Chemical Looping Gasification for Sustainable Production of Biofuels

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Deliverable D1.1:

Analysis of Selected Feedstock

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1 Introduction

While the CLARA-project aims for validating and demonstrating chemical looping gasification (CLG) of biogenic residues in a relevant environment, the consortium is encouraged to keep in mind also constraints with regard to the market introduction and scaling potential of the investigated biomass-to-end-use chains. The CLG-process will yield bio-gasoline and bio-diesel. However, the feedstock basis for this process can be shifted to lower quality biogenic carbon sources compared to the sugar-, starch- and oil plants used for conventional liquid biofuels. Therefore, competition with food production can be avoided and dependencies regarding the restriction of land use or regarding limited potentials of used cooking oil can be reduced. Moreover, the process can be driven feedstock-flexible using a broad and variable portfolio of biogenic residues. Using biogenic residues also has the advantage of being in line with the EU's biofuels policy documented in the RED II directive. In this directive residue based biofuels (or so-called advanced biofuels) will be promoted by quotas that will have to be fulfilled by the mineral oil sector.

Since feedstock prices often contribute substantial shares in bioenergy deployment costs, they can pose a major barrier (e.g. (Müller, 2013)). Especially also when considering seasonal aspects for feedstock sourcing and pricing, this flexibility will have a significant effect on the economic feasibility and on unit scaling potentials of the individual CLG-plants and on the market upscaling potential of biomass gasification in general.

However, for the market introduction of CLG of biogenic residues a trade-off between lowest hanging fruits in terms of best performing feedstocks and highest market potentials have to be found. The aim of this deliverable is to select a reference feedstock, representative agricultural and forestry residues for further testing and development in the CLARA-project. Parameters that should be considered include among others ash melting point, contents of volatile inorganics like alkali and chlorine as well as physical properties with regard to transport, handling, storage and feeding into the reactor. Feedstock potentials, sourcing costs or biogenic carbon carrier prices (e.g. pellet prices) need to be investigated such as feedstock yields, seasonal availabilities and logistical accessibilities. Biomass feedstocks exhibit lower carbon densities, higher water contents and a higher heterogeneity in contrast to fossil based feedstock. Transportability, storeability and tradeability can be enhanced through densification. Thus, optimisation through drying, torrefaction, leaching and adding additives prior pelletising is part of this project and has also to be considered during feedstock selection.

In the upcoming sections, we discuss criteria for the feedstock selection and provide a long-list and project specific classification of possibly relevant biogenic residues and biogenic carbon carriers. Based on the long-list, representative feedstocks for the CLARA-project have been selected in a climate neutral online meeting on 13.November 2018. The selected feedstocks include:

- As a representative for agricultural residues; Wheat straw from sourcing regions close to the CENER facility in Spain and close to ABT facilities in Sweden
- As a representative for forestry residues; Pine wood residues from sourcing regions close to the CENER facility in Spain and close to ABT facilities in Sweden
- As a representative for commoditised & densified biogenic carbon carriers; Industrial wood pellets of considerable low grades should be used in CLARA

Based on the market- and technical assessment reports (Section 5 and Section 6 respectively) the representative feedstocks are presented and their implications for the CLARA-project discussed in this deliverable.

2 Criteria for feedstock selection

Selection criteria have been discussed within the consortium. They can be summarised in (1) technical and (2) market criteria for the raw materials and for the respective densified bioenergy carriers.

The objective function of the selection exercise is to identify biogenic residues that maximise the chances for reaching a higher TRL while at the same time being a good representation for feedstocks with high European sustainable sourcing potentials.

Technical criteria:

- Heating value
- Moisture content
- Bulk density
- Ash content & composition
- Ash melting temperatures
- Nitrogen, Chlorine and Sulphur content
- Handling and feeding behaviour, e.g. particle size distribution, particle density, angle of repose
- Pre-treatment properties, e.g. millability, drying energy
- Gasification properties, e.g. elemental composition
- Handling, storage and feed-in properties

Market criteria

- Sustainable sourcing potentials
- Price developments, sourcing costs composition
- Transport costs, storability and storage costs
- Feedstock yields, seasonal availabilities and logistical accessibilities
- Fulfilment of RED II directive

3 List of relevant feedstocks and bioenergy carriers

An internationally accepted classification of feedstock types as well as their traded forms and raw materials can be found in the ISO 17225-1:2014 standard on “solid biofuels – fuel specifications and classes”. Although this feedstock list excludes currently important bioenergy feedstocks such as used biogenic municipal solid waste, manure, used cooking oil and sewage sludge further referred to as “post-food & feed”, it represents the best starting-point for the discussion of woody-, herbaceous-, fruit- and aquatic biomass. In the follow-up documents (ISO 17225-2 until ISO 17225-8) standards for firewood, wood chips and thermally treated and un-treated (woody and non-woody) pellets and briquettes are outlined (ISO, 2018).

In Table 1 a long-list and classification of potentially relevant feedstocks is shown. Numbering and classifications are adopted from the ISO 17225-1 standard. Therefore, all “blends and mixtures” are excluded, even though blends and mixtures will be used in the CLARA-project. However, this classification doesn´t serve a purpose for the primary identification of feedstocks that can later be blended or mixed anyway. Furthermore, all "chemically treated" biomass types are excluded. These types have special traits due to the treatment and are expected to reduce the comparability to other feedstock types, rendering these biomass types a poor representative feedstock.

Additionally, the following classifications from the standard are **excluded** to increase the straightforwardness of the selection process:

- “1.1.2 Whole trees with roots”; efforts in cleaning the root biomass from soil or high ash-contents of un-treated root biomass would reduce the TRL of the gasification project.
- “1.1.4.1 & 1.1.4.2 Fresh/Green biomass”; increased moisture and ash content would reduce the TRL of the pre-treatment & gasification combination. Water content can be minimised through storing and cleaning from leaves and needles is generally necessary as discussed for the classifications 1.1.4.3 and 1.1.4.4.
- “1.1.5 Stumps/roots”; efforts in cleaning the root biomass from soil or high ash-contents of un-treated root biomass would reduce the TRL of the gasification project.
- “2.1.1.1, 2.1.13, 2.1.14 Whole plant, grains or seeds, husks or shells from cereal crops” are excluded due competition with food- & feed use, respectively husks or shells are too volatile and pose too high explosion risks at handling. They also exhibit critical ash-melting behaviours

- “2.1.2.3 & 2.1.2.4 Seeds and shells from grasses” do not exhibit potentially relevant yields (for seeds) resp. are too volatile and pose too high explosion risks at handling.
- “2.1.3 Oil seed crops”; competition with food- & feed use or 1st generation biofuels, thus not in the focus of this project. Residues from oil production however are included under “2.1.3.2 Stalks and leaves” and also cover empty fruit brunches e.g. from palm oil production
- “2.1.4 Root crops”; efforts in cleaning the root biomass from soil or high ash-contents of un-treated root biomass would reduce the TRL of the gasification project. Overbred root crops with higher cleaning-yield ratios like potatoes are used for food- & feed production.
- “3.1 Orchard and horticulture fruit”; direct competition with food- & feed industry, respectively their residues are included in the classification 3.2 anyway.
- “3.2.1.1 and 3.2.1.3 berries & stone/kernel fruits/fruit fibre residues”; no substantial biomass yields are expected for this biomass types, rendering these types feasible only for very specific use cases. However, their stone/kernel fruits/fruit fibres and crude olive cake is included in the long-list (3.2.1.2 & 3.2.1.4 respectively).
- “4.1, 4.2 & 4.3 algae, water hyacinth and lake and sea weed”; these types are harvested with too high moisture contents for processing to solid biofuels and can be used rather for direct pyrolysis, (centralised) gasification or anaerobic digestion but are not interesting for decentralised feedstock flexible gasification.

As already discussed an additional classification “post food/feed biomass” is introduced. Under “6.1 food processing residues”, the classification “6.1.1 mixed biogenic municipal solid wastes” should be considered too in the selection process, while “6.1.2 used cooking oil” is not in the scope of this project. Under “6.2 Metabolised residues”, “6.2.1 and 6.2.2, manure and sewage sludge” are excluded since they are also not interesting for decentralised feedstock flexible gasification.

In the next step we discuss major traded forms of solid biofuels. In Junginger et al., (2014) global net solid biofuel trade is outlined based on Lamers et al. (2012a). They estimate about 300 PJ (about $18*10^9$ kg) internationally traded biomass for direct biomass consumption in 2010 with the highest share in **wood pellets** followed by similar shares for **fuelwood** and **wood waste** and a small amount of **roundwood** and **wood chips**. However, “industrial roundwood dominates absolute international woody biomass trade volumes.” While on average, 40-60% of roundwood ends up in material use, the remaining is used as bioenergy in the form of black

Table 1: Long-list and classifications of primary feedstocks potentially relevant for the CLARA project. Source: Own selection, modification and illustration based on ISO 17225-1/2014.

1. Woody biomass	1.1 Forest plantation and other virgin wood	1.1.1 Whole trees w/o roots	1.1.1.1 Broad-leaf
			1.1.1.2 Coniferous
			1.1.1.3 Short rotation coppice
			1.1.1.4 Bushes
		1.1.3 Stem wood	1.1.3.1 Broad-leaf with bark
			1.1.3.2 Coniferous with bark
			1.1.3.3 Broad-leaf w/o bark
			1.1.3.4 Coniferous w/o bark
		1.1.4 Logging residues	1.1.4.3 Stored, Broad-leaf
			1.1.4.4 Stored, Coniferous
		1.1.6 Bark (from forestry operations)	
		1.1.7 Segregated wood from gardens, parks, roadside maintenance, vineyards, fruit orchards and driftwood from freshwater	
	1.2 By-products and residues from wood processing industry	1.2.1 Chemically untreated wood by-products & residues	1.2.1.1 Broad-leaf with bark
			1.2.1.2 Coniferous with bark
			1.2.1.3 Broad-leaf w/o bark
			1.2.1.4 Coniferous w/o bark
			1.2.1.5 Bark (from industry operations)
	1.3 Used wood	1.3.1 Chemically untreated wood	1.3.1.1 Without bark
			1.3.1.2 With bark
			1.3.1.2 Bark
2. Herbaceous biomass	2.1 Herbaceous biomass from agriculture and horticulture	2.1.1 Cereal crops	2.1.1.2 Straw parts
		2.1.2 Grasses	2.1.2.1 Whole plant
			2.1.2.2 Straw parts
			2.1.2.5 Bamboo
		2.1.3 Oil seed crops	2.1.3.2 Stalks and leaves
		2.1.5 Legume crops	2.1.5.1 Whole plant
			2.1.5.2 Stalks and leaves
			2.1.5.3 Fruit
			2.1.5.4 Pods
		2.1.6 Flowers	2.1.6.2 Stalks and leaves
		2.1.7 Segregated herbaceous biomass from gardens, parks, roadside maintenance, vineyards, fruit orchards	
	2.2 By-products and residues from food and herbaceous processing industry	2.2.1 Chemicall untreated herbaceous residues	2.2.2.1 Cereal crops and grasses
			2.2.2.4 Legume crops
			2.2.2.4 Flowers
3. Fruit biomass	3.2 By-products and residues from fruit processing industry	3.2.1 Chemically untreated fruit residues	3.2.1.2 Stone/kernel fruits
			3.2.1.4 Crude olive cake
4. Aquatic biomass	4.4 Reeds	4.4.1 Common reeds	
		4.4.2 Other reeds	
6. Post food/feed biomass	6.1 Food processing residues	6.1.1 Mixed biogenic municipal solid wastes	

liquor, bark, sawdust and wood chips. This indirect trade of biomass for energy is estimated with 400 PJ of industrial round wood and 200 PJ of pulp chips & particles in 2010. Fuelwood and **charcoal** are considered to be "traditional energy carriers" which are typically not traded internationally and exhibit no political impetus of extending its importance. No distinguished statistics on international trade of **wood briquettes**, thermally treated biomass (**torrefied pellets**), **square- & round bales** or densified herbaceous biomass such as **straw pellets** are known by the authors. This indicates currently vanishing importance for bioenergy trade of these biogenic carbon carriers, even though the extension and commoditisation of some of these carriers will be decisive in the future for phasing out fossil based carbon carriers (Schipfer, 2017).

In summary the following densified biogenic carbon carriers have to be considered additionally to the long-list of primary biomass feedstocks outlined in Table 1:

- Woody, herbaceous & aquatic biomass compressed mechanically to pellets or briquettes.
- Woody, herbaceous & aquatic biomass thermally pre-treated and compressed to torrefied pellets or torrefied briquettes.
- Woody, herbaceous & aquatic biomass compressed to small square bales (0.1m^3), to big square bales (3.7 m^3), or round bales (2.1 m^3) according to ISO 17225-1.

Furthermore, the following processed raw materials exhibit preferable properties (e.g. energy density and durability) for transportation, handling and storage:

- Wood chips
- Crushed, cutted, debarked, bundled, planed wood
- Grain or seeds
- Fruit stones or kernels
- Fibre cake
- Wood wastes and other tradeable biogenic waste fractions

4 Selection of feedstocks

Based on the discussion of the long-list and the experience and interest of the consortium three feedstocks have been selected on which we perform technical and market related assessments in a climate neutral online meeting on 13 November 2018. The following feedstock classifications from Section 3 will be considered for the market and technical assessment:

- As a reference material, standardised industrial grade wood pellets have been selected to build upon a highly available biogenic carbon carrier, which is on the one hand as homogenised as possible for gasification test during the project duration at different purchase locations. On the other hand the CLARA-project aims at developing industrial gasification projects thus rendering high-quality wood pellets graded for household economic unfeasible.
- Representing the section “forestry residues”, pine wood chips have been selected due to their availability and potentials in Europe and worldwide, including different regions relevant for the CLARA-project and consortium. In the classification the feedstock falls under “1.1.4.4 Stored coniferous logging residues” from forest plantations and other virgin wood. Also spruce wood chips fall into the same category and will be discussed and tested throughout the project.
- As a feedstock representing “agricultural residues”, wheat straw delivered in the form of bales has been selected due to its availability and potentials in Europe and worldwide, including different regions relevant for the CLARA-project and consortium. In the classification the feedstock falls under “2.1.1.2 Straw parts” under cereal crops in the herbaceous biomass section.

5 Market assessment of selection

5.1 Industrial graded wood pellets

Wood pellets available in Europe contain mainly saw-dust from the wood processing industry, imported pellets from the US and Canada can also be based on whole trees (AEBIOM, 2015). In the ISO17225-2 graded wood pellets are defined for commercial- (combined heat and power and/or district heating) and residential applications (A1, A2 & B) and industrial use (I1, I2 & I3). The standard contains ranges for shapes and sizes, moisture-, ash content, mechanical durability, fines-, additives contents, net calorific values (NCVs), bulk densities, particle size distributions of disintegrated pellets and chemical compositions further discussed in Section 6 (ISO, 2014). For the market assessment especially the NCV-values (16.5 $GJ*t^{-1}$) and the bulk density (600 $kg*m^{-3}$) are important as well as the mechanical durability which render this biogenic carbon carrier an internationally tradeable and biomass feedstock that can be eventually stored over months.

Thrän et al., (2018) discusses a global demand for wood pellets (industrial, commercial & residential) of about 25 Mt in 2015 with the USA being the largest producer (6.3 Mt in 2016), followed by Canada (2.4 Mt in 2016), Germany (2.2 Mt) and Sweden (1.5 Mt). Largest consumer is the United Kingdom with 6.7 Mt and Denmark with 2.8 Mt in 2015 mainly for electricity production in refurbished coal fired power plants. The USA consumed about 2.9 Mt and Italy about 2.1 Mt mainly in the residential sector. Reported installation pelletising capacities are at about 43 Mt globally with the USA representing 32% of this capacity. However, compared to the production of about 26 Mt in 2015, the overall capacity utilisation rate is at about 60%.

Industrial graded wood pellets are either purchased on the wholesale market in the Baltic Sea region and at the Amsterdam-Rotterdam-Antwerp (ARA-) ports or often also traded using over-the-counter and long-term contracts or are even produced by an affiliated company (vertical integration). Prices for wholesale markets are provided posthumously by the FOEX Indexes Ltd. and the Argus Wood Pellets Index respectively. Between 2009 and 2015 these spot market pellet prices ranged between a maximum of 185 $US\$*t^{-1}$ (mid 2014) and 113 $US\$*t^{-1}$ (December 2016) and an average of 169 $US\$*t^{-1}$. (Thrän et al., 2018) While seasonal fluctuations can be observed for residential consumer graded pellets due to consumption peaks for heating during winter (Schipfer, 2017), prices for industrial pellets used for base load electricity production do not necessarily exhibit these seasonality but can be still influenced by single events such as fires in power plants as seen after an accident in the RWE-plant in Tilbury in 2012 and in a Drax power plant in 2017. How a possibly ongoing integration between these markets (residential and industrial) will influence prices and availability in the future remains to be discussed.

While the ISO 17225-2 covers technical standardisation aspects, aspects concerning sustainability criteria such as CO_2-emissions throughout the entire biomass-to-end-use chain are not addressed. For wood pellets especially sustainable forest management (SFM) is a precondition for this bioenergy pathway to contribute to reaching the climate goals. Existing certification schemes include the Forest Stewardship Council (FSC), Programme for the Endorsement of Forest Certification (PEFC) and the Sustainable Biomass Program (SBP). The latter is gaining significant market shares with about 50% of the market in the UK, Belgium and Denmark being covered by SBP certified pellets in 2017 (4.7 Mt) (Thrän et al., 2018).

Torrefaction, as a mild form of pyrolysis, is heating biomass up to 250-350°C in the absence of oxygen and can convert biogenic raw-materials into biofuels with improved properties such as energy density, grindability and hydrophobicity, especially when combined with pelletisation. In Q4-2016 the ISO 17225-8 was published to outline technical specifications of respective energy carriers for the international market. According to Thrän et al., (2016) eight torrefaction plants have been in operation and three under construction in Europe and the USA in 2015. Due to the early implementation phase of this technology, no considerable trade streams of torrefied pellets can be outlined so far.

5.2 Pine wood residual wood chips

Francescato et al., (2008) discuss market relevant values for wood fuels. Wood chips of coniferous trees and a moisture content (on wet basis) of 30% are discussed with about 220 $kg*m^{-3}$ and 12.2 $GJ*t^{-1}$. While wood chips derived from roundwood are mainly used for pulp and paper production in Europe, wood chips for energy purposes can be either sourced from recovered/waste wood or from harvesting residues “such as branches, tops, thinnings or other inferior wood not suitable for material or pulp and paper production” (Junginger et al., 2014). However, also parts of high-quality wood chips end up in energy use in the form of black-liquor combustion in the pulp and paper industry.

Density and NCV for wood chips are considerably lower than for wood pellets, however international trade is still feasible especially for shorter trade distances. Main sourcing areas are on the one hand the Baltic states and Russia for markets in Sweden, Denmark and also Germany, on the other hand Italy imports large volumes from Balkan countries but also Spain and France. Exact numbers on traded volumes for traded logging residues pine wood chips for energy use are not available. International trade of wood chips is discussed to be dominated by high quality chips for the pulp and paper industry as well as wood chips from waste wood (Junginger et al., 2014).

Forest residues potentials in Europe are estimated with about 1.200 PJ (120 Mt) in the Horizon 2020 SECTOR project (Alakangas et al., 2013). A later study assesses the sustainable potential of biomass for energy use in the EU28. Hoefnagels et al., (2017) based their estimates on the EUwood study and the EFSOS-2 and the EFISCEN-Model calculating annual EU-forest increments of about 11.000 PJ until 2030. The report discusses between 16-26% of the forest biomass potential to be logging residues potentials. The H2020-Rehap project estimates the production share between different wood types. Thorenz et al., (2018) presents the results with the focus on bark as a residue from different wood types based on the assumption of a bark-to-wood ratio of 10-15%. A ratio between broadleaf and coniferous wood is discussed with 40:60 with spruce as the more important coniferous feedstock compared to pine wood. This would add up to a range of 600-1.000 PJ per year pine wood residues without bark based on the EU-forest increments. Main pine wood sourcing countries are discussed to be Sweden, Finland and Germany.

Similar to trade statistics for low quality wood chips for energy use from pine wood residues, there are also no official prices time series. To give an estimate for local sourcing and supply the following case studies can be considered: For thinning operations with a chainsaw Francescato et al., (2008) assume production costs of about 15 €*m_{bulk}^{-3} (about 70 €*t^{-1}) based on chainsaw thinning, tractors forwarding, chipping with a high power chipper and delivery to the end-user with a truck and trailer. The price at the power station in this example is stated with 80-90 €*t^{-1}. For main felling in a coniferous stand and chipping of the road side residues in a high power chipper and forwarding to the end-user with a truck and trailer costs can be reduced to 5 €*m_{bulk}^{-3} (about 20 €*t^{-1}). The price at the power station in this example is stated with 30-40 €*t^{-1}. Wood chip prices from forestry residues did most likely undergo an increase in the 10 years after the discussed publication. For a comparison with high quality wood chips for pulp and paper we state prices from the FOEX Index Ltd. which reports price levels of 196 US$*$t^{-1}$ (172 €*t^{-1}) for softwood and 178 US$*$t^{-1}$ (156 €*t^{-1}) for hardwood at the time of writing of this report (November 2018) (FOEX, 2018).

Nilsson, (2016) discusses the operational method of “dried-stacking” for providing a “dry forest fuel containing a small quantity of needles, as requested by the energy-conversion industry.” Therefore, logging residues are left to dry during the felling period over summer. Through defoliation (needle fall-off) nutrients can be furthermore left in the forest and moisture content decreases over time. Forwarding is then carried out in an optimised time window not too early in the season but also not too late in autumn for not risking re-moistening and biological degradation of the residues. However, logging residues can also be sourced e.g. during winter if logging is scheduled to minimise the (soil-) damage of machinery by logging on frozen grounds or when severe weather conditions ask for additional maintenance measures. Logging residues from thinning operations account for a significantly smaller although growing share compared to logging residues from final felling today.

5.3 Wheat straw bales

In the Horizon 2020 BioBoost-project logistical issues of suppling wheat straw bales have been discussed. Rotter and Rohrhofer, (2014) define square bales with the dimensions of 2.4 x 1.2 x 0.9 m and a weight of 500 kg resulting in a bulk density of 193 kg*m^{-3} and 14% moisture content (166 kg* m^{-3} dry matter). The NCV is estimated with about 13 GJ*t^{-1}. International trade of straw bales for energy purposes is not known to the author and is considered to be not feasible due to the discussed low energy and bulk densities. Bales are rather sourced and used locally and transported with a farm tractor and platform or drawbar trailer and handled with front-end loaders or telescopic handlers directly to the end-user or via an intermediate storage.

Current EU-28 demand for straw (2014) was assessed in the H2020 Rehap-project. Thorenz et al., (2018) estimate about 29 Mt to be used today for agricultural, energy and industrial applications. The main share is used for cattle bedding & fodder (12 Mt) followed by pig, sheep and horse bedding & fodder. About 2.5% of cereal production is cultivated organically where 100% of the straw is returned into the soil. The European Commission (2013b in Thorenz et al., 2018) discussed about 5 Mt straw mulching for organic and non-organic production. Another 3 Mt are used for other agricultural products mainly for mushrooms through composting and to a smaller extent for strawberries for protection against frost in the higher latitudes. About 2 Mt have been furthermore used in 2014 in 15 combined heat and power plants in the EU28 (mainly in Denmark) and vanishing amounts for second-generation biofuels (e.g. in Crescentino, IT) and other biochemicals for material use or for insolation in the building sector.

Agricultural residues potentials reviewed in Hoefnagels et al., (2017) range from 1.000 – 7.100 PJ for Europe depending on the potential definitions and residues considered (straw, cuttings, pruning residues, sunflower and rice husks and bagasse). Wheat straw however contributes most to the total share of residues with 42% on average. The report concludes with an ecologically sustainable agricultural residue potential of 2.400 PJ for energy purposes in 2030 in the EU28 (thus about 1.000 PJ for wheat straw). Thorenz et al., (2018) conclude with a current technical potential for wheat straw of about 57 Mt. In this publication, the potential is defined after discounting for issues of law, sustainability and technology, as well as without refining residues but including competitive applications. To provide a comparison, this value would equal to about 740 PJ based on a heating value of 13 $GJ*t^{-1}$.

Harvesting of wheat takes place either around July until end of August for winter strains or starting with August until around October for spring-sown strains (USDA, 2018). Winter wheat production generally dominates in the EU28 with an average of 80% of the total area of wheat production with most significant outliers including Finland and Estonia (Eurostat, 2018).

For the BioBoost-project (Pudelko et al., 2015) estimated straw prices of about 50 $€*t^{-1}$ (3.6 €/GJ) if 50% of straw on the field is sourced and the rest remains as a natural fertiliser and soil conditioner for the next crop cycles. Another 12 $€*t^{-1}$ can be estimated for handling and forwarding to the gasification plant within a radius of 50 km (Rotter and Rohrhofer, 2014).

Scarlat et al., (2010) report an average sustainable removal rate for wheat, barley, rye and oat residues of 40% based on experts' estimates and literature analysis. The remaining part plays a crucial role in "maintaining or improving soil characteristics, protecting the soil from erosion, maintaining or increasing soil organic matter, maintaining mineral nutrients in soil and improving water retention" (Nelson, 2002 in Scarlat et al., 2010).

6 Technical assessment of the feedstock selection

The biomass feedstocks have physical and compositional differences: moisture, ash content, net calorific values (NCV), bulk densities and chemical composition. This section is focused on feedstocks ash chemical composition and its effect on ash fusibility behaviour during thermal conversion.

Low ash, low moisture content feedstock have higher NCVs (as received) and are therefore preferred from the technical point of view leading to higher process efficiencies. In this sense, wheat straw is interesting due to its typical lower moisture content (10-25%) compared with pine (30-55%). In addition, bulk density and flow behaviour is also relevant due to the impact in logistics cost and gasification process. While wood chips have typical bulk densities of 180-220 kg/m^3, chopped wheat straw is below 120 kg/m^3, so pelleting can decrease transport cost through increasing biofuel density to over 600 kg/m^3.

Although it is not part of the objective of this section, particle size distribution and bulk densities of the biofuels for gasification should be considered and measured throughout the project. These parameters are relevant from a technical operation point of view, especially when talking about gasifier feeding system and its fluidised conditions. However, physical properties can be improved by means of chipping, chopping, drying and pelleting, etc., these kinds of pre-treatments don´t affect chemical composition.

Selected feedstock have been characterized and compare against bibliographic data (ECN, 2019; Lemus et al., 2013) in order to check it´s representativeness. The following tests have been carried out for the characterisation based on international measurement standards:

Table 2: Reference standard for characterisation test. Source: own illustration

Test	Reference Standard
Ash Content	UNE-EN ISO18122
Elemental Analysis (CHN)	UNE-EN- ISO 16948
Net Calorific Value	UNE-EN 14918
Sulphur and Chlorine content	UNE-EN- ISO 16994
Ash Composition-Major elements (Al, Ca, Fe, Mg, P, K, Si, Na and Ti)	UNE-EN- ISO 16967
Ash Composition-Minor elements (As, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, V and Zn)	UNE-EN- ISO 16968
Ash melting behavior	CEN/TS 15370-1

Most biomass has significant contents of inorganic matter (Coulson et al., 2004; Demirbas, 2004; Jenkins et al., 1998) and many of the problems in thermochemical processes are related with its quantity and behavior. According to the profile of inorganics in biomass ashes, shown in Figure 1, the following classification is proposed by Boman et al., (2013):

- Woody fuels are calcium oxide and carbonate based ash
- Grassy fuels are silicate based ash
- Fast growing crops are phosphate based ash.

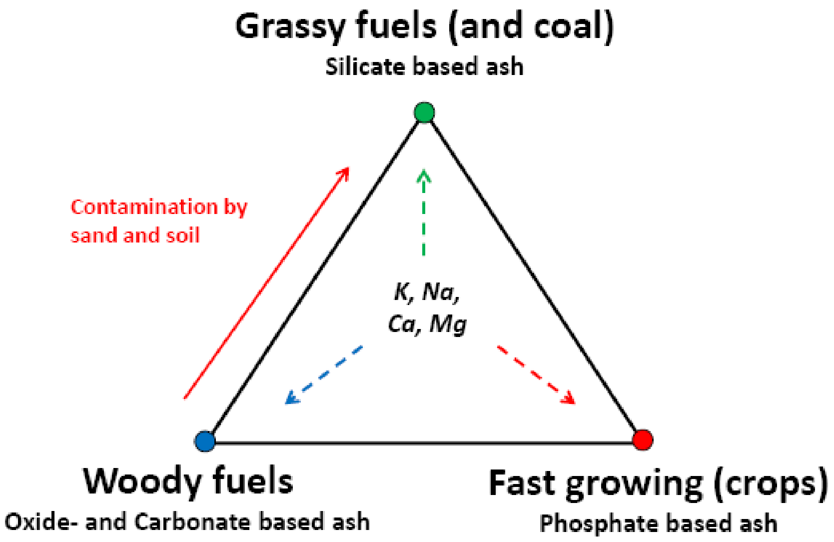

Figure 1: Compositional differences for different types of biofuels. Source: (Boman et al., 2013)

The compositional differences in the inorganic matter influence fate of elements in the gasification process and also the behaviour of the produced ashes. Some of these elements present in the inorganic matter like alkali metal compounds, phosphorus compounds and some heavy metals have a higher tendency to volatilise.

Besides, the behaviour depends on the presence of silica, sulphur and chlorine and their interactions affecting the amount of condensable vapours released to the gas stream. In this sense wheat straw (ash >6%; Table 3) is problematic feedstock due to its high ash, potassium and chlorine content (Table 3 and Table 6). Hence, leaching pre-treatment will be needed to decrease ash content on wheat straw in order to reach lowest quality industrial pellet specifications (Table 5). On the other hand, there are also differences between woody biomasses such as wood chips (ash <0.5%;) and bark (ash 5-7%) reported by Obernberger et al. (in Nussbaumer et al., 2001; Obernberger et al., 2003) which can be also checked from other bibliographic data measured from CLARA pine forest residue (Table 4). In the case of bark, a higher calcium content is observed in ashes than in the chips (Table 7) causing a higher release of calcium oxide which facilitates later condensations.

Sulphur content must be considered as a key element during the project, not only because of its interactions with other elements in the gasifier bed, but also by its H_2S-release to the product gas.

Table 3: Proximate Analysis, Ultimate Analysis and NCV - Wheat Straw. Ẋ represents the average. Source: own measurements and ECN, (2019)

Sample		Wheat Straw (CLARA)	Wheat Straw (Bibliography)		
Parameter	Units	Measured	Range		Ẋ
Ash	% (d.b.)	7.5	1.3	13.5	6.4
C	% (d.a.f.)	48.2	46.3	52.6	48.9
H	% (d.a.f.)	6.5	3.2	6.4	5.9
N	% (d.a.f.)	0.43	0.3	2.1	0.7
O	% (d.a.f.)	44.9	39.4	47.9	44.1
S	% (d.a.f.)	0.11	0.03	0.46	0.15
Cl	% (d.a.f.)	0.05	0.002	2.3	0.4
Net Calorific Value (NCV)	Mj/Kg (d.a.f.)	19.9	15.2	20.5	18.2

a.r.: as received
d.b.: dry basis
d.a.f.: dry and ash free

Table 4: Proximate Analysis, Ultimate Analysis and NCV - Woody Biomass. Ẋ represents the average. Sources: own measurements and ECN, (2019)

Sample		Pine Forest Residue (CLARA)	Pine			Pine Bark			Pine Needles
Parameter	Units	Measured	Range		Ẋ	Range		Ẋ	Ẋ
Ash	% (d.b.)	2.0	0.07	1.0	0.4	1.6	10.7	3.8	1.5
C	% (d.a.f.)	52.7	48.8	55.0	51.8	53.4	56.1	54.5	48.9
H	% (d.a.f.)	6.4	5.8	7.0	6.3	5.8	6.2	5.9	6.7
N	% (d.a.f.)	0.39	0.02	0.4	0.13	0.1	0.8	0.4	n.d.
O	% (d.a.f.)	40.5	38.1	45.1	41.6	37.5	40.3	39.2	44.4
S	% (d.a.f.)	0.05	0.01	0.6	0.05	0.03	0.1	0.07	n.d.
Cl	% (d.a.f.)	0.007	0.01	1.2	0.06	0.009	0.03	0.016	n.d.
Net Calorific Value (NCV)	Mj/Kg (d.a.f.)	20.2	17.9	21.5	19.4	19.7	20.5	20.2	19.0

a.r.: as received
d.b.: dry basis
d.a.f.: dry and ash free

Table 5: Ash, N, NCV, S and Cl– Industrial Wood pellets. Source: External test report of the pellet trader

Graded Wood Pellet		CLARA	I1	I2	I3
Parameter	Units	Certified			
Ash	% (d.b.)	0.73	≤1.0	≤1.5	≤3.0
N	% (d.b.)	≤0.05 [a]	≤0.3	≤0.3	≤0.6
S	% (d.b.)	0.005 [a]	≤0.05	≤0.3	≤0.6
Cl	% (d.b.)	≤0.005 [a]	≤0.03	≤0.05	≤0.1
Net Calorific Value (NCV)	Mj/Kg (a.r.)	17.96	≥16.5	≥16.5	≥16.5

a.r.: as received
d.b.: dry basis
d.a.f.: dry and ash free

Although ISO 17.225-2 does not detail any criteria about major elements content for industrial wood pellets its analysis is recommended. Due to interactions of major elements, especially alkaline, alkali earths and silicates and depending on chlorine and sulphur availability ash fusibility temperatures can cause troubleshooting. Typical ash related problems at the gasifier bed could be bed sintering or agglomeration causing bed defluidisation.

Table 6: Major elements as oxides-Wheat Straw. Ẋ represents the average. Sources: Own measurements and ECN, (2019)

Sample	Wheat Straw (CLARA)	Wheat Straw (Bibliography)		
Oxide (% ash)	Measured	Range		Ẋ
P2O5	1.6	1.2	7.9	2.8
SiO2	44.5	27.3	72.5	55.4
Fe2O3	1.2	0.09	2.2	0.6
Al2O3	4.3	0.09	3.9	0.9
CaO	9.1	2.6	17.0	7.6
MgO	2.2	0.75	4.3	2.1
Na2O	0.4	0.06	9.8	1.3
K2O	34.0	5.9	36.7	18.8
TiO2	0.6	0.01	0.2	0.07

Table 7: Major elements as oxides - Woody Biomass. Ẋ represents the average. Sources: own measurements and ECN, (2019)

Sample	Pine Forest Residue (CLARA)	Pine			Pine Bark			Pine Needles
Oxide (% ash)	Measured	Range		Ẋ	Range		Ẋ	Ẋ
P2O5	4.2	0.1	4.8	2.5	n.d.	n.d.	4.8	n.d.
SiO2	17.1	5.6	10.2	n.d.	1.3	39.0	13.9	n.d.
Fe2O3	1.6	2.0	2.1	2.0	0.3	3.0	1.2	n.d.
Al2O3	2.7	1.8	2.5	2.1	5.3	14.0	8.2	n.d.
CaO	39.5	12.3	32.9	25.7	25.5	40.6	35.6	n.d.
MgO	6.3	0.6	1.6	1.1	4.5	6.5	5.2	n.d.
Na2O	0.4	0.2	0.9	0.7	0.4	1.3	0.7	n.d.
K2O	27.6	1.0	6.8	4.8	6.0	7.6	7.1	n.d.
TiO2	0.6	0	0.8	n.d.	0.08	0.2	0.12	n.d.

Table 8: Minor elements - Wheat Straw. Ẋ represents the average. Source: own measurements and ECN, (2019)

Sample	Wheat Straw (CLARA)	Wheat Straw (Bibliography)		
Compound (mg/kg d.b.)	Measured	Range		Ẋ
As	<0.8	<0.8	1.2	1.0
Cd	<0.2	<0.2	0.3	0.2
Co	<1	<1	6.4	3.9
Cr	9	9	60	2.6
Cu	5	5	11.4	3.8
Hg	<0.05	<0.05	0.05	n.d.
Mn	46	46	100	19.9
Mo	<2	<2	2.2	1.1
Ni	5	5	4	0.9
Pb	3	3	3.1	0.2
Sb	<1	<1	n.d.	n.d.
V	5	5	6	0.4
Zn	10	10	60	15.7

In the case of minor elements, some of them are specified in the ISO 17.225-2 (see Table 8). The composition of selected feedstock meet by far these requirements in all cases: CLARA industrial wood pellets (Table 5), wheat straw (Table 8) and pine forest residue (Table 9). Notwithstanding the above, during the gasification process its concentration on the bottom ash or in the gas phase depending on the volatilisation tendency must be checked to avoid unforeseen problems.

Table 9: Minor elements – Woody Biomass. Ẋ represents the average. Source. Own measurements and ECN, (2019)

Sample	Pine Forest Residue (CLARA)	Pine			Pine Bark			Pine Needles		
Compund (mg/kg d.b.)	Measured	Range		Ẋ	Range		Ẋ	Range		Ẋ
As	<0.8	n.d.	n.d.	n.d.	0.1	4	n.d.	0	0.3	0.2
Cd	<0.2	<0.05	0.5	n.d.	0.2	1	n.d.	0	0.3	0.1
Co	<1	<0.2	0.5	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Cr	2	0.2	10	n.d.	1	10	n.d.	0.5	0.8	0.6
Cu	2	0.5	10	n.d.	3	30	n.d.	2.0	5.0	3.6
Hg	<0.05	<0.004	0.05	n.d.	0.01	0.1	n.d.	0	0	0
Mn	25	40	200	n.d.	9	840	n.d.	n.d.	n.d.	n.d.
Ni	<1	<0.1	10	n.d.	2	20	n.d.	0.5	2.5	1.4
Pb	<2	<0.5	10	n.d.	1	30	n.d.	0.2	2.8	1.3
Sb	<1	0.01	0.5	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
V	<1	0.2	2	n.d.	0.7	2	n.d.	n.d.	n.d.	n.d.
Zn	19	5	50	n.d.	70	200	n.d.	10.0	50.0	31.4

Biomass ash behaviour is studied according to the CEN / TS 15370-1 Solid biofuels, Method for the determination of ash melting behaviour, within which the following terms related to ash fusibility are defined (shown in Figure 2):

- Shrinkage Start Temperature (SST): temperature at which the area of the sample tested decreases to 95% of the initial area at 550 ° C.
- Deformation Temperature (DT): temperature at which the first signs of rounding of the axes due to fusion occur.
- Hemisphere temperature (HT): temperature at which the sample tested forms a hemisphere; for example when its height is equal to half the diameter of the base.
- Fluid temperature (FT): temperature at which the ash expands on the surface, the height is half of the sample at HT.

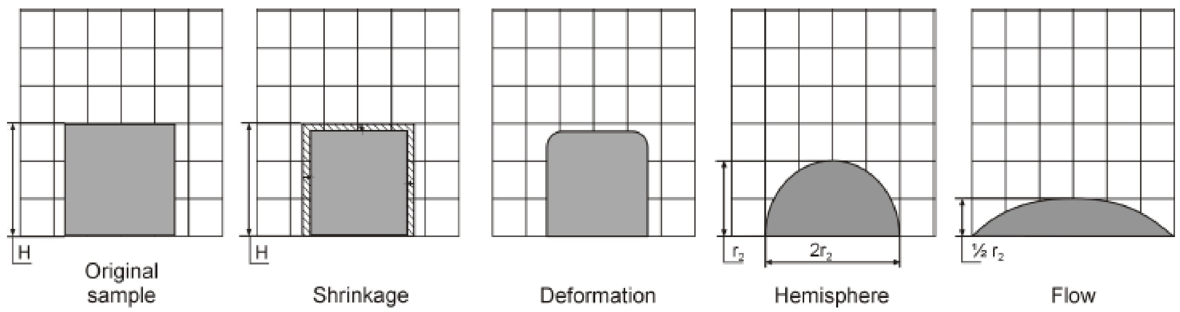

Figure 2: Ash Fusibility Test Temperatures in CEN / TS 15370-1

Ash fusibility temperatures are shown in Table 10 for wood pellets, pine forest residues and pine (bibligraphic data for pine, pine bark and pine needles), and in Table 11 for wheat straws (both, measured form selected feddstock and bibligraphic data). The main difference between feddstocks is base on the initial defornation temperature (IDT). As bed temperaures during gasification are in the range of 850-950 ºC, straw could be a more problematic feedstock causing bed defluidisation due to its IDT compared whith pine or pine residues which IDT is higher (>1.140 ºC).

Table 10: Ash Melting Temperatures in oxidation atmosphere – Woody Biomass. Ẋ represents the average. Sources: own measurements and ECN, (2019)

Sample	Wood Pellet (CLARA)	Pine Forest Residue (CLARA)	Pine	Pine Bark			Pine Needles
Temperature (ºC)	Certified	Measured	Ẋ	Range		Ẋ	Ẋ
IDT	1.140	1.100	1.150	1.210	1.340	1.275	1.270
SOT	1.150	1.480	1.180	1.249	1.525	1.387	n.d.
HT	1.200	>1.490	1.200	n.d.	n.d.	1.650	n.d.
FT	1.200	<1.490	1.225	1.288	1.650	1.469	1.370

Table 11: Ash Melting Temperatures in oxidation atmosphere - Wheat Straw. Ẋ represents the average. Sources: own measurement and ECN, (2019)

Sample	Wheat Straw (CLARA)	Wheat Straw (Bibliography)		
Temperature (ºC)	Measured	Range		Ẋ
IDT	870	780	1.080	896
SOT	1.080	800	1.110	968
HT	1.130	1.040	1.280	1.130
FT	1.190	1.080	1.500	1.255

7 Discussion and conclusions

Possible relevant biogenic feedstocks and densified biogenic carbon carriers for CLG have been clustered and a long-list of categories has been provided. Decisive feedstock criteria for the CLARA-project and the market introduction and –diffusion of CLG have been outlined. Based on the long-list and criteria, representative feedstocks for the CLARA-project have been selected in a climate neutral online meeting on 13.November 2018 and an overall market- and technical assessment was performed.

Wheat straw, pine wood chips and industrial graded wood pellets are selected for testing in the CLARA-project. However, fuel-flexibility of fluidised bed gasification is expected to be high enough and for example, it is expected that an industrial straw gasifier design should also be able to use other feedstocks such as palm oil shells and residues from olive oil production. The flexibility topic will be further discussed throughout this project.

The market assessment for each feedstock outlined in this report focuses on the feedstock definition and its overall form and function as purchased on the markets, overall key market points such as demand, production capacity and trade streams, price ranges and the formation of prices as well as relevant standards and sustainability considerations. While wood pellets mainly based on saw dust can be seen as an internationally traded commodity with functioning market mechanism, wheat straw and pine wood chips for energy purposes are traded rather regionally and no international statistics, standards or prices exist. How commoditisation of pellets and torrefied pellets based on these feedstock could develop will also have to be discussed in this project (WP6 & WP7).

Challenges related to the selected feedstock are especially (1) feeding of pellets in small plants and (2) low-temperature ash melting phases. The first issue can easily be solved by crushing/milling of the pellets. The second issue will be comprehensively investigated in WP2 and WP3. Also, the inorganic matter content and its composition, especially alkaline, chlorine and sulphur have to be analysed on the different feedstock. Their presence and fate during gasification is an important issue in order to avoid problems related with generated ash melting behaviour.

However, it is expected that feedstock properties will vary during the project, depending on sourcing area and region. Therefore, all batches will have to undergo an analysis prior gasification in order to derive generalizable results and insights into the potentials and challenges of chemical looping gasification.

The technical material testing reports of this deliverable and of the ongoing testing of the various batches throughout the project are collected in a standardised Excel format and will be stored on CLARA server accessible to all CLARA participants.

The results will be directly used in Task 1.2 “Basic definition of process chain” as well as in Task 2.1 (Lead by CENER) “Biomass pre-treatment method development”. One main goal of the CLARA project is the demonstration of the full process chain in the pilot plant at TU Darmstadt. The pilot plant requires pellets as feedstock to enable continuous feeding in large amounts (i.e. around 70 t per test campaign). Therefore, ABT will produce pellets from pine wood residues and wheat straw for pilot tests in WP5 at TUDA (Task 5.3) and deliver them to Darmstadt in bulk by truck. Pellets are also expected to be used as feedstock for full-scale commercial plants due to feeding issues (particularly at elevated pressure) as well as decentralised sourcing and pre-treatment of the biomass. All tests in small and large pilot plants will use the same (or at least similar) pellets in order to allow for a direct comparison of the results. TU Darmstadt acquires industrial wood pellets that will be used as a reference feedstock in both small-scale and pilot-scale tests. CENER will produce torrefied pellets from wheat straw and non-torrefied pellets from pine wood residues for the lab-scale tests in WP2/WP3 at CENER, FZJ, UNIVAQ, CSIC and CTH (Task 2.4). Feedstock cost-supply curves will be furthermore developed and used in Task 7.1 (CENER + VUT).

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