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Identification of Japanese Solid Biowaste for Conversion into Biochemicals and Energy via Thermochemical Biorefinery

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Japan has one of the best stated and regulated waste management programs worldwide, befitting its shortage of disposal sites and sheltered resources. While being the 5th largest biomass market worldwide, its efficient waste management accounted for a biomass recycling ratio near 71 % in 2020. Still, Japan has a substantial capacity for bioenergy and carbon capture, as its supply chain remains mostly non-renewable-based. A biorefinery prospect is to foray into biochemical production integrated with bioenergy technologies. However, detailed insights are scarce in the literature for a thermochemical biorefinery to biochemicals in Japan. This study aims to clarify the more promising Japanese solid biowastes for thermochemical conversion into bioproducts and bioenergy. For this, technical and regulatory analysis was deployed to fit the interests of the Japanese administration. Conversion of biomass to biochemicals (light olefins and BTX) was calculated using overall rates in the literature. Forest residue (leftover/thinning), rice waste (straw/husk), and cardboard waste were the most available lignified biowaste in 2018. The work forecasted a theoretical substitution near 21 % of the Japanese petrochemical olefins or BTX produced in 2018. The current research precedes a more comprehensive study that deploys the simulation and optimization of thermochemical pathways, assessing the environmental impact and techno-economic feasibility.

1. Introduction

Almost 20 years have passed since the resolution from the Japanese Cabinet on the “Comprehensive Biomass Nippon Strategy”, the first inter-ministerial policy that promotes biomass utilization as a national project (Kuzuhara, 2005). This establishment to prevent global warming through a "biomass-based circular economy" has been materialized since 2002 by enacting sustainable and nationwide environmental normativity (Honma and Hu, 2021). Current regulation encourages a "recycling-oriented society", where residual biomass is a keystone in waste management that accounts for nearly half of the nationwide residues generated (Ministry of Environment, 2020). Accordingly, the “Comprehensive Biomass Nippon Strategy” envisioned for 2030 that the biomass utilization increases to 26 Mt-C/y by extending the recycling of waste and unused biomass above 80 % and 25 %, respectively (Minami and Saka, 2005). According to the New Energy and Industrial Technology Development Organization - NEDO, Japan forecasts by 2030 a Bioenergy generation between 3.7 % and 4.6 % of its energy consumption, and a relevant substitution for petrochemicals above 40 % (Sugie, 2019). The achievement of former goals entails an extensive insight into conversion pathways, embracing the technologies scaling up, economic feasibility, and environmental impact.
Japan has a substantial capacity for bioenergy and bioproducts as its supply chain remains mostly non-renewable-based, ranking the 4th largest consumer of petrochemicals and oil (Wu et al., 2020). A steady rise in biomass-based plastics production (PE, PP, PET, PTT) reached 174 kt/y in 2018, projecting a market size of 360 billion yen. Life cycle analysis of bioproducts also suggests the mitigation of steam-cracking petrochemicals promoting bioplastics through biomass-derived ethylene, propylene, and BTX (Kikuchi et al., 2017). In the framework of a Bioeconomy, an integrated biorefinery enables the production of these compounds (Kikuchi et al., 2017). Still, most commercial-scale biorefineries are designed to yield biofuels, commonly employing biochemical conversion of low lignin content biomass (Ubando et al., 2020). A former analysis of bio-residues in Japan points out a relevant availability of lignified biomass like forest and rice residues, which are hardly

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converted through biochemical pathways (Mayorga et al., 2020). In this regard, lignified biowaste can be converted alongside thermochemical pathways that easily decompose lignin. Unfortunately, few studies evaluate thermochemical conversion of Japanese lignified biowaste, involving the synthesis of biomass-derived oligomers, aromatics, or resins (Ubando et al., 2020).

This study aims to identify and quantify the current availability of lignified biowaste in Japan for its thermochemical conversion to added-value products. The last valorization encourages a novel approach for a diversified waste management beyond conventional energy usage. This work initially examines the national regulation of waste management to understand the current legislation on biowaste and administration agencies. In this regard, the presented study focused on official statistics and reports to reliably estimate biowaste production by each residue type. Hereafter, special attention is paid to sort abundant biowaste using its recycling ratio, streams for incineration, and landfill disposal. In the second section, the suitable synthesis of bioproducts from lignified biowaste is evaluated by contrasting and selecting pathways in the literature. The last section summarizes the theoretical production of bioproducts to substitute steam-cracking petrochemicals like olefins and aromatics in Japan.

2. Estimation methodology

The present article employed official statistics of waste production reported by the Ministry of Environment and the Ministry of Agriculture, Forestry, and Fishing. Complementary data was also obtained from reports of the Statistic Bureau of Japan, the NEDO Agency, and the New Energy Industrial Forum. The Annual Report on Environmental Statistics, issued by the Ministry of Environment, incorporates a national survey conducted by 47 prefectures every five years (Ministry of Environment, 2020). The latest report covers the statistics in 2018 of the production of wastes and 23 sub-treatments (including direct recycling, natural reduction, commercialization, incineration, and landfill disposal). The last document also classifies biomass in municipal, industrial, and general, reporting 25 subcategories of biomass effluents.

For the present article, the mass flows of the treatments are merged in recycled stream ($\dot{m}_{recycle}$), reduced stream ($\dot{m}_{reduct}$), and landfill disposal flow ($\dot{m}_{landfill}$). $\dot{m}_{recycle}$ consists of direct and indirect recycling, natural reduction, commercialization, and conversion to feed or fertilizer. $\dot{m}_{reduct}$ accounts for the stream to direct incineration, weight loss by drying, and conversion to fuel. The matter unused ($\dot{m}_{unused,i}$) is calculated with the sum of $\dot{m}_{reduct,i}$ and $\dot{m}_{landfill,i}$. Biowaste is reclassified in "wood and forest residues", "wastepaper", "agricultural residues", and "non-lignocellulosic biomass". For this study purpose, the lignified biowastes correspond to solid waste biomass that presents a lignin content above 15 % g/g on a dry and ash-free basis (Aristizábal-marulanda et al., 2019). The recycle ratio ($Recycle_{ratio,i}$) and unuse ratio ($Unuse_{ratio,i}$) are calculated with Eq(1) and Eq(2), respectively.

$$Recycle_{ratio,i} = \dot{m}_{recycle,i} / \left(\dot{m}_{recycle,i} + \dot{m}_{reduct,i} + \dot{m}_{landfill,i}\right) \tag{1}$$

$$Unuse_{ratio,i} = \left(\dot{m}_{reduct,i} + \dot{m}_{landfill,i}\right) / \left(\dot{m}_{recycling,i} + \dot{m}_{reduct,i} + \dot{m}_{landfill,i}\right) \tag{2}$$

The theoretical bioproduct flows ($\dot{m}_{j,i}$) for the light olefins ($j = olefin$) and aromatics ($j = BTX$) are calculated with Eq(3) for each biowaste i, where the overall ratio of biomass to bioproduct ($\mathrm{BT_j}$) for olefins production is taken from Arvidsson et al. (2016). Likewise, $\mathrm{BT_j}$ for BTX production is estimated with data of Xiang et al. (2015). The ratios of biochemical to petrochemical ($Bio/Petro_{j,i}$) are calculated with Eq(4) for both olefins and BTX. In 2018, the Japanese petrochemicals production ($\dot{m}_{petro,j}$) was 13.33 Mt/y for olefins ($j = olefin$, including ethylene and propylene), and 12.85 Mt/y for aromatics ($j = BTX$, including benzene, toluene, and xylene) (Sumitomo Chemical Co. Ltd, 2021).

$$\dot{m}_{j,i} = \dot{m}_{unused,db,i} \times \mathrm{BT_j} \tag{3}$$

$$Bio/Petro_{j,i} = \dot{m}_{j,i} / \dot{m}_{petro,j} \tag{4}$$

The power cogeneration during the thermochemical conversion of Biomass to Olefins – BTO ($\dot{W}_{with\,BTO,i}$) is estimated with the specific energy output reported in the literature (Arvidsson et al., 2016). Finally, the power generation using biomass exclusively as fuel ($\dot{W}_{without\,BTO,i}$), is calculated assuming a Rankine power generation cycle with thermodynamic efficiency ($\eta_{th,gen}$) of 30 % (Kuzuhara, 2005). The characterization of the residues is obtained from the database Phyllis2, developed by the Energy research Centre of the Netherlands – ECN. The statistic control also includes literature per residue for the contents of moisture (MC_{ad}), Lignin, and the Lower Heating Value (LHV_{ad}) (Zhou et al., 2014).

3. Solid residual biomass in Japan

After the rapid economic growth period and the later expansion of waste incineration capacity, the Japanese government redirected awareness on waste reduction ascribing the Basic Act for Establishing a Sound Material-Cycle Society and the 3R scheme in 2000 (Ministry of the Environment, 2014). As a result, major biomass-related measures from the “Comprehensive Biomass Nippon Strategy” evolved to the foundation of the "Biomass usage promotion council", “The Fundamental Law of Promoting Usage of Biomass” in 2009, the “Biomass commercialization strategy” in 2012, and the “New basic plan for biomass usage promotion” in 2016. Recycling in Japan involves strict regulations based on diverse residue acts (containers and packings, food, construction, etc.) (Dente et al., 2020). Residues are principally categorized in municipal waste, industrial waste, and general waste, with a mass sharing near 12 %, 69 %, and 19 %, respectively (Ministry of Environment, 2020). Based on current regulations, the responsibility of biowaste management falls on business operators for industrial waste and on municipalities and waste-generators for municipal waste. Table 1 presents the production of wastes in Japan during 2018 per residue type and treatment.

Table 1: Production of wastes in Japan during 2018. Non-lignocellulosic biowaste includes food waste, livestock manure, and sewage sludge. Other than biomass includes minerals and fossil wastes.

Residue	Production	$\dot{m}_{recycling}$	$\dot{m}_{reduct}$	$\dot{m}_{landfill}$	$Recycle_{ratio}$	$Unuse_{ratio}$
Unit	Mt/y	Mt/y	Mt/y	Mt/y	% g/g	% g/g
Total waste production 2018	548	310	223	14	57	43
Biomass residue	297	127	167	2.8	43	57
Wood and forest residue	21.0	13.0	7.8	0.2	62	38
Leftover treetop/branch	5.88	1.41	4.47	0.00	24	76
Wood from thinning	3.92	0.94	2.98	0.00	24	76
Sawmill factories waste	5.32	5.10	0.10	0.12	96	4
Wood chips from construction	5.90	5.57	0.23	0.10	94	6
Wastepaper	22.9	18.3	4.6	0.03	86	14
Cardboard / Paperboard	2.42	0.65	1.78	0.00	27	73
Fine paper, Magazine, sanitary	7.60	4.74	2.82	0.03	62	38
Old newspaper	2.32	2.32	0.00	0.00	100	0
Kraft and corrugated containers	10.58	10.58	0.00	0.00	100	0
Agricultural residue	18.0	11.7	6.2	0.05	65	35
Rice straw	7.93	6.71	1.22	0.00	85	15
Rice husk	1.73	1.51	0.23	0.00	87	13
Wheat straw	1.05	0.77	0.28	0.00	73	27
Others (potato, bamboo, grass)	7.27	2.24	4.48	0.05	36	64
Non-lignocellulosic biowaste	235	84	148	2.5	36	64
Others than biomass	251	183	56	11	73	27

Residual biomass is a cornerstone in waste management that accounted for 54 % of the total residues generated in 2018. Although the production of solid biowaste has remained mainly constant in the last 20 years, the recycling ratio has steadily increased from 29 % in 1998 to 43 % in 2018 (Ministry of Environment, 2020). Nearly 52 % of industrial waste and 20 % of municipal waste were recycled in 2018, while 41 % was incinerated and only 3 % proceeded to landfills. Lignocellulosic biomass represents 20 % of the biomass residues, with a recycling ratio of 69 % in 2018. Less than 0.5 % of the lignified biomass reached land disposal, and the recycling ratio in most of the residue was above 90 %. Incineration of biowaste is caused by techno-economic issues for expensive collection, poor quality of recycled products, or unsuitable processing with current technologies.

While most wood residues like sawmill and construction wood waste report a high utilization ratio, forestry residues represent a challenge for the administration. Besides forest floor disposal, the logistic and transportation of treetops and branches are currently expensive. Yoshioka et al. (2006) find the development of suitable systems for the steep Japanese topography crucial, suggesting an in-forest mobile chipping approach as the best way to reduce manipulation cost. Other effective measures to reduce this cost are trailers and high-grade forest roads that directly transport leftovers to the landings of logging sites. Still, the Japanese Forest Agency plans to improve the recycling ratio above 30 % by 2030. For this effect, the Feed-in Tariff (FIT) impelled the generation capacity with woody power plants to 16,815 GWh in 2020 (Fushimi, 2021).

Wastepaper is efficiently recycled to produce a wide range of paperboards and printing paper. The recovery of paper is currently limited because of sanitary paper (incinerated) and waterproof paper (expensive to retreat). Cardboards and paperboard report the lowest recycling ratio in the paper industry. The excessive recovery of paperboard reduces the quality of the cellulose due to numerous loops of reuse. Consequently, the production

of paper from cardboard is limited because of processing troubles (screen clogging) and poor appearance (whiteness deterioration). Cardboard presents an interesting potential for its utilization out of the paper industry. Agricultural residues are mainly disposed to compost, livestock feed, soil replacement, and mulch. Indeed, rice waste utilization is one of the most important improvements of the waste management reforms, contributing to 28 % of the reduction in the Japanese environmental impact since 1998 (Dente et al., 2020). Unfortunately, almost 14 % of rice straw and husk are burned in situ to be reincorporated into the soil since their low density makes transportation expensive without improving profitable usage. While the former approach adds value to rice straw and husk as fertilizers, the burning of these biowaste contributes to biogenic CO_2 emissions that represented 8 % of the Japanese environmental impact in 2010 (Dente et al., 2020).
Biomass utilization is currently prioritized in Japan with the "Biomass 5F" scheme (Fushimi, 2021), a hierarchical valorization of the biomass from higher to lower as Food, Fiber (chemicals and materials), Feed, Fertilizer, and Fuel (biofuels and power generation). The "Biomass 5F" scheme recognizes the diversity of biowaste utilization, regarding bioenergy and biofuel as large biomass consumption methods that release low-value compared to other products (such as food, chemicals, or fertilizers) (Fushimi, 2021). Biofuels and bioenergy emerged from the pressure to mitigate CO_2 emissions in the energy industry that values of 43 % in the global breakdown, blurring the urgency in the chemical industry that only shares 4 %. By fortune, the current developments on solar, wind, and hydrogen-based power help to spur biomass toward added-value products embedded in a coherent Bioeconomy (Yadav et al., 2020). The upcoming analysis concerns the production of added-value chemicals that can store biogenic CO_2 through a biorefinery with integrated power generation.

4. Conversion of lignocellulosic biowaste to biochemicals in Japan

This study employs the design methodology proposed by Aristizábal et al., to lay out a biorefinery, defining the feedstock, products, platforms, and processes (Aristizábal-marulanda et al., 2019). Figure 1 summarizes the selected pathways from biomass to bioproducts (olefins or BTX) using thermochemical conversion (gasification or pyrolysis). Table 1 reveals an attractive availability of biowaste in Japan as feedstock, following the classification of lignocellulosic residues like forest residue, rice waste, and cardboard (accounting for low recycling ratio or burned in situ). Most commercial-scale lignocellulosic biorefineries employ biochemical conversion of low lignin content biomass (Ubando et al., 2020). Lignified biowaste can be converted with thermochemical processes because gasification and pyrolysis ease the thermal decomposition of aromatic compounds present in lignin (Talero et al., 2019). Consequently, an initial "lignin platform" is selected in terms of the feedstock, followed by gasification and pyrolysis.

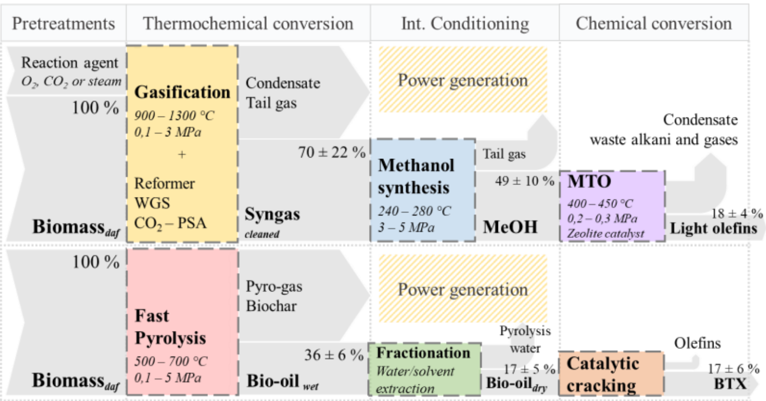

Figure 1: Conversion pathways in a thermochemical biorefinery integrated with power generation and oriented to produce biochemicals (light olefins and BTX). Mass flows consider average values reported in the literature.

Regarding the biorefinery products, the building block in conventional petrochemistry to produce several polymers and intermediate chemicals are the steam cracking olefins (mainly ethylene, propylene) and the catalytic reforming aromatics (benzene, toluene, and xylene) (Kikuchi et al., 2017). The substitution of crude oil-derived olefins or BTX extends the migration to biochemicals by employing developed polymerization technologies. For this reason, the present work focuses on light olefins and BTX, excluding the polymerization process to PE, PET, or others. It is still supposed that biomass-derived olefins and BTX achieve quality standards for polymers production. Additionally, the production of light olefins is preferred over BTX. Light olefin is favoured for the carbon-carbon double bonds, allowing a broader application to polymers, or even enabling the production of BTX via steam cracking.
According to Figure 1, the gasification process leads to a "syngas platform" converted to olefins via methanol synthesis and catalytic conversion of Methanol to Olefins – MTO. The "methanol platform" and MTO grant a

mature technology, as most methanol is commercially produced from natural gas by steam reforming and from coal by gasification reactions (Kansha et al., 2019). Biomass pyrolysis evolves a "Pyrolytic liquid platform" known as Bio-oil. The Pyro-gas and Biochar only generate energy for simplification of the study, but the potential for chemical conversion is proposed for future study. The pre-concentration is performed with water/solvent extraction to increase the thermal stability of the Bio-oil. These enriched fractions are converted to bio-aromatics with a catalytic cracking upgrading system using microporous zeolites and metal oxides.
The overall ratio $\mathrm{BT_{olefin}}$ is estimated from the literature between 12.7 % and 24.8 % (Xiang et al., 2015). Likewise, the overall ratio $\mathrm{BT_{BTX}}$ is estimated from the literature between 14.1 % and 24.6 % (Yan and Li, 2021). The specific energy released after the olefins production is reported in the literature between 144 kWh/t and 189 kWh/t, excluded from thermal and electrical auto-demand. Prior yields are used in Table 2 to estimate a theoretical maximum and minimum production of olefins and BTX from the Japanese lignocellulosic biowaste in 2018, including only residues that exhibit high availability for a low recycling ratio or field burned.

Table 2: Estimated maximum production of biowaste-based olefin, BTX, and power cogeneration from Japanese lignocellulosic waste biomass in 2018.

	Residue	Forest residue (leftovers)	Rice waste (straw/husk)	Cardboard waste
MC_{ad}	% $g/g_{BM.db}$	21.8 ± 7.5	9.6 ± 1.2	8.1 ± 1.8
Lignin daf	% $g/g_{BM.db}$	26.9 ± 2.6	24.1 ± 10.5	20 ± 1.0
LHV_{ad}	MJ/kg	16.8 ± 0.9	19.7 ± 1.0	17.1 ± 0.8
$\dot{m}_{unused.db}$	Mt/y	5.82	1.31	1.63
Light olefin production				
$\dot{m}_{olefin}$	Mt/y	1.45 - 0.83	0.32 - 0.19	0.41 - 0.23
$Bio/Petro_{olefin}$	% g/g	12.8 - 7.3	2.9 - 1.6	3.6 - 2.1
BTX production				
$\dot{m}_{BTX}$	Mt/y	1.43 - 0.82	0.32 - 0.18	0.4 - 0.23
$Bio/Petro_{BTX}$	% g/g	11.1 - 6.3	2.5 - 1.4	3.1 - 1.8
Energy cogeneration				
$\dot{W}_{with\ BTO}$	MW_e	125 - 96	28 - 22	35 - 27
$\dot{W}_{without\ BTO}$	MW_e	933	245	265
$\dot{W}_{with\ BTO}/\dot{W}_{no\ BTO}$	% MW_e/MW_e	13.44	8.80	10.30

The conversion of all the highly available biowaste to biochemicals should substitute from 21 % to 17 % of the Japanese consumption in 2018 of petrochemical olefins or BTX, respectively. The surplus energy during the production of biochemicals is 90 % below the power generation if biomass is used as fuel. The last approach forecasts a promising economic scenario since the market price of wood pellets is 250 USD/t compared to the average value of 1,250 USD/t for ethylene and propylene (Yadav et al., 2020).
Several challenges remain for the integration of thermochemical conversion to biochemicals in Japan. While former studies point out the importance of a decentralized utilization of biomass in small rural areas with a capacity of approximately 20 t/h, future studies must identify the most promising locations in Japan (Kuzuhara, 2005). Besides, it is essential to assess the price and demand of process feedstock chemicals in the international market (Kansha et al., 2019). The direct gasification of biomass also evolves a relevant amount of condensate matter, reducing the sensibility of olefins. Thus, a comprehensive recognition among conversion pathways is advised in future studies, assessing recent advances in Catalytic Fast Pyrolysis - CFP, Syngas to Dimethyl ether to Olefins - DMTO, or direct catalytic Syngas to Olefins - STO. It is outlined that Fischer-Tropsch to Olefins - FTO yields gasoline over light olefins, adverse to diminishing biomass fuels (Arvidsson et al., 2016).

5. Conclusions

The last analysis has elucidated the potential of a thermochemical biorefinery with Japanese lignified biowaste. Forest residue, rice waste, and cardboard waste rendered the greatest available biowaste in 2018, accounting for 57 % of incinerated lignocellulosic biomass. The utilization of forest residues and rice waste represents a challenge due to expensive and ineffective collection. Cardboard waste emerges as the best residue for thermochemical conversion, complementing its availability within industrial areas. Although this research contributes as an introduction to more comprehensive biomass utilization in Japan, additional insights are recommended for future works regarding detailed simulations and optimization of pathways, the assessment of environmental impact, or techno-economic feasibility. Moreover, the most promising locations for thermochemical biorefineries in Japan must be recognized in coming studies, reducing feedstock manipulation and transport costs. The maximum production of light olefins and BTX is estimated at 2.16 and 2.17 Mt/y with uncoupled gasification and pyrolysis. Still, primary pyrolysis before gasification depicts an alternative to be evaluated to increase olefins selectivity. This study suggests the production of light olefins over BTX, allowing broader application to polymers (PE, PET, etc.), intermediate chemicals, or even BTX via steam cracking.

Nomenclature

ad – as determined basis
BT – Biomass To (Olefins or BTX)
$Bio/Petro$ – Biochemical to Petrochemical ratio
daf – dry and ash free basis
db – on dry basis
i – subindex for each waste biomass
j – subindex for each Olefins or BTX
LHV_{ad} – lower heating value

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