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Review

Overview of municipal solid wastes-derived refuse-derived fuels for cement co-processing

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ABSTRACT

The global municipal solid waste (MSW) generation rate is 2.01 billion metric tonnes annually with an average of 0.74 kg waste/person/day. Approximately 92 % of the MSW originates from organics composition (e.g., food waste; plastic; paper; garden waste/woods, and textile), where 33 % of overall MSW is improperly managed in an efficient and environmentally safe manner. One of the promising methods to solve MSW management issues is to convert MSW into refuse-derived fuel (RDF) that can be used for the clinker burning process in cement kiln to replace the usage of fossil-based solid fuel. Thus, the potential of local MSW composition in energy recovery; suitability of RDF production technology; as well as international-industry requirement on RDF in co-processing and environmental concerns are discussed. Due to heterogeneous composition and sizes in nature, high moisture, and substantial amount of chloride content in MSW, it needs to undergo pre-treatment processes to enhance the RDF's physio-chemical properties that comply with RDF ASTM/EN standards, where expected high heating value (HHV) is > 20 MJ/kg, ash (<10 %), Chloride (Cl) (<0.80 – 1.00 %), Sulphate (S) (<1.50 %), Nitrate (N) (<1.00 %). As the development of upgraded MSW to RDF is still new in the commercial phase, there is minimal information and data on the techno-economic analysis, as well as recent industrial-scale of thermos-chemical conversion technologies for RDF preparation. Hence, the present work provides a clear picture on overview of municipal solid waste (MSW) generation, MSW composition, MSW pretreatment, and application as co-processing in cement industry are discussed. Besides, recent thermochemical upgrading process (torrefaction, dry carbonization, and hydrothermal carbonization) of MSW from R&D to commercial scale was further highlighted. In summary, this review serves as basic criterion and strategies to explore the new path of upgrading the waste into RDF for the purpose of sustainable energy recovery that adopting in circular carbon economy framework.

1. Introduction

With the global population reaching 8 billion as of 2023 [1], massive amounts of municipal solid waste (MSW) are generated each year, estimated to be more than 2.01 billion tonnes globally, with a projected increase to 3.40 billion tonnes by 2050 [2]. This poses a serious threat to global health and the environment. In managing municipal solid waste, limited land remains a crucial problem today as there are few appropriate locations for site treatment plants and disposal. While effort has been made to instill recycling practices in communities, glaring issues in implementation can be seen such as inadequate legislation, poor enforcement, and citizen noncompliance can be noticed. Currently, the majority of MSW is landfilled (including unidentified sites) and dumped both illegally and legally [3]. The accumulation of inappropriate and unregulated wastes at landfills can result in methane gas (CH_4) emissions, an anthropogenic greenhouse gas with a global warming potential 34 times larger than carbon dioxide gas (CO_2). These disposal methods are unsustainable because they require huge tracts of land to bury the trash, resulting in land scarcity, leachate leakage, and soil contamination. Furthermore, 38 of the world's 50 largest dumpsites have been

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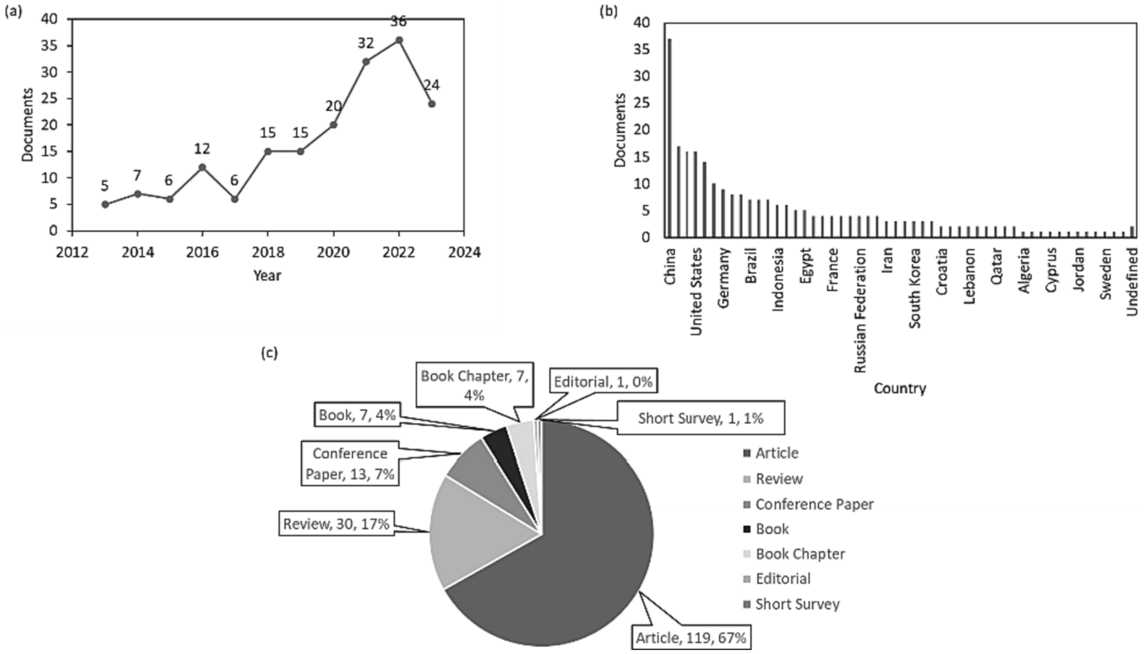

Fig. 1. (a) Distribution of papers by year, (b) Distribution of papers by country, (c) Distribution of papers by category.

identified as indirect sources of marine dumping, damaging marine life, and causing coastal erosion [4].

Rapid economic and population growth raises electrical and resource demand for construction materials such as cement [5]. Cement production is an energy-intensive process that necessitates high temperatures above 1450 °C and a large amount of energy (3000–6500 MJ), resulting in high production costs [6]. To address these problems, the government and cement industry are both exploring sustainable waste management systems and energy sources. One approach is to extract recyclable and combustible components of MSW and convert them into densified solid fuel called refuse-derived fuel (RDF). RDF, in the form of fluff, pellets, or baled paper and plastics, has been widely employed in power generation facilities and incineration plants due to its partial carbon neutrality and reduced sulphate (S) and nitrate (N) content when compared to fossil-based solid fuels such as coal [7].

The cement industry provides several features in the clinker kiln that enable RDF co-processing activities. The clinker kiln runs at high process temperatures of around 2000 °C in an oxidizing environment with a long residence period, permitting thorough decomposition of organic compounds for good combustion [5]. Globally, RDF replaces fossil-based solid fuels in the cement industry at a rate of roughly 13 % in Canada, 16 % in the United States, and 44 % in the European Union [8].

In addition, the trend of publication on relevant subjects concerning MSW management and refuse derived fuel for cement co-processing in the cement industry has expanded significantly over the past decade, as evident in Fig. 1 **(a)** from the Scopus database. Fig. 1 **(b)** indicated that a total of 178 documents were published by authors from 59 different countries, with China leading in term of the highest number of publications, followed by India, Poland, the United States, the United Kingdom, and Australia. This demonstrates a substantial interest in this topic among scholars, particularly from developing nations like China and India. Among 178 technical publications, only 119 articles and 30 reviews were identified within this span of a decade. The overview distribution of these 178 articles across different categories is shown in Fig. 1 **(c).**

The assessment of previous literatures in the last ten years (2013 to 2023) validates the research gap on refuse-derived fuel that employs municipal solid waste (MSW) as feedstock for the purpose of co-processing energy resource in cement industry. As refer to the relevant review studies outlined in Table 1**,** it is worth noting that it lacks specific focus on the practical and feasible technologies for conversion of MSW into RDF that comply with fuel standard for cement co-processing. Most of the studies focused on broader interest such as circular economy using alternative fuels/refuse-derived fuel, assessment of pollutants during co-processing of solid wastes in cement kilns, and technical solutions for waste-derived alternative resources.

Although MSW as a solid fuel feedstock has been extensively researched, yet there is limited knowledge regarding its suitability and feasibility as co-processing fuel in the cement industry. As a result, there has been a paucity of studies into viable conversion technologies for producing a fuel that meets the alternative fuel standards required by the cement industry. Based on this notion, the current study intends to comprehensively review refuse-derived fuel (RDF) production from municipal solid waste (MSW) for sustainable co-processing in the cement industry. This paper attempts to systematically address the following research questions: 'Are MSW composition suitable for energy recovery?', 'What are the present challenges encountered in cement co-processing activities?', 'What are the viable MSW-based RDF conversion technologies?', and 'What are the cement-industry requirements for RDF co-processing and its environmental concerns?'. The review also sheds light on contemporary commercial-scale MSW upgrading technologies for RDF production, and their techno-economic analysis. This approach makes it feasible to strengthen the circular economy and improve solid waste management practices in society. Hence, the present review retains its originality and distinctiveness by addressing this research gap.

1.1. *Composition profile of MSW*

Understanding the characteristics and compositions of generated MSW is crucial prior to the selection of waste-to-energy (WtE) technologies for RDF production, as these two factors significantly impact the optimal recovery of energy from wastes. Generally, MSW is generated from numerous sources such as residential, commercial, industrial, construction, institutional, agricultural, and municipal services. Residential (households) have been recorded as the highest generation of solid waste [20,21].

Effective waste sorting is necessary to ascertain the precise composition of MSW for further upcycling process. Due to varying regional

Table 1
Summary of literature.

Year/Author	Year	Title	Summary
Present study		Overview of Municipal Solid Wastes-derived Refuse-derived Fuels for Cement Co-processing	MSW composition and characteristic for energy recovery; criteria of MSW-derived RDF for cement co-processing; feasible and practical RDF production technology; techno-economic analysis of RDF.
Mateus et al. 2023 [9]	2023	Modern Kiln Burner Technology in the Current Energy Climate: Pushing the Limits of Alternative Fuel Substitution	Existing kiln burner technologies that used to combust alternative fuels are reviewed to access maximized alternative fuel processing and minimizing environmental impact.
Kahawalage et al. [10]	2023	Opportunities and challenges of using SRF as an alternative fuel in the cement industry	Waste generation problems are discussed, detailed overview of SRF is provided including its pretreatment methods and characterization, and scenarios of cement kiln systems that can be used for coprocessing of SRF are reviewed including its challenges.
Kusuma et al. 2022 [11]	2022	Sustainable transition towards biomass-based cement industry: A review	The importance of sustainable biomass transition policies in the cement industry is underlined, which include disseminating niche ideas, expediting transition, and reconfiguring systems by destabilizing current regimes that impede system shift. Intervention policies for biomass-based cement systems are also recognized across different locations.
Sarquah et al. [12]	2022	Bibliometric Analysis; Characteristics and Trends of Refuse Derived Fuel Research	The progress of RDF research trends for the last 30 years is examined using bibliometric analysis from the SCOPUS database across knowledge domains such as conversion processes, applications, and management.
Jąderko-Skubis [13]	2021	Production of alternative fuels from waste: assumptions for the design of new fuel recipes	Appropriate proportion of municipal and industrial waste streams is investigated, key properties of waste-derived fuels are identified with a set of variables and interactions that contribute to the design process are recognized. The novel waste streams as alternative fuels in the cement industry are investigated.
Liu et al. [14]	2020	Review on the current status of the co-combustion Technology of organic Solid Waste (OSW) and Coal in China	The physicochemical features of OSW were examined, as well as a synopsis of co-combustion fundamentals, pollutants emissions, pretreatment methods, and considerations on future coprocessing technology of OSW with coal. The work highlighted on understanding of OSW and coal coprocessing technologies as guidelines for further usage in power plants.
Yang et al. [15]	2019	Unintentional persistent organic pollutants in cement kilns co-processing solid wastes	There is information offered on ways for reducing emissions of various inadvertently created persistent organic pollutants (POPs) from cement kilns co-combustion solid waste.
(Thanos) Bourtsalas et al. [16]	2018	Use of non-recycled plastics and paper as alternative fuel in cement production	Total energy consumption from cement industry in the United States is computed, waste-to-energy capability and related reduction in landfilling are evaluated, dioxin emissions are investigated, and CO2 emissions are determined using life-cycle assessment.
Shehata et al. [17]	2016	Role of refuse-derived fuel in circular economy and sustainable development goals	The function of RDF in achieving the United Nations Sustainable Development Goals (SDGs) and the circular economy is examined, as well as the impediments to RDF usage. Prospective study directions and metrics for incorporating solid waste into the circular economy and SDGs are presented.
Rahman et al. [18]	2015	Recent development on the uses of alternative fuels in cement manufacturing process	The energy values, benefits and drawbacks, GHG emissions, and environmental effects of several alternative fuels currently employed in the cement business are discussed. For further investigation, a multi-criteria decision-making technique is advised.
Aranda Usón et al. [19]	2013	Uses of alternative fuels and raw materials in the cement industry as sustainable waste management options	Technical, economic, and environmental evaluation of the cement industry's focus on municipal solid waste (MSW), meat and bone animal meal (MBM), sewage sludge (SS), biomass, and end-of-life tyres (ELT).

profiles, the availability of data documentation on MSW compositions remains highly limited. Typically, the dominant portion of MSW in urban area composed of food waste (39.78 %), plastic (16.67 %), paper (13.28 %), diapers (7.89 %), garden waste (4.81 %), glass (3.40 %), metal (3.15 %), textile (3.30 %), tetra pak (1.96 %), rubber (1.93 %), wood (1.44 %), and leather (0.37 %). The physical composition of MSW is categorized into two groups of waste, which are organic and inorganic waste as listed in Table 2 [22,23]. On average, the main components of MSW are highly organic and have the potential to be converted into energy sources like biofuels. Wet organic fractions of MSW, such as food waste and sewage sludge, may be utilized as material for the generation of biogas via anaerobic digestion (a biological process in which microbes dissolve down organic substances in the absence of oxygen, producing methane-rich biogas), whereas other organic fractions of MSW may be transformed into biofuels via fermentation and pyrolysis. Most biofuels

Table 2
Physical-chemical composition of MSW [22–24].

Physical composition	Basic classification	Composition (%) *	Examples	Chemical composition
Organics	Food waste	46.09	Vegetables, meats	Cellulose, carbohydrate, protein, lipid
	Garden/Yard waste	5.58	Dried leaves, twigs, cut grasses	Cellulose hemicellulose, lignin
	Textile and rubber	6.06	Clothes, leather products	Nylon
	Paper and corrugated box	15.39	Newspaper, different types of paper and box	Cellulose
	Mixed plastic	19.31	Disposable water bottles, food packaging/ container, plastic film, tubes, polyethylene bag, plastic toys	Polyethylene terephthalate, high density polyethylene, Polyvinyl chloride, Low-density polyethylene, Polypropylene, Polystyrene, Multilayer plastic
Inorganics	Glass	3.93	Different types of mixed glasses from daily usage, restaurant, laboratory, etc.	Formers (Silicon dioxide); Fluxes (Sodium carbonate, Potassium carbonate) Stabilizers (Calcium carbonate); Boric Oxide; Lead Oxide
	Metal	3.64	Ferrous products, zinc, chromium, and vary type of metal products	Aluminum, tin-coated steel (tinplate) and electrolytic chromium coated steel (ECCS)

*Average of 5 years: no major changes in trend of composition waste from 2013 to 2017.

made from MSW are typically liquid fuels like bioethanol (fermentation of sugars and starches found in organic waste), biodiesel (lipids and fats from MSW through transesterification), and syngas (mixture of carbon monoxide and hydrogen through pyrolysis or gasification), but this study focuses on biofuels in solid form.

2. Future opportunities and challenges in cement industry

2.1. Co-processing of alternative solid fuels in cement industry

Cement is a hydraulic binder which means it sets, hardens, and

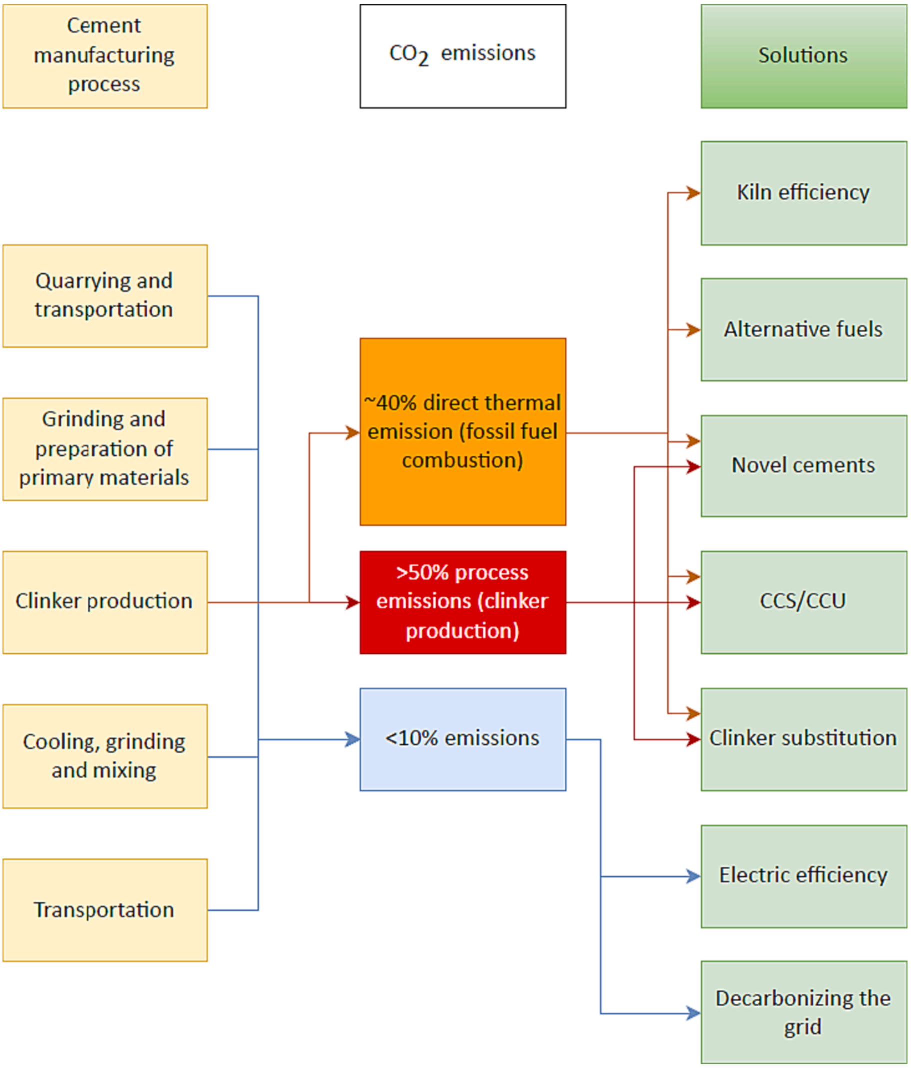

Fig. 2. Emissions and mitigation solutions along the cement manufacturing process [30].

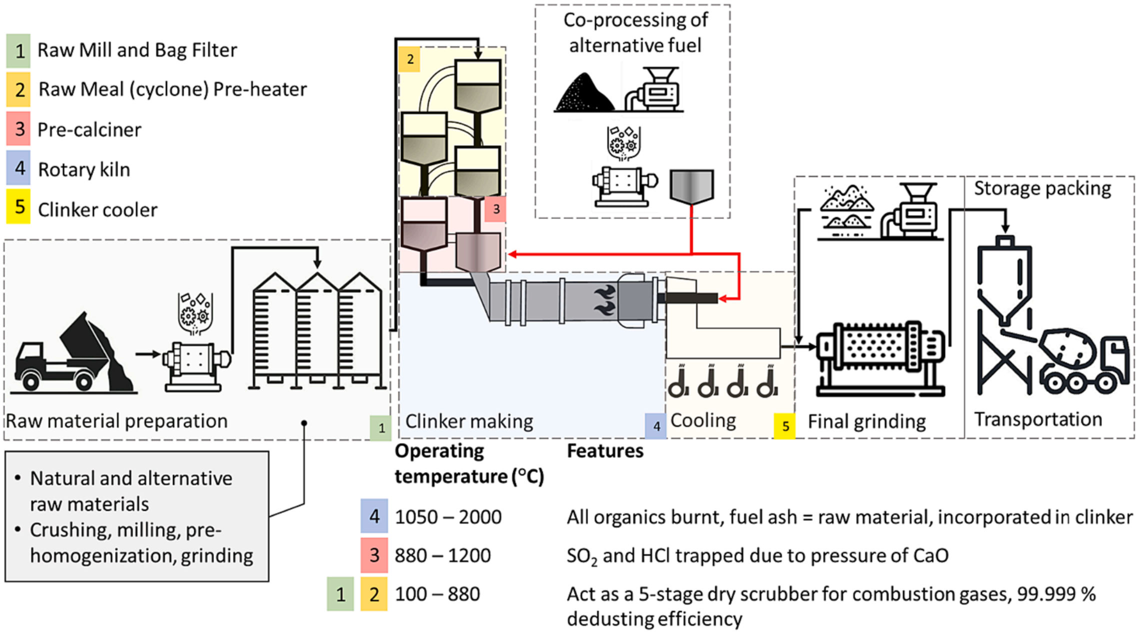

Fig. 3. Co-processing of RDF in clinker kiln.

achieves proper strength characteristics even underwater conditions, after its fine aggregate is blended with water [25,26]. The principal raw materials needed to form the resulting hard intermediate product called clinker are typically limestone ($CaCO_3$), and other materials such as chalk, shale, or clay and ferrum containing calcium oxide (CaO), silica dioxide (SiO_2), aluminium oxide (Al_2O_3), and small amount of iron oxides (Fe_2O_3) [5].

The process of cement production begins with, 1st stage: raw materials extraction and preparation (quarry); followed by 2nd stage: pyro-processing, and finally 3rd stage: cement grinding and distribution [27]. The process involves quarrying or preparation of raw materials, including excavation of limestone. The extracted raw materials are then crushed and subsequently transported to the cement plant for intermediate storage, proportioning and grinding of raw meal through milling process. Following that is pyro-processing where the grounded raw meal is preheated and then sent to the rotary kiln to around 1450 °C using hot gases from the kiln to burn off the impurities. Rotary kiln continuously mixes the ingredients and then subsequently sends them to clinker cooler for cooling process. To control how quickly cement sets, a little amount of gypsum is added to the clinker during the final stages of cement grinding and distribution. Gypsum and fly ash combination is finely pulverized to create pure cement. After all the processes are completed, the cement is then stored in the silos before being packed and loaded for delivery.

Cement manufacturing is an energy and carbon-intensive process which consequently a noteworthy contributor to CO_2 emissions [28]. More than 50 % of the natural release of CO_2 is associated with pyro-processing (calcination) process under intense heat, in which $CaCO_3$ is transformed into lime (CaO) in the following reaction: $CaCO_3 \rightarrow CaO + CO_2$ [29] and a further 40 % of CO_2 emission is caused by the fossil-based solid fuel combustion for burning the clinker to high temperature and running the pre-heaters and other operations [30]. Another remaining 10 % of CO_2 emission is caused by transporting raw materials. About 0.9 tonne of CO_2 is released for every tonne of cement produced. Besides, enormous energy sources are needed in the precalciner tower and at the end of the kiln.

Worldwide, fossil-based solid fuel like coal is primarily needed for the thermal energy generation during the calcination process in preheater tower and during the clinkerization process in the kiln [31], which accounted for 30–40 % of the industry's cash expenses [32]. High operating costs due to fluctuations in natural energy prices have urged cement manufacturers worldwide to seek for potential replacement of fossil-based fuel using various types of feedstocks [33]. Fig. 2 shows the substitution of fossil-based solid fuel by alternative fuels as one of the solutions made by cement industry to reduce operating cost and lowering emissions [30]. The use of refused-derived fuels (RDF) like tires, scrap rubber, plastic, sewage sludge, graphite waste, petroleum pellets, electrode coke, paper waste, household waste, agriculture waste, wood, etc is a viable option as alternative biofuels at cement industry. Some of the merits of refuse-derived fuels (RDF) that make them attractive as material for production of biofuels are that they are deemed cheaper to adapt in a cement kiln than to build specific incinerator for the waste incineration [34], their resources are rich, abundant, and never-ending (sustainable) compared to coal. Furthermore, MSW was offered as a re-burning fuel and re-generable biofuel during co-firing since 10–20 % of total heat input reduced NOx emissions by about 70 % [35].

The substitution rate varies from country to country depending on the local waste markets and permitting conditions in every country but most of the European countries are already way ahead than the rest of the world [18]. It was reported that usage of alternative fuels in different countries has substituted the usage of conventional fuels in clinker kiln at about 7.8 % in Australia (2013), 15.5 % Japan (2012), 45 % Sweden (2011), 41 % Switzerland (2012), 85 % Netherlands (2011), 11.3 % Canada (2008), 53.6 % Germany (2010), 18 % EU (2012), 45 % Poland (2010), 22.4 % Spain (2011), 60 % Belgium (2011), and 8 % USA (2004) [18]. By using liquid waste in the main burner, some plants may reach a permanent substitution rate of up to 80 %, and some can achieve 100 % [36]. Alternative fuels breakdown in the European Union shows that 37 % are plastics, 18 % are mixed industrial wastes, 15 % are tyres, 7 % are other fossil-based wastes, 7 % are biomass, 6 % are animal bone meal (ABM) and fats, 5 % are solvents, 3 % are impregnated sawdust, and 2 % are waste oil [37].

Cement plants also meet general requirements of applicable laws that follow procedures for incinerating waste [51]. As shown in Fig. 3, the main two locations for co-processing activities of alternative fuels in clinker kiln are at the pre-calciner and through the main burner firing at rotary kiln outlet. The European Integrated Pollution Prevention and Control Bureau has recognized several features in the clinker process that allow the use of alternative fuel for co-processing activities as

Table 3
Characteristics that are most desirable for alternative fuel co-processing in cement plants [a][42], [b][43], [c][44], [d][45], [e][46].

Properties		Value	Unit	Possible impacts
Calorific value		> 14 [a]	MJ/kg	Release energy into the burning chamber for combustion process. [b]
Moisture content		< 20[c]	%	High amount of moisture may lower kiln system productivity and efficiency.
Ash content				Affects the cement chemical structure and may necessitate a modification in the raw material mix composition. [b]
Chloride content		< 0.5[d]	%	i. Excess chloride may result in dust from the cement kiln or bypass (which may necessitate construction of a bypass). [b] Cause the creation of acid gases, which may promote corrosion on the cement kiln surface if these gases accumulate. [d] Affects the general quality of both concrete and cement, as well as the clinker strength. [e]
Size	2D	< 20[c]	mm	Subject to specific cement plant process limitations. [c]
	3D	< 70[c]	mm	
Sulphate content		< 2[c]	%	Most of the sulphate is trapped by the clinker's alkaline matrix. Sulphate content might have an impact on overall product quality.[a]
Heavy metals content		< 2500 [a]	ppm	i. Mercury (Hg) < 10 ppm, Cadmium (Cd) + Thallium (Tl) + Mercury (Hg) < 100 ppm. [a] Non-volatile heavy metals are successfully integrated into clinker, whereas semi-volatile metals are caught in the clinker stream or dust. Highly flammable metals like Hg and Cd are an exception. [d]
Phosphate content				Influences setting time [b]
Chromium				Sensitive users may experience allergic reactions. [b]
Sulphate				SO2 emissions are possible. [b]
Organic carbon				Carbon monoxide (CO) and volatile organic compound (VOC) emissions are possible. [b]

follows [5,6]: (1) Rotary kilns operate at maximum temperature: 1050 to 2000 °C or higher in the main firing system and flame temperature with a gas retention time of about 8 s; (2) Gas retention periods of more than 2 s inside the secondary firing system at temperatures higher than 850 °C (880 – 1200 °C) in pre-calciner; (3) Oxidizing conditions with good blending conditions assuring good combustion and avoiding the generation of CO and other toxic compounds in the rotary kiln; (3) High thermal inertia and thermally consistent conditions with sufficiently long retention times in the kiln completely destroy organic pollutants; (4) Uniform burnout conditions for load fluctuations; (5) High capacity for particle-bound toxic substances retention; (6) Ash retention in clinker, long residence times; (7) Any non-volatile residues from combustion are trapped in the clinker matrix; (8) Alkaline environment inside the kiln; (9) Large furnace area; and (10) Significant length of the kiln. To ensure optimum kiln performance, complete combustion, and minimal emissions, it is crucial to identify the proper feeding points (primary firing, secondary firing) and conveying technology for the fuel. This is due to the temperature variations between various process components, particularly the pre-calciner and main burner. Primary firing is where the primary fuels (typically petroleum coke, coal, or natural gas) are utilized to provide heat to cement kiln where materials are fully calcinated to produce clinker which is then mixed with about 8 % gypsum to create Portland cement. Besides, secondary firing is where secondary fuels (natural gas, petroleum coke, coal, chipped tyres, solid-recovered fuels, bio-fuels, non-hazardous liquid wastes, etc.) are used to provide heat to precalciner where raw feedstocks are combusted to remove volatiles and start calcining the feedstocks. Appropriate feeding points will be chosen depending on the suitability of the alternative fuels based on the physical, chemical, and elemental properties. Due to the high temperature and lengthy retention time, high-quality fuel with a substantial energy value, including fuel containing volatile organic components (VOC), should be added to the primary burner in order to ensure complete combustion. Meanwhile, medium-grade fuel with an average energy content can also be supplied to the pre-calciner burner [38]. In general, the most common feeding points in a cement production plant are [39]: (1) At the rotary kiln outlet end, through the main burner; (2) At the riser duct at the kiln inlet via secondary burners; (3) At the pre-calciner via pre-calciner burners; (4) At the pre-calciner using a feed tube (for lump fuel).

2.2. Criteria of alternative fuels in cement industry

Due to the mixture of various wastes are used as alternative fuels, the specifications of the fuels must be evaluated to comply with the health, safety, and environmental standards before delivering them to the cement plant (pre-calciner/kiln). The desirable characteristics of the alternative fuels and possible impacts on the environment and cement quality are listed in Table 3**.** The requirements include suitable chemical composition of the component and its organic content, as well as physical characteristics that are reasonably homogeneous and manageable for transportation (size and density) [40]. The fuels must also have high calorific value (CV) that can release large amount of energy into the burning chamber for combustion process [41].

Table 4
Fossil-based and alternative solid fuels used in cement kiln.

Feedstock	C (%)	CO_2 (m^3/kg)	S (%)	SO_2 (m^3/kg)	N (%)	HHV (MJ/kg)	Challenges	Ref.
Bituminous coal	70.60	1.309	1.30	0.009	1.20	27.40	- High sulphur concentration	[18,21]
Petcoke	89.50	1.659	4.00	0.027	1.71	33.70	- Poor igniting and burnout properties (low volatile)	[18,21]
Meat and bone meal	42.10	0.781	0.38	0.003	7.52	16.20	- High levels of ash and chlorine (most chloride is found as salt (NaCl)). - N content is ~ 7 – 8 times higher than in coal - High potassium and sodium alkali levels (which cause cracking in concrete and mortar and clog preheater units)	[18,21]
Sewage sludge	40.50	0.751	0.12	0.001	0.84	15.80	- Ash contain high SiO_2, Al_2O_3, and Fe_2O_3 (affect cement quality) - High chlorine $> 0.2 – 0.5$ % (can cause clogging) - Contain high nitrogen	[18,21]
Car tyre scrap	79.60	1.476	1.54	0.010	0.43	35.50	- Excessive zinc oxide in tyre may retard setting time and strength of cement (causing incomplete combustion) - Cannot be finely shredded economically	[18,21]
Car tyre rubber	87.00	1.613	0.80	0.005	0.33	35.60		[18,21]
Coal/coke mix	75.10	1.392	3.00	0.020	1.70	29.00	- CO_2, SO_2, and NO_x pollution - Reduce boiler performances	[18,21]
Fluffy RDF (mixed plastic and paper)	70.60	1.309	0.20	0.001	1.20	25.02	- Higher levels of moisture and chlorine - Variations in quality and composition	[21,34]

Table 5
RDF used globally and its purposes in different countries [58].

Country	Type of waste	Energy value (MJ/kg)	Method	Industries	Yearly production (Mt/year)
Japan	Dry and non-hazardous industrial	> 25.00	Mechanical-biological treatment	Cement, steel, and pulp industries, district heating facilities	2.70 (2013) 3.00 (2015)
Thailand	40 % plastics, 30 %-yard waste, 10 % food waste, less than 10 % paper, and 10 % non-combustible	~ 19.60	Mechanical treatment	Cement kilns and power boilers	~ 2.46
India	Plastic, wood, pulp, or organic waste, chlorinated	~ 19.60	Mechanical treatment	Thermal power stations, incineration plants, cement industry	~ 1.37
Egypt	Urban wastes, agriculture waste, RDF, shredded scrap tyres	11.70 – 16.70	–	Cement kilns	0.22 (2015)
Germany	MSW and industrial or commercial	13.00 – 27.00	–	Incineration plants	8.70
Austria	Selected or mixed non-hazardous waste	12.00 – 25.00	Mechanical treatment	Incineration plants, cement kilns	1.50
Ireland and United Kingdom	MSW	9.00 – 20.00	Mechanical treatment	Incineration and co-incineration plants, cement industry	3.20
France	Recoverable waste	12.00 – 20.00	Mechanical treatment	Cement kilns	2.30–8.00
Italy	Dry and bio-dried waste fractions	15.00 – 20.00	Mechanical-biological treated	Incineration plants, cement kilns, coal co-combustion	4.49 (2017) 0.15 (2017)
Finland	MSW, industrial/commercial waste streams, construction, and demolition wastes	20.20 – 22.40	Mechanical treatment	Gasification facilities, incinerations /co-incineration plants	0.5 (2014)

2.3. *Environmental perspective and limitations of alternative fuels*

The MSW-derived alternative solid fuels usage in cement co-processing includes its impactful effects on the environment. Such as conserving resources (reduction of non-renewable fossil fuels like coal), maximizing recovery of energy, reducing waste disposal in landfills, and decreasing emissions of NO_x, SO_2, and CO_2 [34]. Table 4 indicated the emission profile of fossil-based and alternative solid fuels combustion in cement kiln with CO_2 and SO_2 emission calculated based on following formula [35]:

$$CO_2\left(m^3/kgfuel\right) = \frac{1.854 \times C(\%)}{100}$$

$$SO_2\left(m^3/kgfuel\right) = \frac{0.680 \times S(\%)}{100}$$

The CO_2 emissions of derived alternative solid fuels is considerably lower than bituminous coal, pet coke, and coal/coke mixed (1.309 m^3/kg). Generally, higher carbon content resulted in the higher emission of CO_2. Sulphur content also has direct relationship with its emission where higher sulphate content emits higher SO_2. In this case, the usage of alternative solid fuels has the potential to reduce SO_2 level to 0.001 m^3/kg when apply as co-processing in cement kiln. In comparison to coal, RDF has a noticeably low N content of roughly 1.20 %, implying a negligible contribution to NOx generation. Although SOx contents were ineffectual at steady calcination temperatures, NOx emissions were lowered by around 17 % when alternative solid fuels were added to the raw combination of fossil fuels. Legislative restrictions on NOx and SOx emissions have compelled accountable organizations to consume the RDF mix at high temperatures (more than 950 °C) in order to decrease NOx emissions [47]. At this point, estimating NOx emissions from nitrogen concentration is impossible owing to NOx generation, which is affected by kiln temperatures, residence periods, burner types [45], and nitrogen in combustion air.

In regards with alternative solid fuels's energy efficiency, higher heating value (HHV (MJ/kg)) or energy content in the fuel is taken as key criteria, shown in Table 4**.** Most studies found that the HHV of waste-derived solid fuels was comparable to that of the majority of traditional fossil fuels, such as coal. For example, alternative solid fuels made of paper and plastic mixture has about 25.02 MJ/kg, while meat and bone meal has about 16.20 MJ/kg, sewage sludge has about 15.80 MJ/kg, and car tyre rubber has about 35.60 MJ/kg.

Nevertheless, there are also several challenges in adopting alternative solid fuels in the cement industry as shown in Table 4**,** which depending on the types of fuel precursors, potential heterogeneity, and characteristics of the fuel [31]. For instance, the main challenge of using MSW as feedstock for alternative solid fuels is that it is heterogenous in nature and contains high moisture which could affect its potential for energy recovery and calorific value [48]. High water content in MSW prompt microbial contamination and that escalates the formation of extremely strong leachate [49]. MSW also contains a high concentration of chlorine that might result in dioxins emission and cause corrosion. According to the partitioning behaviour during combustion, chlorine is the most volatile matter, which generates chlorinated organic pollutants that are harmful to the environment. This causes technical problems such as reducing the cement strength and inducing corrosion of steel bars in reinforced concrete structures [50]. Other than that, due to its larger particle size, alternative solid fuels need longer residence time for the water to be transported to fuel particle surface for drying which might need significant amount of energy to heat and dry the fuel properties [10]. Because of these reasons, pre-treatment and thermal upgrading process are compulsory to overcome the technical problems by enhance its properties to become solid fuel called refuse-derived fuel (RDF). As for the chemical and elemental properties, the minimum criteria that should be complied by RDF to be accepted for co-processing in the cement plant are based on size (<50 mm or < 20 mm depending upon use in In Line Calciner (ILC) and Separate Line Calciner (SLC), respectively), ash (<10 – 15 %), moisture (<10 – 20 %), chlorine (<0.5 – 1 %), sulfur (<1.5 %) [42]. It requires additional cost associated with capital and operation of the MSW for drying and treating before being able to use in cement kiln [51]. Thus, the latter part of this study will discuss RDF production from MSW, characteristics, technologies, limitations, and techno-economics.

3. Refuse-Derived fuel

The term “RDF” refers to a compacted solid fuel that is extracted from the combustible portion of MSW using two separate physical pre-treatment methods: either by separating the waste at source (collection in various bags or bins with various colours that have been separated based on types of waste groups) or separating the waste in plants using mechanical treatment (mechanical separation through crushing, shredding, screening, magnetic separation, eddy current separation, wind separation, and removing most of the biodegradable fractions, metals, and glass) [38]. It is necessary to use pre-treatment techniques to recover as many recyclable materials as possible, lower the moisture content of the wastes, homogenize them, and stabilize their combustibility for simpler handling, transportation, and storage procedures [52].

The waste composition, typology (fluff, dry fluff, and pellet), and technological aspects of the plants are significant contributing elements to the ultimate life cycle performances of RDF, as well as direct emissions of WtE facilities, for effective use of RDF in the cement industry [53]. It is therefore necessary to make sure that a strict internal control procedures for analyzing the specification and characteristic of the RDF, which are complying with the existing global standards so that the minimum quality criterion for the fuel standard is achieved [54]. Seven different forms of RDF have been categorized by the American Society for Testing and Materials (ASTM) International Standard according to their physical characteristics [42,55] as can be referred in **Table S1**. Among all the RDF classed in the ASTM standard, RDF-2 and RDF-3 are typically used. Another study by Punin et al. [56] stated that RDF-1 and RDF-5 are also commonly used, but there are several problems encountered with the use of RDF-1, which makes it less desirable of usage. This is due to difficult handling process of RDF-1, and usually burned in suspension that resulted incomplete of combustion, as well as problems in ash disposal [56]. The typologies of RDF were classified based on original source and/or sizes in the form of fluff (dimensions from 10 to 100 mm), pellets (dimensions < 25 mm), or briquette (dimensions from 40 to 200 mm) [57].

Table 5 summarizes the different types of RDF and its purposes in different countries. Large countries like China and India are developing national standard/guidelines for the domestic wastes pre-treatment for RDF production [58]. This helps to manage large number of wastes, as well as satisfy domestic demand for energy usage. The guidelines will overcome the market barriers that are caused by the lack of common denomination and procedures to establish sufficient quality of RDF. Other than market barriers, some obstacles of using RDF that should be emphasized are such as: (1) lack of waste management policies and its degree of local implementation standards; (2) insufficient supply of high-quality RDF due to infrastructural issues (under development in pre-processing facilities, insufficient waste treatment system and unbalance of the local WtE system), (3) high local bureaucracy, and (4) lack of public acceptance [58].

3.1. RDF composition and characteristics

The quality and composition of RDF are highly influenced by careful consideration of effective integrated technologies and methods used to produce the RDF including waste sorting, recycling, and recovering [38]. The heterogeneity of generated MSW makes energy recovery difficult, hence proper MSW fractionation and sorting associated with MWS pretreatment process are needed before subjected to further energy conversion technology like thermo-chemical process. Generally, MSW sorting and segregating can be done successfully by continuous participation and support from government, private sector, and public to actively practicing waste separation at original sources according to the types/groups of wastes composition. It is also worth noting that waste composition is subjected to other socio-economical factors including people’s waste sorting behaviour, lifestyle, economic status, political regulation, and environmental factor [59]. Other than that, it could be affected by the direct relation between solid waste composition and social activities in community. Thus, the composition of MSW is not consistent and varies from time to time which contributed to its variable physical and chemical characteristics.

RDF compose main fractions of plastic (9.3 – 42.3 wt%), paper or biogenic waste (5.3 – 25.6 wt%), and textiles (2.1 – 18.8 wt%). In addition, RDF containing non-combustible inorganic components is indicated as ash content with ranges around 5.7 wt% to 30.6 wt% [60]. The RDF can be typically categorized by its energy content of about 10 – 25 MJ/kg with a substantial biogenic content (~50 – 65 %) based on an

Table 6
Proximate and ultimate properties of MSW-derived and fossil-based solid fuel produced from various sources.

Fuel	Composition	Operating condition	Proximate analysis (%)				Ultimate analysis (%)					Cl (%)	HHV (MJ/kg)	Ref.
			Moisture	Volatile	Fixed carbon	Ash	C	H	N	S	O			
Fossil-based solid fuels	Anthracite coal	–	1.96	12.81	73.85	13.34	78.70	2.80	1.10	14.90	0.04	0.35	29.90	[70,71]
	Bituminous coal	–	1.50	28.71	65.10	16.24	75.40	4.90	1.40	11.40	6.90	1.15	31.20	[70,71]
	Lignite coal	–	2.46	36.96	53.55	19.42	68.40	4.40	1.20	6.70	19.30	1.10	24.40	[70,71]
	Petcoke	–	–	9.83	88.63	1.54	80.50	3.60	2.50	13.36	0.04	–	33.30	[70,71]
Waste-derived solid fuel	Meat and bone meal	200—–1000 °C	5.50	54.08	15.33	25.09	57.83	9.81	8.71	0.30	23.13	0.22	19.18	[72]
	Sewage sludge	150—–400 °C, 0 – 50 min	82.20	15.20	–	2.60	46.93	6.83	7.40	0.54	23.23	0.07	19.86	[73]
	Pyrolyzed black carbon (tyre-derived solid fuel)	25—–550 °C, 0 – 50 min	1.16	2.50	79.79	16.55	95.42	0.77	0.22	3.29	0.12	0.19	28.70	[74]
	RDF from MSW (50 % paper and fibre, 28 % wood, 9 % plastic, 7 % food waste, 6 % incombustible)	500—–900 °C	–	76.70	10.40	12.90	43.50	6.40	1.10	0.10	48.90	0.40	17.90	[75]
	RDF from MSW (gardens, trees, grass, timber waste at landfills)	150—–500 °C	0.48	12.20	32.90	54.90	36.60	1.60	0.74	0.15	60.00	0.18	13.70	[76]
	Wood (Acacia nilotica)	220—–280 °C, 40 min	6.18	81.77	11.35	0.69	43.84	7.88	0.42	–	47.86	–	19.31	[77]
	Mixed plastic waste (from reject fraction of MSW in Granada, Spain)	450—–550 °C, 90 min	0.61 – 0.84	51.00 – 24.90	13.35 – 6.40	35.04 – 67.87	55.24 – 17.82	5.74 – 0.55	0.46 – 0.24	–	3.31 – 12.95	0.60 – 11.57	23.94 – 3.69	[78]
	Agricultural wastes (from corn fields)	180—–260 °C, 60 min	–	78.60	18.42	2.98	46.24	6.52	0.58	0.38	43.30	–	18.13	[79]

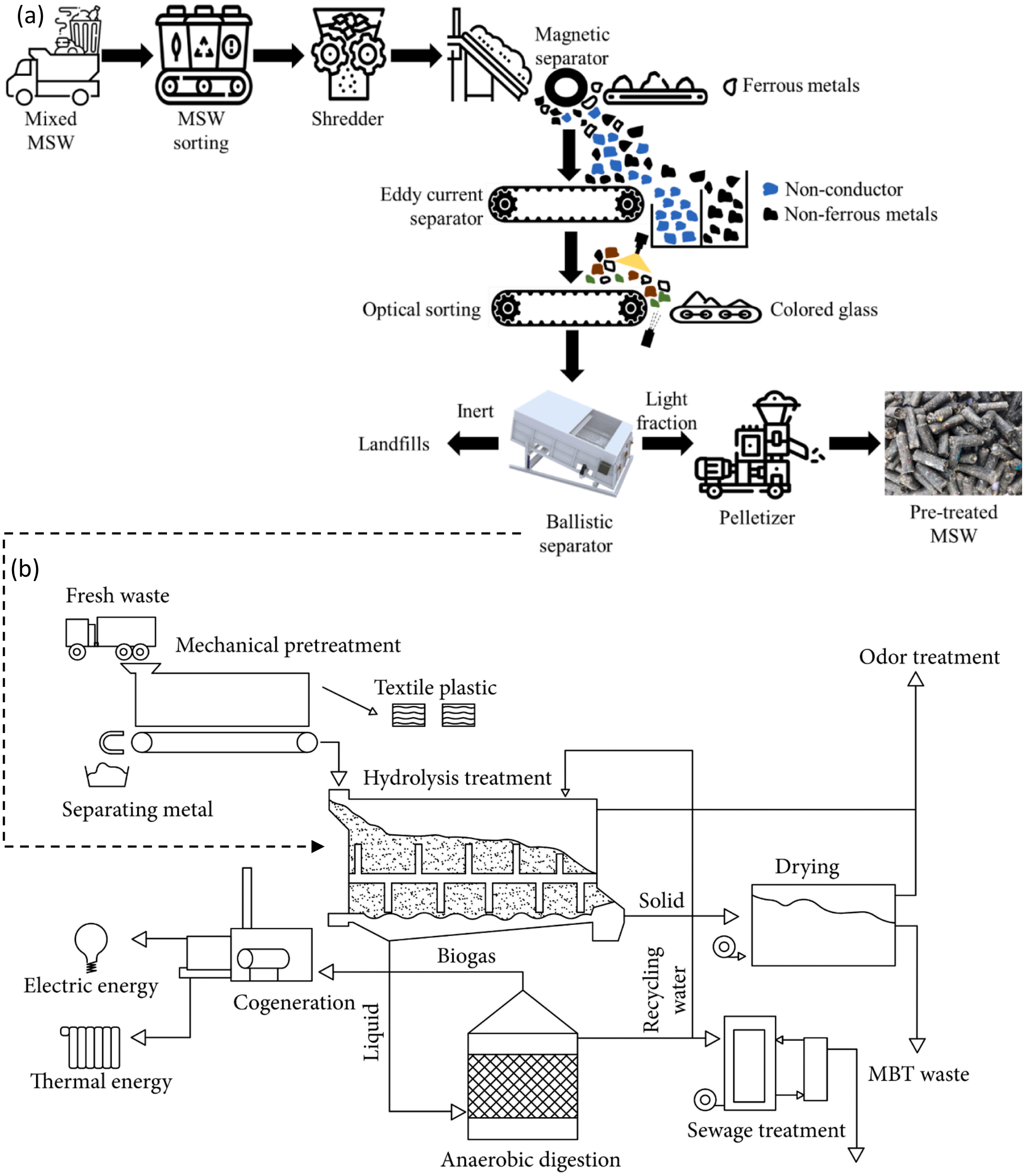

Fig. 4. (a) Mechanical pretreatment; (b) Mechanical-Biological pretreatment of MSW. The figure is redrawn from Ref. [65].

energy basis. There are two types of RDF that can be employed in cement industry: (1) RDF for secondary firing with calorific value ranged 12 – 18 MJ/kg, and (2) RDF for primary firing with calorific value ranged 18–25 MJ/kg [60].

The characteristics of RDF components are determined by proximate analysis and ultimate analysis which are two important parameters to select best alternative management and consideration as potential feedstock for energy recovery systems [43]. Table 6 shows the physical and chemical properties of several alternative solid fuels which include MSW-based RDF and fossil-based solid fuels to reflect the diversity of these fuels' properties. These properties deviate based on different factors, such as the waste's origin, prevailing weather conditions, preprocessing methods, operating parameters, composition, and storage conditions [10].

The proximate analysis studies the moisture, volatile matter, ash, and fixed carbon. High moisture content is usually prohibitive for combustion process as it will reduce the energy recovery of the fuels, which resulted low thermal efficiency and low quality of gas emission during combustion process [61]. Moisture content $<$ 15 wt% is favorable for cost-effective operation in the energy recovery plants [62]. The main challenge of using local MSW (especially food waste) as RDF is that it has a high moisture, various composition of pollutants, and salt content, which making conversion process a slightly challenging task [63]. In addition, untreated wet waste can become a problem with steam production in terms of heat and pressure fluctuations [64]. Volatile matter of RDF is released in the form of vapor consisting of water, tar, oil, and gas when the material is heated [65]. After combustion is completed, an incombustible inorganic solid residue that remains in oxidized form represent as ash content [66]. Ash range between 0.49 and 12.58 % has the possibility of having small quantity of salts, heavy metals, chloride, and organic pollutant which is an advantage to waste management and environment [61]. The ash content and ash composition can affect the performance of combustor, as ashes will undergo agglomeration and melting during high heating temperature, which causes slagging and clinker formation in combustor system. Generally, no slagging will happen when ash content is below 5 % [62]. Final stage in the proximate analysis study is the determination of fixed carbon [65].

On the other hand, ultimate analysis specifies the major elemental composition and concentration of heavy metal properties of the solid waste. The percentage of H and C usually influence the HHV of the samples while S and N affect the corrosiveness of the equipment and gas emissions due to the combustion products of sulphate and nitrate are SO_x and NO_x, respectively [67]. During combustion process ($\sim$1000 °C), oxidation occurs to oxidize carbon into carbon dioxide (CO_2), hydrogen into water (H_2O), nitrogen into nitrogen gas/oxides (N_2/NO_x) and sulfur into sulfur dioxide (SO_2). For oxygen determination, the elemental analyzer operates by pyrolysis mode in a second run [68]. The ultimate profile can be used as indicator of material balance, as well as calculating calorific value of the fuels based on established equation [69].

RDF quality and grade are based on key parameters like combustibility, calorific value, moisture content, ash content, sulphate, chloride contents, and metal contents (Hg, Cd, Pb, and As) [80]. The properties of RDF from different countries like United Kingdom [81], Korea [82], India [83], Indonesia [84], Pakistan [85], Malaysia [86] are compared with coal as summarized in **Table S2**. From this table, the common problem with RDF is regarding the inconsistent calorific value (about 16.04 to 30.03 MJ/kg) and high moisture content (about 15 to 26 %) due to the different composition of MSW [36]. High moisture content in MSW is not desired as it contributes to low energy density, an increase in operating cost of transport and energy usage for drying process [87]. In contrast to coal, RDF contains higher ash content (about 10 to 25 %), as well as lower carbon content (40 to 50 %) which shows that RDF needs further upgrading process to improve its performance. However, from the elementary analysis result, RDF contains a lower concentration of sulphate and chloride than coal. This is beneficial for the environment as lower sulphate content can reduce the effect of acidification due to lower SO_2 emissions during RDF co-combustion and lower chloride content can reduce the generation of chlorinated organic pollutants to the environment which may reduce the cause of technical problems such as reduce the cement strength and induce corrosion of steel bars in reinforced concrete structures [50]. Although RDF shows a lower calorific value than coal which can significantly reduce electricity yield (or power or heat) supplied to cement kilns, RDF still shows the possibility for co-processing fuel as it is still above RDF standard criteria.

3.2. *Conversion technologies of MSW to RDF*

3.2.1. *MSW pre-treatment Processes: Mechanical treatment (MT) and Mechanical-Biological treatment (MBT)*

The pretreatment process of MSW includes the approach of mechanical treatment (MT) or mechanical–biological treatment (MBT) (Fig. 4). This pretreatment stage aims to enhance the capacity of energy recovery and optimize the type of waste that can be potentially upcycled into energy. In MT plants, the procedures involve mechanical sorting operations, size reduction (shredding techniques with different particle sizes, milling, chipping, crushing), separation and screening (magnetic separation (removal of Fe-metals), eddy-current separation (removal of non-Fe-metals)), sieving steps, blending, drying, and pelletizing [38].

It should be emphasized that MT method will be more efficient, cost-effective, and technologically feasible if MSW is readily separated at source. When MSW is not sorted beforehand, additional production line that consist of numerous stations and laborers is necessary to separate the undesirable waste components. Following the MT process, the remaining combustible materials like wastepaper, waste plastic, and waste cardboard are used for energy recovery, which can be incinerated directly, co-incinerated in the power plant or pelletized for subsequent upgrading process [50].

On the other hand, MBT plant involves two integrated mechanical (M) processing and biological treatment (BT) reactors. By integrating biological stabilization technology into the mechanical treatment platform, it becomes possible to employ bacterial-driven biological

Table 7
MT and MBT studies of various types of MSW for pretreatment stage.

Raw material with MT and MBT pretreatment stages	Findings	Limitation	Refs.
Sludge sewage, sawdust, and wheat straw MT: $\sim$15 mm for sawdust; $\sim$10 cm for wheat straw pieces MBT: Stainless steel reactor	1. Highest CV was 13.07 MJ/kg High air flow rates improved moisture removal and decelerated bio-degradation. Sawdust provided better fuel quality due to higher bulk density. C and N levels were decreased throughout the bio-drying process.	1. Decrease of carbon values due to rapid consumption of volatile organic compounds (VOC) during bio-drying. Maximum of bulking agent 20 % to protect energy content of the product.	[91]
Food, paper, and garden waste MT: n.a MBT: n.a.	1. Bio-drying increased the CV of food waste in the pelletizing process. CV of RDF pellets was increased when composition of garden waste was increased. CV of RDF was decreased in the presence of high moisture of food scraps (51 %).	1. Density and hardness did not have any impact on the CV relationship.	[96]
Plastic, glass, metals, mineral and biodegradable paper, as well as textiles MT: 0 – 80 mm MBT: Full-scale bio-drying industrial reactor	1. Temperature (55 °C) in bio-drying is crucial to reduce loss of carbon. Decreased in moisture (by 50 %) will enhance CV (by 45 %).	1. Carbon content expressed as the loss of ignition (LOI) decreased in waste to 66 %. LOI decrease was not favorable of using MSW as RDF.	[97]
120 tonnes of fresh MSW MT: < 150 mm and < 15 mm MBT: Conveyor system	1. CV is increased by almost 600 % from 2.97 MJ/kg to 14.55 MJ/kg. CO_2 emissions was avoided by not more than 65.8 tonnes/day,. GHG emission tax reduced to 0.35 USD/day and coal saving was about 70 USD/day.	1. Requires large investment.	[98]
33 % food waste, 23.3 % organic compost, 17.4 % glass beads, 11.7 % paper, 7 % plastics, 5.4 % wood chips and 2.2 % rice straw MT: 50 mm, followed by the addition of distilled water MBT: Stainless steel reactor	1. CV was increased by 3.6 – 11.4. High moisture level led to more carbon being used during the bio-drying process to evaporate the water, which produced more leachate.	1. Bio-drying during the addition of water could not be accomplished.	[99]
Compost-Like-Output (CLO) (biologically stabilized organic fraction): mixture	1. RDF rendered CV of 16.70 MJ/kg. Moisture content of the tested waste was < 20 %.	n.a.	[100]

(*continued on next page*)

Table 7 (*continued*)

Raw material with MT and MBT pretreatment stages	Findings	Limitation	Refs.
of organic, paper and plastic materials MT: 10 – 35 mm and 35 – 80 mm MBT: Bio-reactor	Total recovery of organic, plastics and paper were ranging from 70.2 % to 95 %.		
42.53 % organic waste, 27.33 % plastics, 13.60 % papers, 3.07 % rubbers, 1.87 % metals, 0.53 % clothes, 0.87 % cement and ceramic, 2.87 % wood, 4.93 % glasses, 2.40 % others. MT: n.a. MBT: Lab-scale lysimeter	1. Inconsistent bio-drying of MSW due to condensation of moist vapor. MSW became dried by bio-drying process became dried within 5th – 7th day.	n.a.	[94]
About 50 tonnes of mixed MSW MT: < 100 mm and < 150 mm MBT: n.a.	1. Increased in heating value by 58 % with 15.58 MJ/kg. Bio-drying process resulted in increased heating value because of waste moisture reduction. RDF produced had high calorific value, low water content and satisfactory chloride content.	n.a.	[92]
32 % kitchen waste, 26 % agriculture waste, 12 % paper, 16 % plastics and 14 % others. MT: < 76.2 mm MBT: Laboratory horizontal-column reactor	1. Moisture content decreased from 63 % to 13 % within period of 8 days. Slow rotation and preheating provided adequate conditions for effective bio-degradation by increasing degradation temperature. HHV is increased from 4.89 MJ/kg to 19.19 MJ/kg within period of 8 days.	1. Convection heating process and moisture content reduction were slower.	[93]

Abbreviation: n.a. is not available; CV is calorific value; HHV is high heating value; LHV is low heating value.

treatment for the processing of MSW residue that left-over from MT treatment, as well as MSW containing substantial quantities of organic (biodegradable) elements. This process can generate valuable of organic soil conditioner or biogas (CH_4) as outcomes [88]. The mechanical process is similar to MT system, while additional biological treatment involves bio-drying, bio-stabilization, composting, anaerobic digestion [89].

The bio-drying process reduces the moisture content of MSW through the heat generated by forced aeration during microbial metabolism, along with a reduction in odor through biological stabilization [90]. Additionally, this process enhances the calorific value of the treated MSW, resulting in a thermally dried and highly mineralized organic material [38]Bio-drying process shares similarities with composting treatment, but unlike composting, the treated product is partially stabilized to preserve the organic content of the feedstock [91]. It requires appropriate degree of temperature and air flow rates to optimize moisture removal to produce high energy density of RDF [91,92]. According to Patcharavongsiri et al. [93], increased of rotation speed resulted rapid decrease in moisture content due to high rate of evaporation. Consequently, the moisture content decreased from 63 % to 13 % within 8 days. On the other hand, slower rotation and preheating created favorable conditions for effective biodegradation by raising the degradation temperature. In addition, Sutthiprapa et al. [94] reported that GHG emissions (CH_4, CO_2, N_2O) were lowered at the aeration rate of 0.2 L/min as compared aeration rate of 0.5 L/min. The moisture content was reduced significantly by 40.6 – 94.5 % in treated wastes after 14 days.

However, the biological treatment process has common challenges, which require long treatment period for more than 1 week to months, along with the presence obnoxious odors [95]. This treatment raises system complexity such as, adding inherent system losses and induce additional energy consumption [57]. In addition, RDF presents several challenges such as high moisture levels, large granulometry, substantial of ash content, and low calorific value when compared to fossil-based coal. These limitations prevent its effectiveness usage for cement co-processing due to inadequate quality. Therefore, it becomes essential to enhance the physico-chemical properties of MSW precursors through pretreatment process, prior for RDF production through the thermo-chemical conversion process. Table 7 summarizes the MT and MBT studies of various MSW for pretreatment stage.

3.2.2. *Thermo-chemical conversion technologies for RDF production*

Generally, Waste-to-Energy recovery technologies can be categorized into three main pathways (bio-chemical, physico-chemical, and thermo-chemical) as shown in Fig. 5. These pathways are determined by the physico-chemical characteristics of the raw MSW compounds, as well as the intended usage of the final product [101,102]. Pyrolysis is one of the thermo-chemical technologies that can convert waste to energy which includes dry pyrolysis at atmospheric pressure such as torrefaction (mild pyrolysis, 200–300 °C) and carbonization (moderate pyrolysis, 300–650 °C), as well as wet pyrolysis such as hydrothermal carbonization at pressures ranging from 10 to 80 bars (hydrous pyrolysis, 180–280 °C). [103].

These thermal conversion technologies exhibit variations in their operating conditions, including temperatures, heating rate, and oxygen supply. These variations result in distinct ranges of product distributions, such as heat and power, syngas, bio-oil, and bio-char. Additionally, each conversion process has a different gas emission profile. Torrefaction is thermal pretreatment process that operates at lower temperature ranged 200 – 300 °C with longer treatment time, where the majority of product yielded is bio-char/bio-coal/green coal (used as fuel or soil amendment) [104,105].

As for pyrolysis, this process is a carbonization process that works under temperatures 300 – 650 °C, higher than torrefaction where this process produces a different range of products depending on its heating rate and temperature. The pyrolysis will produce a majority of liquid products (e.g., bio-oil and condensable gases) under a fast-heating rate within a short residence time, while solid char is produced under a slow heating rate with longer residence time, and syngas yielded under flash pyrolysis with high heating rate [104]. Another type of pyrolysis is called hydrous pyrolysis (hydrothermal carbonization) and the thermal/heating treatment is done in hot compressed water with elevated pressure. Although bio-oil is the main product of pyrolysis, however, direct usage of the bio-oil is restricted due to its high acidity, water, and oxygen (O) content, which need further upgrading process before use [106].

The partial oxidation process of gasification, on the other hand, entails more complicated procedures, starting with the conversion of wastes into a synthesis gas (specifically, syngas, a combustible gas mixture rich in carbon monoxide (CO) and hydrogen gas (H2)) at a

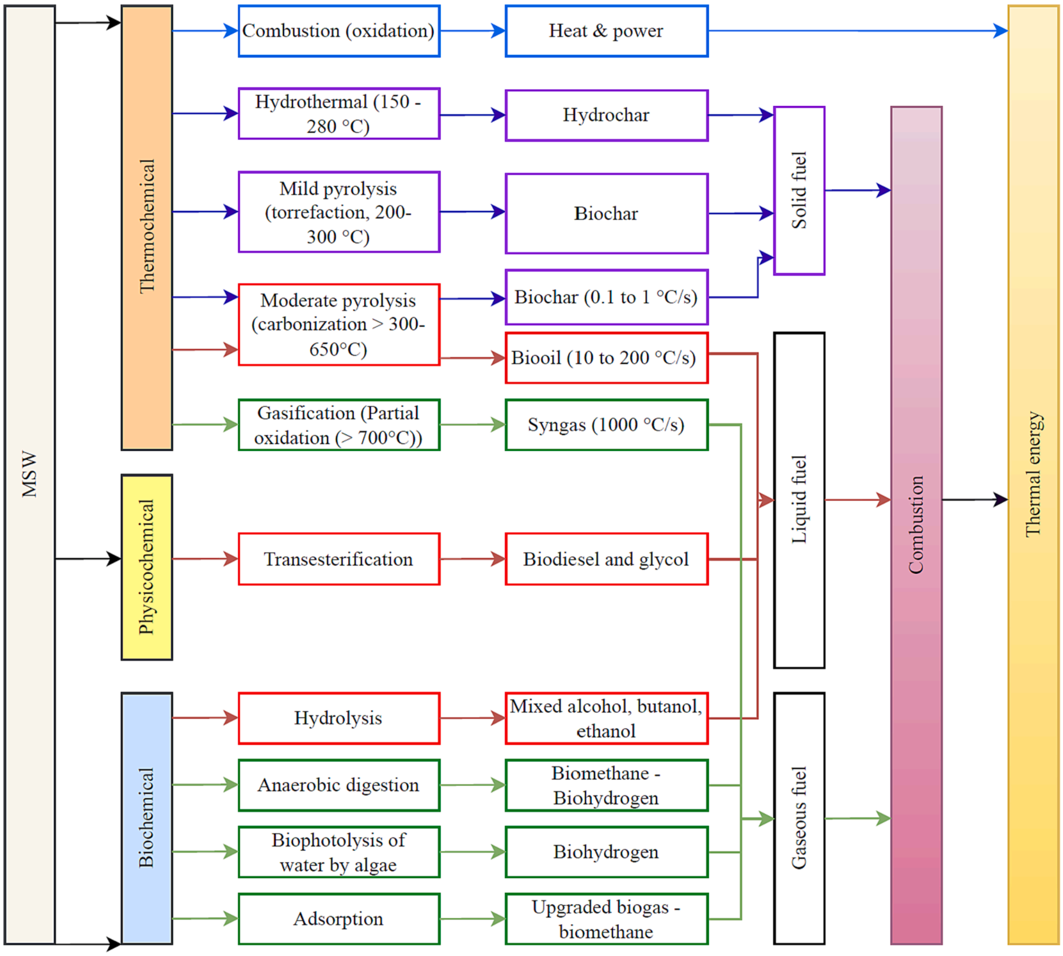

Fig. 5. The pathways of Waste-to-Energy technologies and their products. The figure is drawn based on information from Ref. [101,102], with permission.

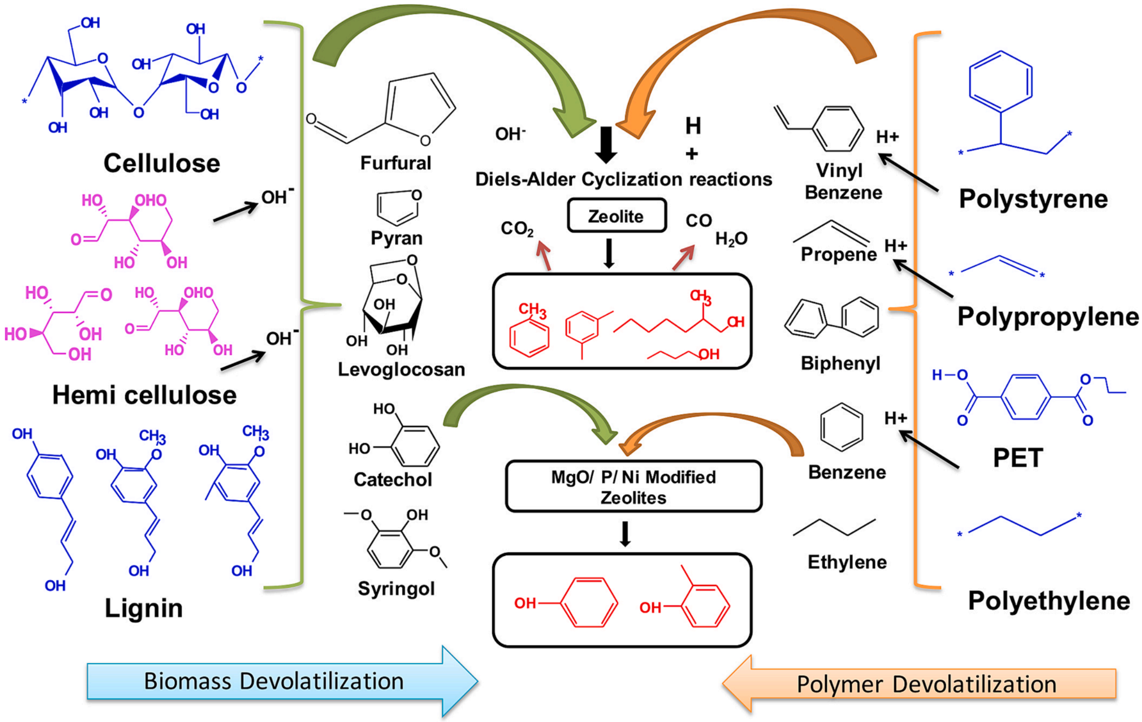

Fig. 6. Devolatilization mechanism between plastics and biomass. From Ref. [48], with permission.

temperature above 700 °C. The resulting syngas was then further transformed into pure H2, which could be used as a clean fuel gas in a standard burner or connected to a boiler or steam turbine [107,108]. In the reactor, contaminants like tar, alkaline compounds, halogens, and heavy metal agglomeration are concentrated at high levels, which is the fundamental disadvantage of gasification for downstream applications [109]. Since converting MSW into RDF solid fuel is the main emphasis of this study, the study does not go into great length to explain the various gasification conversion technologies.

3.2.2.1. Dry pyrolysis (Torrefaction and Carbonization). Torrefaction is referred to as mild temperature pyrolysis, occurring at temperatures between 200 and 300 °C in an inert atmosphere with the use of nitrogen or carbon dioxide as carrier gases to regulate the oxidizing conditions inside the reactor and prevent combustion [110]. Torrefaction involves the overall mechanism of free moisture evaporation (dehydration), followed by primary devolatilization, and secondary reactions (oil cracking and repolymerization) [103,104]. Dehydration reaction changes the torrefied material structure which increases the hydrophobic character, which reduces hydrogen (H) bond formation [111]. This results in the enhancement of reactivity, stability, and grindability due to its hydrophobic solid product and improved resistivity against water (shields against composting) [112]. Besides, when polymers and biomass are torrefied together, the plastic monomer and primary alcohol combine. This causes the polymers to break into monomers, resulting in the devolatilization of biomass which produces OH^{-1} while H^{+} is donated by polymers to the biomass molecules [48]. By removing oxygen from the solid fuel, the interaction of the components as depicted in Fig. 6 will improve the quality and performance of the RDF. RDF production is strongly influenced by key factors such temperature, residence time, and feedstock properties. Torrefaction reaction kinetics depend on temperature, while process features depend on time.

Like torrefaction, the mechanism of carbonization (moderate pyrolysis) also works similarly but under moderate operating temperature at about 300 – 600 °C. In between this temperature range, product yield is highly dependent on its heating rate (slow, fast, or flash) and residence time. Higher yield of liquid products (e.g., biooil and biogas) is produced under fast heating rate (10 and 200 °C/s) within short residence time (0.50 – 10 s), while higher fraction of solid char is produced under slow heating rate (0.1 to 1 °C/s) with longer residence time (450 – 550 s), and higher syngas is produced under flash (very high) heating rate (1000 °C/s) and very low residence (<0.5 s) time [78]. In comparison with torrefaction, solid yield is much lower when temperature increases above 300 °C as most hemicellulose, cellulose, lignin, and polymers start to decompose at this temperature, causing devolatilization and decomposition mechanism to occur actively. In the case of feedstock containing high lignin, pyrolysis at moderate temperature about 500 °C can generate higher solid char yield [88].

It is known that both dry pyrolysis like torrefaction and carbonization can successfully enhance the solid fuel properties, depending on the type of raw material and operating conditions used during the conversion process. In general, after torrefaction and carbonization, lower moisture content and oxygen-to-carbon (O/C) ratios are obtained, resulting in higher high heating value (HHV) and carbon content [104,105]. The HHV of the remaining material slowly increases when the light volatile fraction is removed, hence high thermal efficiency can be achieved as the energy contained in the released volatiles is equal to the heating required in the process [114]. However, torrefaction of high mineral, salt, and protein content from food (organic) waste will result in high ash deposition, chlorine (Cl), nitrogen (N), and sulfur (S) [115]. Also, Wiedner et. al. [116] indicated that high number of polysaccharides are conserved at lower temperatures below 300 °C and do not completely degrade, causing the properties of solid fuel to remain almost like its original feedstock.

E. H. X. Lim et al. [90] carried out the torrefaction of empty fruit bunch (EFB) at mild temperature 200, 250, and 300 °C for 30 to 90 min using a fixed-bed reactor where the authors found an increased in HHV by 40.80 % and energy yield by 90.10 % as temperature increased, with reduced of S from 0.18 to 0.08 %. Besides, the authors also found an increased in N from 0.53 to 0.74 % and ash from 3.50 to 6.90 % after torrefied at 300 °C. Abdul Samad et al. [91] studied the effects of temperatures in the range of 240 to 300 °C for residence time 30 min on food waste and wood waste using a vertical tubular reactor. It was discovered that after torrefaction from 240 to 330 °C, Hydrogen-to-Carbon (H/C) and Oxygen-to-Carbon (O/C) ratios were decreased, and carbon was increased, resulting in HHV increased for both food and wood waste from 17.45 to 28.42 MJ/kg and 19.27 to 31.09 MJ/kg, respectively. Despite this, it was discovered that N, S, and Cl were slightly elevated and greater in food waste than in wood waste due to the presence of salt and protein, coupled with elevated ash levels of 16.89 to 25.16 % and 7.37 to 18.90 %, respectively. In a tubular furnace, Pahla et al [92] looked into the torrefaction behavior of landfill food waste at mild temperatures between 200 and 300 °C. The authors came to the conclusion that torrefaction was more efficient between 240 and 290 °C, producing a more hydrophobic torrefied food waste with high energy density, reduced O/C and H/C, and enhanced HHV. However, the authors did observe that N was higher than coal and that it rose with increased temperature. The potential of olive pomace as a solid biofuel was studied by Gultekin et al. [93]. The authors found that as the response time and temperature increased, carbon was also increased and oxygen was decreased, resulting in enhanced HHV from 26.40 to 29.50 MJ/kg and decreased H/C and O/C ratios. The investigators also found that as the temperature rose, the biochar ash increased accordingly from 1.61 to 20.46 % because of an increase in inorganic elements during torrefaction and an increase in N.

At moderate pyrolysis temperature, Ren's group reported on high N content (7.3 – 12.5 %) with major chlorinated species (e.g., CH_3Cl and HCl) when solid fuel derived from biomass was carbonized at 350 to 400 °C [121]. Anand et al. [122] who investigated pyrolysis of wood and rice residues using batch-type vertical retort reactor at temperatures 400 – 1000 °C observed highest biochar yields were at moderate temperature, 400 °C, ranging from 32.00, 27.69, 49.00, and 45.00 % for wood chips char, wood dust char, rice straw char, and rice husk char, respectively. On the other hand, HHV was found to be the highest at 1000 °C, ranging from 23.90, 24.05, 18.25, and 18.60 MJ/kg for wood chips char, wood dust char, rice straw char, and rice husk char, respectively. Ercan et. al. [123] made similar observations when studying on biochars produced from hornbeam wood using stainless steel fixed-bed reactor where solid yield decreased significantly from 67.20 to 27.70 % while HHV increased from 19.00 to 32.50 MJ/kg when temperature increased from mild (275 °C) to moderate (575 °C). Interestingly, Selvarajoo et. al. [124] found an increase in HHV of dried citrus peels char after pyrolyzed using tubular furnace from 24.01 to 25.73 MJ/kg at moderate temperature 300 to 500 °C but the value decreased at higher temperatures from 25.73 to 24.42 MJ/kg at 600 – 700 °C. The authors also found an increase in ash content from 0.11 % to ~ 3.52 % when temperature increased from 300 to 600 °C and reached highest value of 7.97 % at 700 °C, which could be attributed to the continuous concentration of mineral constituents. On the opposite, Chen et. al. [125] who examined moderate pyrolysis of sewage sludge at temperature 300 to 500 °C using quartz tube found a constant decrease in HHV from 12.52 to 8.21 MJ/kg, which may be due to the increased amount of ash in biochar because organic matter presence in raw sludge. However, the authors reported that N-containing proteins in sewage sludge being transformed to amine-N compounds, which were then followed by heterocyclic-N compounds, and subsequently released of NH_3 at moderate temperatures 300 – 550 °C have caused a decrease in N from 5.11 to 2.77 %. They also observed a reduction in S concentration from 1.29 to 0.59 %, which could be attributed to the breakdown of aliphatic sulfur and weak mercaptan compounds that occurred at temperatures below 500 °C. Additionally, Nobre et. al. [126] who converted non-hazardous

Table 8
Dry pyrolysis (torrefaction and carbonization) studies of various MSW to RDF.

Feedstock	Operating conditions	Reactor	Solid yield (%)	Energy value (MJ/kg)	Ref.
Torrefaction (Mild temperature pyrolysis)					
Food, garden, and wood waste	200 – 280 °C, 60 min, 0—40 rpm	Fixed helical screw induced rotation	97.95 – 52.91	17.25 – 26.69	[127]
Empty fruit bunch (catalyst: eggshell)	200 – 300 °C, 30 – 90 min	Fixed-bed tubular	87.50 – 44.90	18.02 – 23.42	[117]
Lignocellulosic MSW	225 – 275 °C, 60 min	TGA-2000A	93.55 – 77.86	19.32 – 21.47	[128]
Melina wood	240 – 300 °C, 30 – 60 min	Tubular	81.18 – 38.07	20.75 – 29.11	[129]
Landfill food waste	220 – 260 °C, 15 – 60 min	Tubular	94.50 – 66.95	19.15 – 23.90	[130]
Corn residue pellets	260 °C, 10 – 40 min	Fixed-bed tubular	70.00	23.00	[131]
Landfill food waste	200 – 300 °C, 40 min	Tubular	89.50 – ≈ 50.00	19.76 – 26.15	[119]
Olive pomace	250 – 350 °C, 10 – 30 min	Pilot-scale continuous screw	87.85 – 29.24	20.34 – 29.41	[120]
Bamboo	180 – 300 °C for 10 min	Ceramic	100.00 – 43.70	19.00 – 30.00	[132]
Japanese cedar			100.00 – ≈ 50.00	19.20 – 29.90	
Food waste	240 – 330 °C, 30 min	Vertical tubular	81.00 – 48.00	17.45 – 28.42	[118]
Wood waste			77.00 – 46.00	19.27 – 31.09	
Carbonization (Moderate temperature pyrolysis)					
Wood chips	400 – 1000 °C, 0.17 °C/s, 1 h	Batch-type vertical retort	32.00 – 21.43	21.82 – 23.80	[122]
Wood dust			27.69 – 23.00	21.25 – 24.05	
Rice straw			49.00 – 28.80	16.61 – 18.25	
Rice husk			45.00 – 34.86	16.06 – 18.60	
Hornbeam wood (sawdust)	375 – 575 °C, 0.08 °C/s, 1 h	Stainless steel fixed-bed	67.20 – 27.70	19.00 – 32.50	[123]
Sugarcane bagasse	450 – 650 °C, 0.17 °C/s, 30 min	Laboratory-scale fixed bed	26.04 – 21.75	27.93 – 29.99	[133]
Tomatoes	800 °C, 0.17 °C/s	TGA 550	39.81	17.64	[134]
Cucumbers			39.02	20.21	
Carrots			41.43	12.21	
Citrus peel fruit	300 – 700 °C, 0.08 °C/s, 1 h	Tubular	53.62 – 22.01	15.02 – 24.42	[124]
Sewage sludge	300 – 500 °C, 0.17 °C/s, 30 h	Quartz reactor	≈ 65.00 – 51.50	12.52 – 8.21	[125]
Black alder cone-like flowers	400 – 800 °C, 0.17 °C/s, 1 h	Horizontal quartz tube	32.00	25.20 – 27.00	[135]
Black alder waste wood chips			24.00	26.30 – 29.70	
Palm fibre	300 – 900 °C, 0.08 °C/s, 2 h	Electric tubular	54.83 – 26.67	16.61 – 25.61	[136]
Non-hazardous industrial wastes	300 – 400 °C, 30 min	Industry rotary	82.00 – 62.90	16.40 – 23.50	[126]
Algal wastes	350 – 600 °C, 0.17 °C/s, 20 mins	Stainless steel fixed-bed	33.00 – 26.50	14.98 – 23.09	[137]

industrial wastes using industry rotary at moderate temperature 300 and 400 °C noticed an increase in HHV from 16.40 to 23.50 MJ/kg. It was mentioned in their paper that as opposed to non-polar C–C bonds, which were stable up to 400 °C, N and S were partly removed as their polar covalent interactions with carbon atoms were more easily broken. The authors also spotted an increase in chlorine from 0.70 to 1.90 %, which could be ascribed to chlorine recondensation on solid fuel surface or chlorine retained onto the organic and mineral phases, as well as the potential generation of inorganic salts featuring potassium and chlorine chloride. Intriguingly, they found that washing with water treatments can reduce chlorine content to 0.54 %, resulting in a more consolidated structure with higher porosity which was beneficial for chlorine adsorption. Table 8 summarizes the mild and moderate pyrolysis studies on the type of MSW used as feedstock, operating conditions, and findings.

Torrefaction technology uses diverse types of reactors for the development of torrefaction studies within three dissimilar stages which are pilot, commercial, and laboratory. Laboratory stage reactors can only be used for smaller particles and size to investigate the effects of parameters, whereas pilot and commercial stage reactors are capable of processing larger particles. There are six most important torrefaction technologies, such as rotary drum reactor, screw reactor, multiple hearth reactor, fluidized bed reactor, microwave reactor, and moving- or fixed- bed reactor [138]. Most moderate pyrolysis also uses fluidized bed reactors (bubbling and circulating) reactors. Other than that, were fixed bed (100 – 500 °C), vacuum (500 °C), ablative (500—650 °C), auger (500° C), conical spouted (450 – 500 °C), and rotary kiln (500 °C) [113].

The following description of each torrefaction reactor technology is based on an overview of the state of biomass torrefaction commercialization [139]. A rotary drum reactor (proven technology) operates constantly, where in the spinning drum, biomass particles are thoroughly mixed and can be either directly or through indirect heating utilizing steam that is superheated or exhaust gas generated from the inhalation of volatile substances. A screw reactor (proven technology) is also a continuously operating reactor by employing one or more auger screws that move biomass through the reactor regardless of vertical or horizontal motion. It frequently gets heated in an indirect manner by a medium located within the hollow screw; however, the disadvantage of this mode of heating is the likelihood of char formation on the hot zones, which causes agglomeration. A multiple hearth reactor (proven technology for industrial applications) is another continuous reactor that consists of numerous layers with heat provided directly to each reactor layer by internal gas burners or steam injection. In a reactor with multiple hearths, biomass is mechanically driven toward the inside of the reactor from the topmost side, then falls on the second plate, where it is mechanically thrown to the outside and falls through another hole. This process is repeated over a number of layers, leading to uniform blending and progressive heating. This technology can process a broader range of

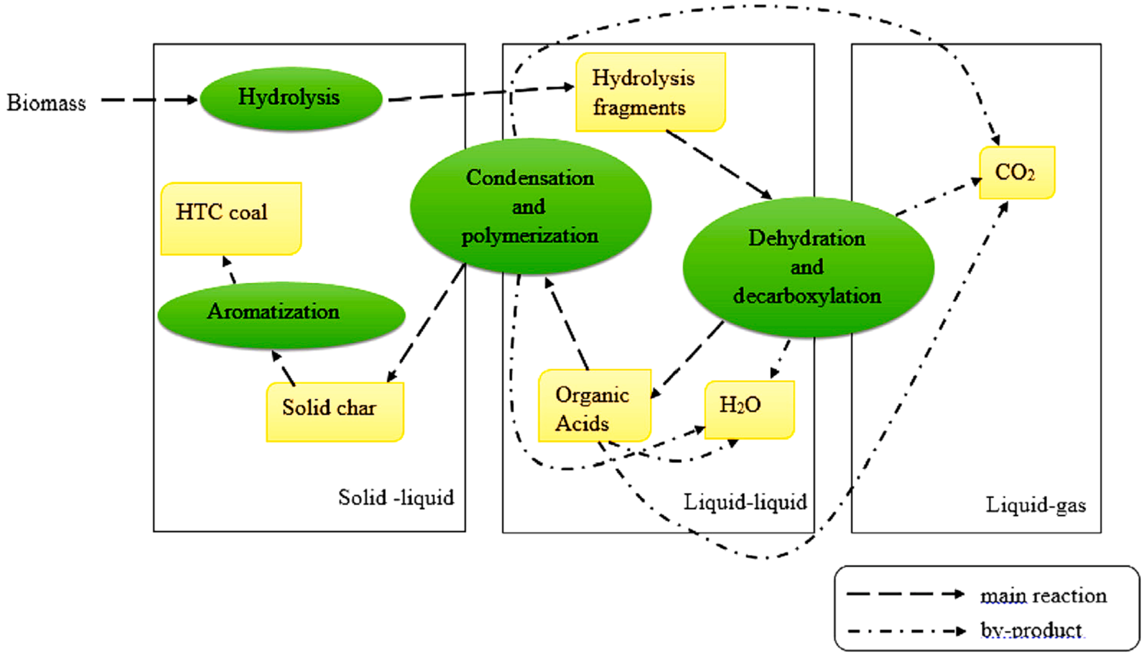

Fig. 7. HTC mechanisms. From Ref. [143], with permission.

feedstock sizes, from sawdust to bigger chips and waste. In general, a fluidized bed reactor features intensive contact between solid and gas phases, resulting in a high heat transfer rate between the two phases. There are two types of fluidized bed reactors: bubbling bed reactors and toroid or torbed reactors. In a bubbling bed reactor, the solid phase is gently fluidized by gases that enter the lower half of the bed, resulting in a dense fluid of solids. Furthermore, in a toroid or torbed reactor, heat is blown at high velocity from the bottom of the bed, passing through stationary and angled blades that cause biomass particles to move in both vertical and horizontal directions, resulting in toroid swirls that rapidly heat the biomass particles and enable torrefaction with short residence times of around 80 s. Microwave reactor technique employs microwave radiation to heat biomass, which produces homogeneous heating from the inside of the material; nevertheless, the microwave technology requires a considerable amount of electricity, which is its main downside. A fixed-bed reactor is the most basic reactor, often employed on a laboratory size, in which biomass is retained inside the reactor and heated by heat conduction from a heater located around the reactor surface. Torrefaction in a moving-bed reactor, on the other hand, occurs as a result of a heat-carrying gaseous medium that enters at the bottom and moves to the top of the reactor. Furthermore, vacuum reactors use lower heat transfer rates and a longer residence time to generate bigger particles than most quick pyrolysis reactors. Ablative reactors, on the other hand, have good heat transmission with a greater heating rate and a smaller contact surface [113]. The auger reactor differs from fluidized bed reactors in that heat is transferred through a molten layer at the hot reactor surface rather than a fluidizing gas [140].

3.2.2.2. Hydrous pyrolysis (Hydrothermal carbonization (HTC)). Due to the high moisture content in MSW, hydrous pyrolysis is a more feasible thermo-chemical treatment that can convert lignocellulosic and polymeric materials into carbon-rich solid fuel called hydrochar. Hydrous pyrolysis, also called as hydrothermal carbonization (HTC) is operated under sub-critical condition at different temperature, pressure, and water content [141]. In contrast to dry pyrolysis, HTC process allows carbonization of feedstock in hot compressed water as green reaction medium. The operating temperature is lower than dry pyrolysis, about 180 – 280 °C. HTC operates in a pressure vessel for 5 to 240 min [104], under elevated pressures with saturation vapor pressure that is corresponding to the reaction temperature. In wet HTC treatments, the individual organic waste components like glucose, sucrose, starch, cellulose, xylan, lignin, and furfural will be decomposed which makes it easy to filter from the reaction solution [142,143]. A study by Mu'min et al. [144] proves that hydrothermal carbonization at low temperatures of 120 °C and 155 °C are sufficient to separate organic and plastic components of mixed waste which produce organic part and plastic part where plastic can be used as fuel with relatively high calorific value (~44 MJ/kg). The water molecules take role as organic solvent, where the water generated ions (H^+ and OH^-) during sub-critical condition that acts as acids or bases catalyst to lower the activation energy levels required for bond cleavage [87]. This favors ion chemistry and suppresses the free radical reactions.

In general, HTC reaction mechanism depends on the composition of feedstock. Hydrolysis, dehydration (production of H_2O), and decarboxylation (production of CO_2 or carbonyls including carboxylic acids) are the primary pathways of HTC, though condensation, polymerization, and aromatization also take place (Fig. 7). At temperatures between 180 and 280 °C for several hours and pressure of 10 – 80 bars, wastes with high organic content may undergo a series of reaction pathways, that are: (1) solid-to-solid conversion through condensation, dehydration, and decarboxylation; (2) liquid-to-solid through polymerization of dissolved intermediates; and (3) aromatization (carbonization) through intermediate solid (coke) [143].

At shorter reaction time and lower temperature, HTC reaction begins with hydrolysis where water in the form of hydronium ions that has high ionic product of H^+ and OH^- [143] will react with glysocidic groups to break ester and ether bonds (typically β-(1–4)-glycosidic bonds) by water autoionization. Subsequently, it produces oligomers like cellobiose, cellotriose, cellotetraose, cellopentaose and cellohexaose, and converting disaccharides (sucrose) and polysaccharides (starch) into their corresponding monosaccharides [145]. Immediately after hydrolysis, dehydration and decarboxylation reaction occurs simultaneously by weakening most of oxygen-containing functional groups (–OH, C = O, and C-O-C) due to the removal of hydroxyl and carboxyl groups to form CO_2 and H_2O as per equation: $4(C_6H_{10}O_5)_n \leftrightarrow 2(C_{12}H_{10}O_5)_n + 10H_2O$. The subsequent reactions stages consist of dehydration and decarboxylation. Dehydration (ring opening and C–C bond breaking) is a dewatering (removal of H_2O) process, where bound water is removed

Table 9
HTC treatments for various MSW to RDF solid fuel.

Feedstock	Operating conditions	HHV	Application	Declorination efficiency	Ref.
Pomelo fruit, PVC, and PP	220 °C, 10 °C/min, 6 h, autogenous pressure, teflon-lined autoclave	22.58 MJ/kg	Co-HTC of porous structure of PP to act as adsorbent/skeleton for PVC condensation (Hydrochar fuel pellets)	93.50 %	[153]
Recycled paper	200, 220 and 240 °C, 3 °C/min, 30, 60 and 90 min, 100 rpm, autoclave	5.04 MJ/m^3 to 17 MJ/m^3	Alternative biofuel (Hydrochar/ solid fuel)	63.57 %	[158]
Organic fraction MSW (OFMSW)	180 °C,1h and 3 h, 220 °C for 3 h, and 250 °C for 6 h, biomass-to-water ratio of 12.9:15.5, 8 – 10 °C/ min, stainless steel (AISI 316) batch reactor	81.6 MJ/kg	Solid fuel (secondary char), soil amendment or environmental adsorbent	n.a	[159]
Woods, plastics, and paper/cardboard from MSW and construction and demolition wastes (C&DW)	250, 300 and 350 °C, 30 min, 0.3 bar, RDF-to-water ratio of 1:2.5, 1:5 and 1:10, 100 rpm, Parr Instruments (stainless steel autoclave reactor)	28 – 33 MJ/kg	Biochars/hydrochars for subsequent energy conversion processes like combustion and gasification	n.a	[142]
Paper, plastics, and food waste	190, 220 and 230 °C, 20, 30 and 40 min, water loading of 70 %, 80 % and 90 %, retrofitted stainless steel tube	15.6 MJ/kg	Biofuel feedstock for standalone fuel or co-feeding with fossil fuels to reduce carbon footprint	n.a	[160]
Industrial refuse-derived fuel produced via mechanical treatment	250, 275 and 300 °C, 30 and 120 min, autogenous pressure, RDF-to-water ratio of 1:15 and 1:5, Parr Pressure Reactor (stainless steel autoclave reactor)	28.1 MJ/kg	Alternative solid fuels	69.2 and 77.9 %	[148]
Food wastes, wood, papers, plastics, textiles, rubbers, and others	180, 230 and 280 °C, 4 °C/min, 60 min, reactor	22.74 MJ/kg	Hydrochar as secondary char and for electrical generation if combine with gasification process	n.a	[161]
Mixed office paper, banana peel, sawdust	190, 210 and 230 °C, 30 and 60 min, Solid load ratio (biomass: aquadest) of 1:3, 1:5, 1:10, SS-304 reactor	24.6, 21.2 and 20.1 MJ/kg for sawdust, paper and banana peel respectively	Solid fuel	n.a	[162]
Plastics, wood, paper and cardboard, foam, textiles, glass, aluminum, others	300 and 400 °C, 30 min, 1 rpm, 4 °C/min, industrial rotary furnace, equipped with emissions collection system	19.9 MJ/kg (torrefied) and 23.5 MJ/kg (carbonized)	Chars	n.a	[126]
Food waste, coal	175, 200, 225, 250, 275, 300, 325 and 350 °C, 60 min, 1L batch high-temperature high pressure vessel	31.1 and 31.4 MJ/kg for food waste and coal hydrochars respectively	Co-HTC of hydrochars and molasses as binder for pelletization	n.a	[46]
Onion skin, banana, chicken, rice, potato peel, orange, fish, potato, carrot peel, apple, beef, pasta, cabbage, banana, lamb, noodle	200, 250 and 300 °C, 1 h, 600 rpm, 1L high-pressure batch reactor	15 to 31 MJ/kg	Co-combustion of hydrothermally converted food waste with coal	n.a	[141]
Food waste (cooked food: meat, seafood, vegetables, rice, noodles and gravy, condiments: salad dressing, ketchup, cocktail sauce) and chopsticks	180 to 260 °C, 60 min, 4 °C/min, 100 rpm, stainless steel autoclave equipped with PID controller and autostirrer	Increased from 17.85 MJ/kg to 31.73 MJ/kg	Hydrochars/solid fuel	n.a	[163]
Food residue and paper content	150 to 280 °C, 30 min, 1.3 to 5.5 MPa, under N_2 gas, 200 L pilot-scale reactor and autoclave reactor and 500 mL autoclave reactor	16.5 to 27.7 MJ/kg for cellulose, 13.9 to 26.5 MJ/kg for hemicellulose, 20.4 to 25 MJ/kg for lignin	Biochar as renewable solid fuel	n.a	[95]
Food waste	150, 250 and 350 °C, 20 min, pressure: 7, 21, 36 bar, Parr stirred pressure batch reactor	17.4 MJ/kg to 26 MJ/kg	Hydrochars and bio-oil	n.a	[164]
Paper (33 wt% from landfills), food waste (rabbit food), mixed MSW (45.5 % shredded office paper, 9.6 % glass (crushed prior to use), 16.4 % shredded plastic bottles, 17.6 % crushed rabbit food, 10.9 % aluminum cans	250 °C, stainless steel tubular reactor	~30 MJ/kg	Hydrochars as environmental adsorbent, soil amendment or a novel material to reduce CO_2-equivalent emissions	n.a	[165]

Abbreviation: n.a., not available.

from the feedstock during HTC because of the increased hydrophobicity in the hydrochar and decarboxylation is a removal of the carboxyl groups process from the extractives, hemicellulose, and cellulose and these materials degrade into monomers like acetic acid, formic acid, and furfurals [146].

Hydrophobicity ensures that the char will not absorb moisture during storage and transportation which makes it more durable and resistance to biological degradation than raw biomass [119]. As a result, atomic O/C and H/C ratio are reduced which means the properties of the hydrochar are upgraded after HTC process, attribute to the decrease of hydrogen and oxygen, and increase of carbon content [147]. Hence, this process contributes to the formation of highly regular and small-diameter particles, yielding solid products with better grindability characteristic [46,148].

This was proven by an experimental study by Triyono et al. [149] as hydrothermal reaction works best at higher temperature (200 C) as it positively changes the product properties, including homogenous shape and uniform size distribution; higher calorific value than coal; and relatively milder condition at 20 bar and 30 min. By increasing temperature or reaction time (further dehydration and decarboxylation), large intermediates monomers are formed through polymerization and cross-linking pathways, where C = C stretching, and C-O-C ether bonds become more intense. This phenomenon reflected an increase in the amount of carbonyl derivatives and formation of aromaticity of carbon-rich structure [147]. Higher temperature and prolong reaction promote the formation of polycyclic structures through condensation of aromatic compounds. Additionally, these conditions shift the condensation reaction towards step-growth kinetics, potentially hindering the access of water into the matrix pores of char by blocking the remaining transport of biomacromolecules, which could trap hydrolysis products [146].

Polymerization reactions are influenced by intermolecular and intramolecular dehydration, while condensation may be induced by aldol condensation and keto-enol tautomerism [143]. This resulting high hydrophobicity and energy yields due to the degrade intermediates from feedstock decomposition [57]. At the same time, aromatization of polymers also takes place. As a result of condensation of aromatized molecules generated in the decomposition of the oligosaccharides and monosaccharides, aromatic clusters were formed which are enhanced by alkaline conditions [150]. The formation of solid char particles in HTC process is facilitated by the connection of aromatic rings, as these rings exhibit high stability under HTC conditions. At higher temperatures, the unreacted waste undergoes fragmentation, promoting aromatization and repolymerization processes. According to a study conducted by Lin et al. [151], it was observed that the aromatic benzene ring = C–H and aromatic C–C stretching vibrations, as well as the aromatic out-of-plane C–H bending peaks, increased significantly with rising temperature. This observation provides clear evidence of the taking place of aromatization reactions during HTC, leading to an increase in fixed carbon content.

Numerous studies have proven that dehalogenation (dechlorination) can be achieved under HTC conditions, which is crucial for the use of hydrochars-based RDF for cement co-processing. A study by Prawisudha et al. [152] observed water-insoluble organic chloride generated from PVC containers was reduced significantly at elevated temperature and pressure from 10,0000 ppm to 2000 ppm. This is most likely because the of the HCl elimination by –OH during the hydrothermal dechlorination process [54]. Wei et al. [153] reported that higher contents of Cl remained in the processing water, which confirms that the chloride traces were successfully extracted into the liquid medium during the HTC. Therefore, the removal of chloride from plastic wastes (e.g., PVC) during HTC process can be summarized in the following equations: $CH_2CHCl + [OH] \rightarrow CH_2CHOH + [Cl]$ and $CH_2CHOH + [O] \rightarrow CH_2COOH + [H]$ [153]. Another mechanism of HTC that improves the efficiency of dechlorination is through co-HTC technique, which more than one material is mixed and torrefied together in the wet environment. As explained by Shen [145], the dechlorination of co-HTC process involves two reaction pathways (elimination and substitution). The reaction initiated with generation of free radicals to form low stability of chloroallylic structures, where HCl will be eliminated to form non-free radical reaction pathway. Once a double bond is created, the allylic Cl-atom on the C-atom that adjoin to the double bond will split off –H and –Cl to structure two double bonds, which in turn activates adjacent Cl to propagate the dehydrochlorination process.

Lignocellulosic components can be added as catalyst to enhance the substitution of –Cl in chloride-containing feedstock. This can be done via degradation of lignin, hemicellulose, and cellulose to form plenty of free –OH bond for the dechlorination reaction process. Based on the studies conducted by Marzbali et al. [154], he concluded that the possibilities of employing catalysts in HTC treatment, which help reduce the operating temperature and time as well as lower the amount of chemical agent and solvent usage. The catalytic process can increase bio-oil yield and HHV of MSW (artificial garbage waste) to 36 MJ/kg. The discoveries further indicated that catalytic HTC is a useful method for producing nano-architectures that can enhance a particular chemical functionality in the hydrochar and hence be a possible approach to treat MSW streams. Examples of catalyst used for HTC of MSW listed in these studies were $ZnCl_2$, Na_2CO_3, and activated carbon [154].

There has been a great interest in the development of MSW as a potential RDF using the HTC technology as summarized in Table 9. The yielded solid indicated as high-grade solid fuel-hydrogen-char, in which the H/C and O/C are reduced for better energy efficiency [155]. As examined by Guo et al. [156] the XRD result of the microcrystalline structure evolution showed that component degradation has led to improve hydrochar structure (highly stable and water-resistant pellets). Generally, RDF prepared from wet torrefaction rendered better energy density (with the same mass yield) as compared to dry torrefaction. Energy density is linked to the amount of chemical energy stored in RDF per unit mass due to the release of volatiles when temperature increases and corresponded with HHV [119]. Even though higher energy density is preferred, the amount of final product that can be obtained and utilized must also be maximized for it to be industrially feasible [112]. The energy density of RDF can be further improved via mechanical [157] palletization process. Pellet quality is also important to be considered as it corresponds to its ability to remain intact during handling and storage [145]. In addition to that, packaging and storage are also important final steps that need to be given extra attention to as the fuel can explode in the storage facilities and surroundings as pointed out by [57]. Pellets crumbling caused by handling and logistic problems can also reduce the combustion efficiency and increase the gas emissions [145].

A study conducted by Acaru et al. [166] examined patent data filed under the Patent Cooperation Treaty (PCT). The study revealed that hydrothermal techniques, which have reached maturity through policies, investment, and knowledge accumulation, primarily focus on enhancing process efficiency. This highlights the need to drive innovation through scale-up and cost reduction using continuous processes and economical reactor designs. The study found that the most frequently cited articles on hydrothermal techniques address the chemical and structural properties of solid residues, the processing of municipal waste, and the solid residues resulting from the processing of lignocellulosic materials. Additionally, there is research on catalysts using feedstocks such as algae, food waste, and sludge to improve cost-effectiveness towards a circular economy [166]. Currently, there are approximately 200 companies and organizations worldwide engaged in researching, developing, and applying HTC technology across multiple markets [167]. Despite the efforts to advance HTC in the industry, there is a lack of information regarding available industrial-scale HTC production. According to Cavali et al. [168], HTCycle technology, a German corporation, produces fertilizers, activated carbon, and coal from sewage sludge and other wet biogenic waste. In 2010, HTCycle became the first company in the world to introduce an industrial-scale HTC plant. However, unfortunately, no information about the reactor can be found.

HTC reactors in manufacturing scale can be grouped into two types of feeding-mode, which are continuous or batch. Most of the reactors are continuous and take more than 5 h to complete one treatment cycle. The batch reactors in industrial plants are usually cylindrical stirred tanks operating in parallel (quasi-continuous multi-batch system) to optimize the process. While continuous reactors are basically smaller than batch reactors that work continuously in a flowing stream under high pressure and temperature, without the need to be cooled down and reheated in between two reaction cycles [169]. HTC plants are based on either vertical ($AVACO_2$, TerraNova Energy) or horizontal reactors (Artec Biotechnologie GmbH, Grenol GmbH) [170]. Whereas the common HTC reactor for laboratory-scale context are hydrothermal autoclave reactor, stainless steel autoclave reactor, polytetra-fluoroethylene-lined stainless autoclave reactor, and teflon-lined autoclave reactor.

Considering the techno-economic viewpoint, it is crucial to consider the expenses associated with (i) material collection, (ii) equipment and devices, (iii) production and operations, (iv) insurance costs, (v) contingency costs, (vi) additional facilities usage, and (vii) energy required to heat the water during the process [171]. The capital investment and

Table 10
Notable advantages and disadvantages of dry and wet/hydrous pyrolysis to convert MSW into RDF.

		Advantages	**Disadvantages**
Dry pyrolysis	Torrefaction (200 – 300 °C)	-Increases hydrophobicity. Enhances reactivity, stability, and grindability. Higher solid product yield at lower temperature. Increases HHV. Simpler operating procedure Lower heating and residence time	High ash deposition, Cl, N & S due to high mineral, salt, and protein from organic waste High number of polysaccharides are conserved at lower temperatures below 300 °C and do not completely degrade.Requires drying (adds to overall process expenditure)
	Carbonization (300 – 600 °C)	Increases HHV at temperature 300 – 600 °C Decreases S & N Increases hydrophobicity, grindability, and homogeneity	Lower solid yield as temperature increase. High HHV when temperature increases above 600 °C. High Cl content Higher heating and residence time
Wet/hydrous pyrolysis	Hydrothermal carbonization (HTC)	Increases hydrophobicity, grindability, and homogeneity. Reduces atomic O/C and H/C ratio. Yields higher HHV and solid product.Increases dechlorination efficiency (reduces chlorine content in solid product) Minimizes pollutant elements. Adding catalyst can enhance product quality. Operates under lower temperature than torrefaction and carbonization	Operates under sub-critical condition at different temperature, pressure, and water content. Needs drying after HTC process completed. Requires high pressures to maintain super-critical conditions. More complex and time-consuming procedure. Corrodes the reactor due to salt and metals present in MSW. Issues in recovery of waste heat. Lack of study on HTC to produce solid fuel to meet basic requirement standard as co-processing fuel in cement industry.

transportation costs for HTC are substantial, as they involve the acquisition of equipment and infrastructure, with the decentralization of wet waste locations, thereby limiting its current industrial utilization. High moisture content with low solids content of MSW necessitate increased energy and usage of larger reactor volume, which contribute to higher operating and capital costs [154] By situating the HTC plant near the wastes source, the cost of transporting feedstock can be significantly reduced, making the operation profitable [172]. A summary of the economic considerations for an HTC plant with a treatment capacity of 78,000 tonnes/year of biowaste (70 % moisture content) and a production output of 15,400 tonnes/year of hydrochar is presented in **Table S3**. Assuming a market price of €180/tonne for the hydrochar and a tipping fee of €50/tonne, the project's internal rate of return (IRR) is 18.7 %. The capital expenditure (CAPEX) is €351/tonne of waste, and the operational expenditure (OPEX) is €20/tonne of waste [173].

4. Recommendations for future works

4.1. Conclusion

The limitations and challenges in conventional solid waste management and handling process such as landfilling, incineration/combustion, 4R, and composting embark the exploration of sustainable waste-to-energy path by converting the MSW into RDF, which act as recovered heat/electricity for co-processing in cement kilns by fulfilling the minimum requirement complementary to the ASTM/EN standards. The large portion of organics component in municipal solid waste (MSW) that rich in biogenic/non-biogenic carbon (e.g. polysaccharides, carbohydrates, lipid, protein, synthetic polymers) are potential energy value that required further treatment prior for the cement co-processing application. This is due to the presence of high moisture content, impurities with heavy metals and hazard compounds, and heterogeneous characteristics of raw MSW that reduce the energy content for efficient usage. Both R&D and commercial phases studies indicated that hydrothermal carbonization (HTC) renders efficacious paths to convert MSW into RDF. This process is well fitted for upgrading MSW with high moisture, where the moisture plays a role as active proton to catalyze the hydrolysis, dehydration, and decarboxylation pathways in converting carbon-based oxygenates in MSW into solid carbon of RDF product (char). The RDF from wet feedstock obtained via HTC process indicates reduction of O/C and H/C ratio of RDF for higher energy quality (>25 MJ/KG), as well as low content of water-insoluble organic chloride that reflecting the effectiveness of HTC to dissolute the inorganic matters in the water medium. However, the present technology requires high capital cost about 50 to 60 % due to the need to invest in the reactors to thermal treat the feedstocks, with large amount of water, which limit their commercialization deployment at this stage. Hence, research and development have been actively conducted to seek for the best waste type, composition, process, and pre-treatment techniques to optimize the operating and capital costs. As a conclusion, RDF produced from MSW is a viable of alternative fuels with promising of zero residual waste left over that ensures complete waste disposal in an environmentally sustainable manner. This approach allows an opportunity of joint effort between municipal waste sector, cement industry, as well as environmental organization to work out on air and land pollution from large MSW disposal. In addition, co-combustion of MSW-derived RDF leads to a substitution of primary fossil, which offers cost savings, minimize waste disposed at landfills or dumpsites, and a net reduction of CO_2 in mitigating climate changes.

4.2. Recommendations

Physico-chemical properties and composition distributions of RDF prepared from HTC of MSW greatly dependent on the process parameters. One crucial requirement is that the solid feedstock must be completely submerged in sufficient water during carbonization in a closed system under saturation pressures. It is noteworthy that the HTC operating parameters (temperature and time) are two important factors that can affect the mass yield and energy densification ratio of the recovered solid. Other significant elements to be considered for high-quality RDF are the water:solid ratio, feedstock size, and mixture distribution of the feedstock. Generally, particle sizes of feedstock between 0.25 mm and 0.60 mm are recommended for optimum treatment.

In addition, the HTC energy input can be improved by lowering water content used during the HTC process, as well as maintaining the optimum recovered solid product (RDF) by lowering the temperature, while prolonging the residence time. Studies indicated that HTC temperature above 300 °C will result in hummus-like product and difficult to be removed from the reactor. In addition, co-hydrothermal carbonization (Co-HTC) was recommended in between organic feedstocks with fossil-based solid fuel (e.g. food waste and coal) at 300 °C, which benefit of low ash yield while maintain high combustion properties and high heating value (>31.4 MJ/Kg). The Co-HTC offers some additional advantages, such as increasing sulphate and chloride removal efficiencies and achieving better homogeneity of MSW-coal blends which are beneficial for co-processing fuel in cement industry. Finally, palletization of RDF is a critical step to enhance energy densities and strength of RDF, which facilitates RDF transportability and storability, as well as enabling the RDF to hold the particles in van interlock matrix. This effort will potentially reduce the high cost of transportation and operation. As shown in Table 10, future problems of adopting HTC for MSW containing high organic matter might include a complicated method requiring more than two experimental factors and high pressure, which could be laborious and costly to operate. Aside from that, drying is necessary after the HTC process to separate the liquid from the solid product. Furthermore, the high salt and metal content of MSW may corrode the reactor over time. As a result, it is suggested that more research and development be conducted on the characterisation and optimisation of HTC parameters.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

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