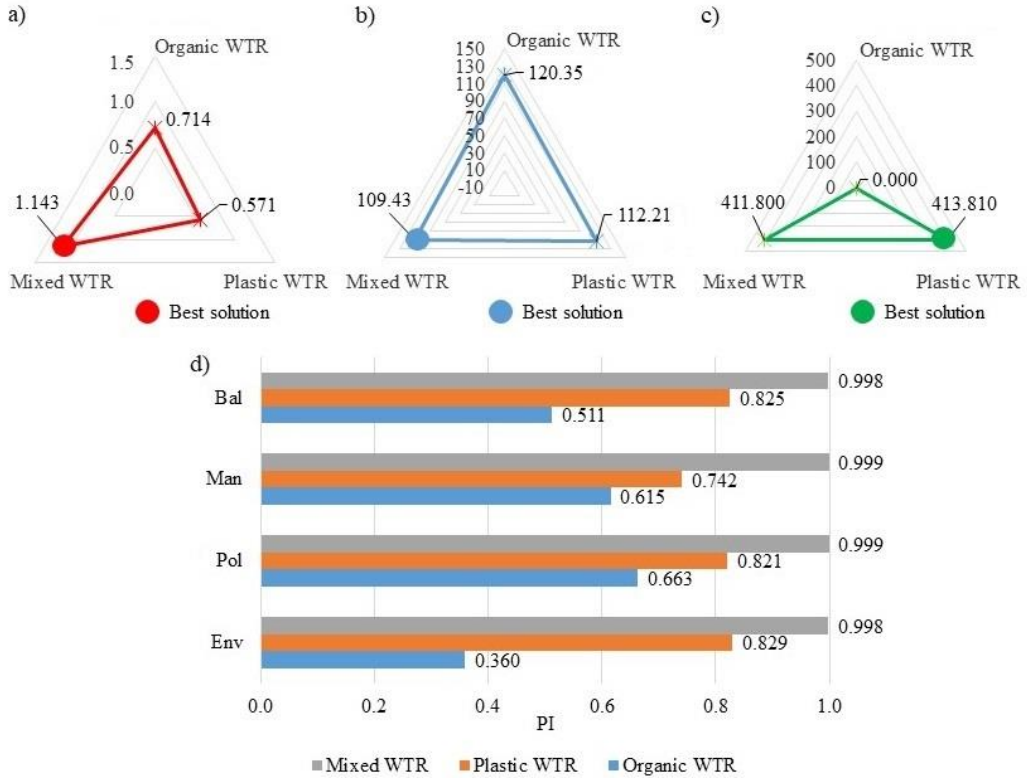


Figure 4.15 - (a) P, (b) C and (c) GHG_a values for each scenario; (d) Preference Index (PI) score for the identification of the best waste treatment route, varying decision- makers.

Results from sensitivity analysis indicated a preference for mixed waste treatment route more than 90% for all decision makers. On the other hand, plastic waste treatment route achieved values that slightly exceeded 80% of preference (except for policy decision maker). Finally, organic waste treatment route occupied the last place, characterized by a lower SC rate (36-66%).

All the waste treatment routes in the ranking of alternatives were similar regardless of varying weights of the evaluation criteria. In particular, the mixed one was every time dominant, confirming how the scenarios ranking clearly indicating it as the best waste treatment route.

These results indicated that, in the current waste management system, the most sustainable effect of CAT waste is obtained from mixed waste treatment route. Aerobic

biostabilization and further incineration seem to be the most preferred technologies for all the stakeholders involved. On the other hand, if CAT is collected with organic waste, the worse effect from the combination technical, environmental and economic criteria is registered regardless the stakeholders point of view. In particular, for the environmentalist (Env) decision maker, this route obtained a preference of 0.36, which is significantly low if compared to the other ones. This means that, nowadays, the current organic waste management system would present the lowest preference among other treatment for CAT, which is in contrast with the general Italian bio-plastics management. Finally, despite plastic waste achieved the worst technical performance, the overall treatment route for CAT showed a suitable preference. The low technical performance is visible from the lowest PI registered for the waste treatment manager (Man) decision maker.

5. DISCUSSION

The principal aim of this work was to point out the effect of a new bio-plastic (the case of cellulose acetate) on the current Italian waste management system. Apart from the collection with organic waste (organic waste treatment route), which is the disposal route that Italy is pursuing, the collection with plastic and mixed waste were also assessed (plastic and mixed waste treatment routes respectively). The idea of this work was to give an objective overview of the technical, environmental and economic performance of each waste treatment chain, providing quantitative results of it.

Results have revealed how the mixed waste treatment is the one that achieves the highest preference among the stakeholders involved, meaning that on the overall this treatment is the most suitable and sustainable according to technical, environmental and economic criteria. On the other hand, the organic waste treatment route revealed the lowest preference, supported by poor benefit or even worsening especially in compost quality. The key factor relies on the not-achievement of the compost quality standards, due to the partial degradation of cellulose acetate during the whole anaerobic digestion and composting process. Indeed, the increase of bio-plastics with organic waste resulted in the quantity of composts that do not meet quality requirements, thereby increasing the costs and environmental impacts of organic waste disposal route.

In this context, it is evident how the organic waste disposal route needs to be improved, if Italy want to continue on this direction. First, thermophilic temperature (higher than 55°C) would be necessary and mandatory in the near future to enhance the degradation under anaerobic digestion and/or composting conditions of most of the bio-plastics commonly used. In addition, the residence time of bio-plastics (but generally organic waste) is an actual issue, which is reported also in other European countries. In

Germany, for example, both anaerobic digestion and composting process are carried out for few weeks, which is too short for a suitable bio-plastics degradation. Indeed, in Germany, but also in other countries, only bio-plastics carrier bags are collected with the organic fraction of MSW while rigid or other bio-plastics packaging are collected with mixed waste. Other strategies to enhance bioplastics degradation under anaerobic conditions are pre- (i.e. thermal or alkaline pre-treatments) or post-treatments (residual bio-plastics screening from compost) (Battista et al., 2021), which actually are is still almost unexplored and the few papers dealing with this topic have reported controversial results (Cucina et al., 2021a). All these strategies are viable since bio-plastics can properly give a significant contribution on methane generation during anaerobic digestion. Assuming 10% of bio-plastics in the organic waste (which is a viable forecast for 2030 looking the increase of bio-plastics in the market), it is reasonable to suppose a contribution of 40% in biomethane potential from the input in anaerobic digester (bio-plastics and organic waste), if suitable process conditions are performed.

The increasing use of bioplastics and their collection within organic waste also challenges the existing regulations concerning compost quality. The limit of 0.5% w/w for the presence of inert materials (e.g. plastics, metals and glass particles with particle size > 2 mm), without any distinction between petroleum-derived plastics and bioplastics, is not only adopted in Italy but was proposed by the European Commission as end-of-waste criteria for biodegradable waste subjected to biological treatments (i. e. compost and digestate) (Saveyn and Eder, 2014). Bioplastics residues in compost should not be taken to count as inert materials (plastics, metals, glass) since they are proved to degrade in natural environments and a potential phytotoxicity of the compost containing bio-plastics residues is averted (Gadaleta et al., 2022a). Indeed, bioplastics disintegration is a necessary part of the whole biodegradation process and leads to smaller particles, which should not be confused with persistent microplastics that remain in the final compost. Even in the case of suboptimal compost processing, the biodegradation process of the disintegrated smaller particles does not stop at this point, but further continues in the soil.

To face bio-plastics waste issue, Italy has introduced in 2020 the national Extended Producer Responsibility (EPR) authority for compostable bio-plastics recycling (called Biorepack) to manage them within the organic fraction of MSW. The creation of Biorepack opens to a main question: If bio-plastics products do not present specific problems or requirements for organic treatment, why should be necessary a specific authority? In addition, Biorepack has fixed the EPR fee for bio-plastics production to 294 €/t (reducing the one of 560-660 €/t previously managed by COREPLA, the authority of plastic waste). This has led and will lead to a significant increase of bio-plastics items on the market. Yet, Biorepack reward the costs for bio-plastics collection and treatment in relation to the quality of organic waste collected. Biorepack set to 1.25% the maximum level of bio-plastics in the organic waste in input as quality range until the whole 2023. This means that only these amounts will be financed by Biorepack to the organic waste treatment facilities, leading to an use of the remaining bio-plastics amount on different treatment chains (mainly mixed waste ones). On the other hand, UK's EPR scheme for packaging (currently being revised) proposes to categorise compostable plastics as non-recyclable (due to lack of sufficient structure), meaning that the strategy of Italy is different from the one adopted by other European countries. It is clear that not only the treatment but also the management of bio-plastics in Italy needs to be improved. Moreover, some of bio-plastics are not compatible with the anaerobic digestion process (e.g PBS) or the composting process is not properly suitable (e.g cellulose acetate). For such polymers, other waste management routes should be preferred and, as revealed by this study, the mixed waste treatment chain could be a viable and suitable option for the treatment of bio-plastics, especially for cellulose acetate, without requiring any significant modification on the chain. The treatment of mixed waste (consisting in aerobic stabilization and incineration) presents the same bio-plastics fate of the one of organic waste treatment route. In fact, during the mixed waste treatment chain, bio-plastics are not recovered in new material but in energy, as well as for the organic waste treatment route where bio-plastics are converted in CH_4 and/or CO_2.

In order to differentiate the preferred disposal route, specific pictograms should be introduced. Indeed, in Europe, the Directive on Packaging and Packaging Waste (94/62EC) states the consumer should also be informed of how to dispose of the product (Van Roijen and Miller, 2022). However, plastics products (either fossil- or bio-based) have already several different label schemes to indicate their chemical composition, whether they can be recycled, are bio-based and/or can be biodegraded and under which conditions. Thus, the introduction of new labels may confuse, rather than inform, costumers (Rosenboom et al., 2022). Then, a common labelling policy should be pursued in order to face not only the widespread unfamiliarity of people about bio-plastics disposal routes but also the misguided purchase decisions and possibly to littering behaviour.

The technical and social problems related to bio-plastics could be reduced with other strategies using more compatible materials, when the properties of plastics are unnecessary. Indeed, in some cases, bio-plastics items could be replaced with other materials like paper or cardboard, which can be treated in an easier way in the organic waste treatment route (Dolci et al., 2021). This strategy is also supported by EU since the European Strategy for Plastics in a Circular Economy suggests that innovative materials and alternative feedstocks for plastic production should be developed and used where the sustainability is clearly improved (European Commission, 2018).

Finally, the possible separate collection of bio-plastics and further recycling should be encouraged in a scenario of increased use of these products. Bio-plastics management could move from the traditional degradation paradigm to a new one where bioplastics are no longer intended as single-use products but are projected to be recovered in a circular economy perspective. This would shift the plastic production from the conventional polymer to the bio-based one, highlighting the benefit of bio-plastics during the production stage. In this context, there is still an open debate in deeming the organic treatment through anaerobic digestion and/or composting as recycling process. Indeed, the Packaging and Packaging Waste Directive (PPWD), which is being revised, do not consider the organic reprocessing as recycling activities and also

require a separated and defined streams for recycling processes in the next 5 years for all the packaging, including bio-plastics.

Based on these scenarios, the governance systems and management strategies for bio-plastics waste that were proposed so far need to consider all these aspects in order to combat the possible future issues from these materials. Politic authority as EU should guide this coordinate transition, supporting bio-plastics development through ambitious and collaborative research that aims to transform and improve the waste management system over the coming years. In this way, EU could cover the almost inexistence of specific regulatory frameworks on bio-plastics waste, considering the different waste treatment conditions around Europe.

6. CONCLUSIONS

The present study has evaluated the technical, economic and environmental effects of cellulose acetate (CAT) on the waste management system, identified as organic, plastic and mixed waste treatment route.

Technical assessment achieved the following results, according to each treatment route:

- Organic: CAT increased the methane production during anaerobic digestion but the degradation did not fulfil the compost quality requirement;
- Plastic: CAT could be sorted mainly with LDPE and PP packaging films streams and that level of contamination can strongly affect their mechanical recycling;
- Mixed: CAT has not altered the aerobic biostabilization of mixed waste; has maintained the shape without significant disintegration and can increase the heating value of processed waste for further incineration.

From technical results, the economic and environmental assessment achieved the following results:

- The three waste treatment routes reached a similar total cost (120-109.43 €/t), where the highest cost was achieved by the organic one for the disposal of not-compliant compost;
- The environmental results confirmed a similar benefit for plastic and mixed waste treatment routes (about -88 $kgCO_2$-Eq/t) but high environmental burden for the organic one (more than 300 $kgCO_2$-Eq/t) for the compost disposal presented above.

The MCDA analysis, combining each assessment, pointed out a preference of more than 90% for mixed waste treatment route of CAT, while plastic and organic waste treatment routes reached an overall preference of 80 and 50% respectively.
Results highlighted the following issues:

- Organic waste treatment chain needs to be improved, considering the increasing amount of bio-plastics in the next future, as well as the compost standard regulation;
- Bio-plastics could be treated with a combination of technologies and treatments, not only to anaerobic digestion and composting;
- Clear and concise labelling should be formulated, in order to correctly inform customers for the right disposal routes of each bio-plastic waste;
- Appropriate materials should be developed and applied, avoiding plastics (either fossil- or bio-based) when it is not necessary.

Bio-plastics research opens to further investigation, which are nowadays still not clear:

- Pre- and post-treatment of bio-plastics should be assessed in order to enhance the degradation during biological process and increase the compost quality at the end of these process;
- Future researches have to gain a better understanding of the origin and creation of microplastics from bio-plastics and their release into and effect on the environment;
- A detailed Life Cycle Assessment for bio-plastics waste needs to be modelled, opening to other impacts from bio-plastics during different treatments and in specific scenarios, in order to limit the assumptions that lead to high levels of variability and uncertainty in environmental impact results.

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This achievement is not and end but just a small stop to enjoy what was done in these three years. But this has to continue since “this is the way of those who leave and arrive to leave again”.

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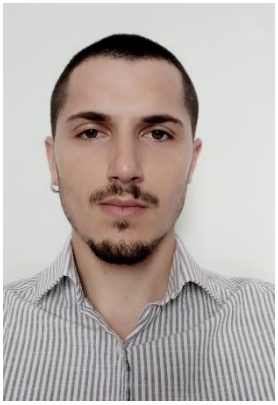

Giovanni Gadaleta was born in Molfetta (Italy) in 1995 and graduated from Polytechnic University of Bari in 2019 with a MSc in Environmental and Territorial Engineering, discussing a thesis in Solid Waste Management entitled “Performance analysis of advanced selection of dry fraction from separate collection of MSW in the facility of Molfetta (BA)”, with supervisor Prof. Michele Notarnicola and co-supervisor Eng. Silvio Binetti and Prof. Sabino De Gisi, with final grade 110/110 cum laude. From November 2019 to December 2022, he was a Ph.D. student in Risk and environmental, territorial and building development Ph.D. program at Polytechnic University of Bari. He spent an year at Hamburg University of Technology (Germany) as visiting Ph.D. student, where he carried out several experimental test on cellulose-based bio-plastics waste treatments in the current waste management system. His research activities aim at developing and investigating environmental engineering activities, process sustainability, management and treatment of waste and wastewater.

PUBLICATIONS

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