

The Influence of Torrefaction on the Combustion Behavior of Selected Agricultural and Waste-Derived Solid Biomass Fuels

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The utilization of agricultural and waste-derived solid biomass fuels for generation of sustainable energy is widely hampered by their “difficult” combustion behavior. Torrefaction can be a well suitable procedure to improve the fuel properties of these fuels. In this study the combustion behavior of five widely available untreated and torrefied biomass fuels - forest residues (FR), sugar cane leaves (SCL), green waste (GW), winter wheat straw (ST) and sunflower husks (SFH) - were investigated by the application of a multi-step laboratory approach (chemical analyses, fuel indices and fixed-bed lab-scale reactor combustion tests). The investigations revealed that SFH (untreated and torrefied) can be recommended as a fuel directly applicable in grate furnaces (also without flue gas recirculation) because of the low slagging tendency, but high aerosol emission needs to be considered. Also FR, SCL and GW (untreated and torrefied) can be applied under consideration of the slagging tendency, whereas ST is not recommendable concerning its severe slagging tendency. However, for all fuels investigated the applicability in grate fired plants needs special considerations, since they do not behave like natural stem wood. Therefore, an appropriate validation by means of experimental investigations with a lab-scale combustion facility is highly recommended before using such fuels. The combustion tests reveal the measures which must be taken to ensure an undisturbed and continuous operation on grate furnaces. By execution of such combustion tests the advantages of torrefied material (e.g., higher calorific value, better storage feasibility, etc.) can be fully exploited.

1. Introduction, Background and Scope

Agricultural and waste-derived solid biomass fuels are available in vast amounts for generation of sustainable energy. Since these fuels usually have rather low heating values, low bulk densities and are disadvantageous concerning their storage behavior, torrefaction of such materials represents an interesting procedure to improve their fuel properties. Torrefaction of biomass is a mild form of pyrolysis at temperatures typically between 200 and 320 °C which changes biomass properties to provide a much better fuel quality for combustion and gasification applications. According to Bergmann et al. (2005) torrefaction leads to a dry product with no biological activity like rotting. The authors concluded that torrefaction combined with densification leads to a very energy-dense energy carrier. Another review article from Ptasinski et al. (2011) concluded that the main advantages of torrefaction are the improvement of energy density and grindability. Jones et al. (2013) showed that torrefaction induces changes in chemical structure that lead to improved hydrophobicity and grindability. They also found that optimization of the torrefaction temperatures maximizes benefits of torrefaction under consideration of good energy yields. Bi et al. (2015) revealed that torrefaction kinetics have been systematically investigated under both inert (N₂) and oxidative (O₂, H₂O) environments for a variety of biomass species. The authors also concluded that the gasification could be improved in terms of both energy efficiency and syngas quality because of the removal of oxygenated volatile compounds from torrefied biomass.

Although several studies are available describing the impact of torrefaction on the fuels properties, only limited literature is available which directly compares the combustion behaviour of thermally untreated and torrefied biomass fuels. Therefore, this study aims to determine the potential of improved combustion properties by torrefaction as a pre-treatment step for selected agricultural and waste-derived solid biomass fuels.

2. Methodology

The combustion behaviour of five widely available (selection criteria) untreated and torrefied biomass fuels, - forest residues (FR), sugar cane leaves (SCL), green waste (GW), winter wheat straw (ST) and sunflower husks (SFH) – should be investigated by the application of a multi-step laboratory approach (chemical analyses, fuel indices and fixed-bed lab-scale reactor combustion tests).

The application of the following analyses methods, which have also been used by the authors, is a result of the FP6 project BioNorm, presented in Bärenthaler et.al (2006), which among other aspects also dealt with the definition of standards and best practice guidelines for biomass fuel analyses. The water content of the fuel is determined according to EN 14774-1. The fuel samples and ash processing for the following analyses are carried out according to EN 14780. The ash content is determined by ashing the fuel at 550°C (EN 14775). Calcium-rich fuels tend to form considerable amounts of carbonates at 550°C. During combustion, however, practically no formation of carbonates occurs; instead oxides are formed exclusively, due to the prevailing high combustion temperatures. As a result, the ash content determined by the standard is overestimated in comparison to the amount of ash produced in real systems. For this reason, the “total inorganic carbon” (TIC) value was used to calculate the so-called “corrected ash content” based on EN 13137. The C, H and N contents of fuels are determined according to EN 15104 by means of combustion and subsequent gas chromatographic separation with an elementary analyser. The chlorine content of fuels is determined according to EN 15289. The concentrations of main and secondary elements in fuels and ashes are determined according to EN 15290 and EN 15297.

The results of chemical analysis are the basis for the calculation of fuel indices, which can be used for a pre-evaluation of the combustion behaviour. In this study the sum of K, Na, Zn and Pb (evaluation of the aerosol emissions expected), the molar Si/K ratio (indicator regarding the K release), the molar 2S/Cl ratio (indicator for high temperature corrosion), the molar Si/(Ca+Mg) and (Si+P+K)/(Ca+Mg) ratio (as indicators concerning ash melting behavior) have been used. In a former study of Sommersacher et al. (2012) the before mentioned fuel indices and respective background information for the derivation as well as boundary conditions of application of these indices are presented.

For the next step thermally untreated and torrefied fuels were combusted in a fixed-bed lab-scale reactor (see Figure 1).

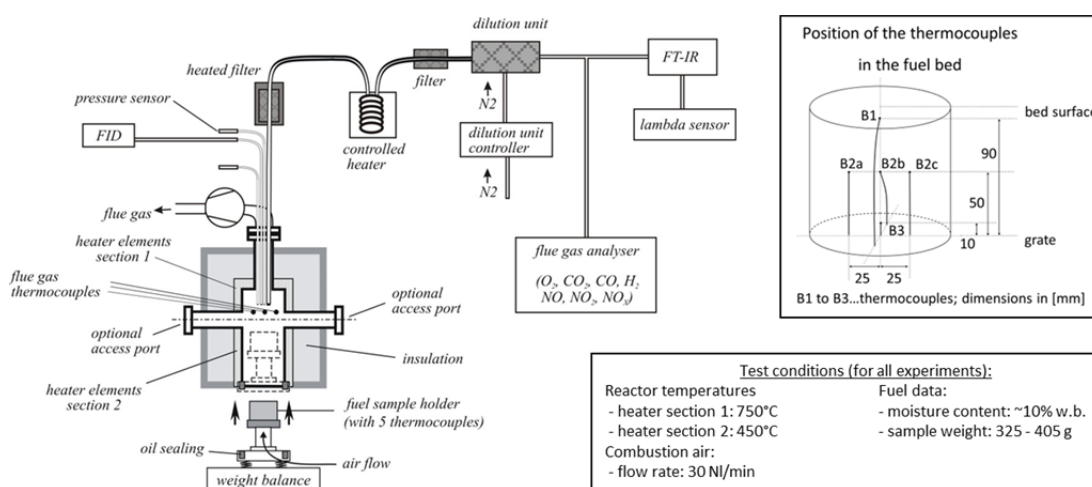


Figure 1: Scheme of the lab-scale reactor including measurement set-up (left), position of the thermocouples in the fuel bed (right up) and definition of common test conditions for all experiments (right below)

The fixed-bed lab-scale reactor (see Figure 1) is a discontinuously operated cylindrical pot furnace in which conditions well comparable to grate-fired plants (>50 kW thermal output) prevail. Biomass is introduced into a cylindrical holder (100 mm height and 95 mm inner diameter) placed inside an electrically heated cylindrical retort. The initial sample mass is, depending on the fuel density, commonly between 100 and 400 g. Dry air is introduced through a porous plate at the bottom of the fuel bed. Five thermo-couples measure the bed temperature at different positions (see Figure 1) and a balance records continuously the mass loss during the experiments. Flue gas composition is measured above the fuel bed by means of the following methods:

(I) Fourier transform infrared spectroscopy (FTIR) (Ansyco Series DX 4000) for CO₂, CO, H₂O, NO, NH₃, HCN, NO₂, N₂O, CH₄, light hydrocarbons and low condensable matters. (II) Flue gas analysis (Emerson NGA

2000), which employs paramagnetism for O₂, nondispersive infrared analysis (NDIR) for CO₂ and CO and heat conductivity for H₂. (III) Flame ionization detector (FID) (Messer Griesheim Austria Model VE7) for carbon amount in organic gaseous hydrocarbons (OGC). (VI) Chemiluminescence detector (CLD) (ECO Physics CLD 700 EL ht) for NO and NO_x. (IV) Wide band λ sensor for the estimation of O₂-concentration.

The reactor setup has been previously described in detail in the studies e.g. Sommersacher et al (2012) and Brunner et al. (2013). Furthermore, the procedure of the test runs, the evaluation of the results obtained and justifications about the results generated with this equipment is presented in these studies.

Experiments with the fixed-bed lab-scale reactor provide information concerning the combustion behavior, duration of drying, release of volatiles and char burnout. The release behavior of volatile and semi-volatile ash forming elements during the combustion process can be determined by calculation of mass and element balances for the ash forming elements. On the basis of this evaluation the emissions of aerosols, SO₂ and HCl can also be estimated. Optical evaluation of ash residues allows preliminary assessment of slagging tendencies.

3. Results

The chemical composition of the thermally untreated and torrefied fuels investigated is summarized in Table 1.

Table 1: Chemical composition of the fuels investigated

Explanation: raw ... thermally untreated fuel, torr ... torrefied fuel, m.c. ... moisture content, a.c. ... ash content, a.c. corr. ... corrected ash content, d.b. ... dry basis, f.b. ... fresh basis

		FR		SCL		GW		ST		SFH	
		raw	torr.	raw	torr.	raw	torr.	raw	torr.	raw	torr.
m.c.	wt% f.b.	13.5	5.1	10.9	4.1	12.9	4.9	8.6	3.3	11.4	6.7
a. c.	wt% d.b.	11.3	12.4	10.0	14.9	2.8	7.3	11.5	14.4	3.7	6.1
a. c. corr.	wt% d.b.	10.5	11.2	9.9	14.8	2.4	6.6	11.2	14.1	3.3	5.4
C	wt% d.b.	51.0	51.8	44.3	52.8	45.3	50.8	43.4	47.1	49.6	59.9
H	wt% d.b.	6.1	5.5	5.7	4.6	5.6	5.1	5.6	5.1	5.9	5.0
N	wt% d.b.	0.41	0.31	0.47	0.64	0.34	0.30	0.55	0.67	0.80	1.09
S	mg/kg d.b.	348	482	841	1,270	1,470	587	917	936	1,550	1,970
Cl	mg/kg d.b.	109	98.8	2,540	975	1,100	324	2,890	3,130	684	634
Si	mg/kg d.b.	3,490	14,700	19,400	28,900	21,000	25,400	31,200	39,900	661	1,480
Ca	mg/kg d.b.	5,460	7,580	4,200	8,210	13,000	15,000	3,770	4,770	3,420	5,670
Mg	mg/kg d.b.	690	1,570	1,780	2,980	2,620	2,990	1,450	1,830	1,940	3,440
Al	mg/kg d.b.	758	3,890	8,420	12,000	5,800	6,310	2,470	3,140	114	190
Fe	mg/kg d.b.	468	2,600	8,230	14,300	4,980	4,740	1,700	2,140	242	340
Mn	mg/kg d.b.	234	456	143	278	176	210	82.8	102	8.2	13.6
P	mg/kg d.b.	429	543	585	854	612	497	569	732	840	1,340
K	mg/kg d.b.	2,490	2,830	3,990	6,230	3,610	3,990	14,600	18,600	11,300	19,700
Na	mg/kg d.b.	173	846	37.5	3,010	1,580	1,500	362	480	39.0	64.4
Cu	mg/kg d.b.	3.7	13.9	8.9		67.7	41.6	4.1	5.1	10.9	19.3
Zn	mg/kg d.b.	57.7	78.3	18.0	45.2	103	112	8.3	13.3	10.8	39.9
Pb	mg/kg d.b.	1.62	15.5	2.3	15.7	26.8	19.6	10.0	10.0	6.1	10.0
Cd	mg/kg d.b.	0.23	0.38	0.16			0.37	0.59	0.70	0.37	0.16

The moisture content of the thermally untreated fuels is in the range of about 10 wt% fresh basis since all of these fuels are pelletized. The moisture content is significantly lower for the torrefied material (also in pelletized form) caused by the torrefaction process. The chemical analysis of the fuel shows (see Table 1) increased contents of C, ash, non-volatile ash-forming elements (Si, Ca, Mg and Al), and semi-volatile elements (e.g. K, Na, Zn, Pb, P) for torrefied material compared to the respective untreated fuel. Concerning Cl lower amounts are present for torrefied material compared to the untreated fuel in case of SCL and GW. The C enrichment leads to higher heating values for torrefied material, whereas the higher contents of certain ash forming elements like K and Si can worsen the combustion behavior.

Based on the chemical composition relevant fuel indices were calculated and summarized in Table 2. According to Table 2, the index K+Na+Zn+Pb generally indicates higher aerosol emissions for each torrefied

fuels compared to the respective untreated fuel. This means that higher aerosol emissions have to be expected for torrefied fuels. The molar Si/K ratio can give a first indication concerning the K release to the gas phase. Very high K release ratios have to be expected for SFH, which also results to high aerosol emissions. Significantly higher molar 2S/Cl ratios for torrefied materials in case of FR, SCL and SFH compared to untreated material were determined. For these fuels an improvement regarding high temperature corrosion can be expected. The values of the indices for prediction of the ash melting tendency (e.g. Si/[Ca+Mg] and [Si+P+K]/[Ca+Mg]) are almost equal for the untreated and the respective torrefied fuels. Here no significant change concerning the ash melting behavior between thermally untreated and torrefied fuel can be expected. For all fuels except for ST (untreated and torrefied) an unproblematic slagging tendency is very likely. For ST severe slagging tendency can be expected due to the high values of the ash melting indices.

Table 2: Values of relevant fuel indices for the fuels investigated

Explanation: raw ... thermally untreated fuel, torr ... torrefied fuel

		FR		SCL		GW		ST		SFH	
		raw	torr.	raw	torr.	raw	torr.	raw	torr.	raw	torr.
K+Na+Zn+Pb	mg/kg d.b.	2,722	3,770	4,048	9,301	5,320	5,622	14,980	19,103	11,356	19,814
Si/K	mol/mol	1.95	7.2	6.8	6.5	8.1	8.9	3.0	3.0	0.08	0.10
2S/Cl	mol/mol	7.1	10.8	0.73	2.9	3.0	4.0	0.70	0.66	5.0	6.9
Si/(Ca+Mg)	mol/mol	0.75	2.1	3.9	3.1	1.73	1.82	7.2	7.3	0.14	0.19
(Si+P+K)/(Ca+Mg)	mol/mol	1.23	2.4	4.6	3.7	1.33	1.40	9.8	9.9	2.1	2.1

The combustion tests in the fixed-bed lab-scale reactor showed that higher durations of the release of volatiles and charcoal gasification (drying phase excluded) for torrefied compared to the untreated fuels are needed to reach a complete burnout. The test results also indicate higher bed temperatures for torrefied fuels during char burnout. Longer residence times for complete burnout and higher bed temperatures have to be considered in the grate design (e.g. grate length, recommendation of flue gas recirculation - to keep the bed temperature below ash softening temperature, etc.).

By calculating mass and element balances the release ratios of relevant aerosol forming elements was calculated. On the basis of the release ratios of the main aerosol forming elements K, Na, Zn and P the potential for aerosol formation can be estimated. These elements are assumed to form solid K_2SO_4 , KCl, Na_2SO_4 , NaCl, ZnO and P_2O_5 . In addition, the formation of carbonates (K_2CO_3 and Na_2CO_3) is possible if there is not enough S and Cl to completely bind the K and Na as chlorides and sulfates. Usually, an excess of S and Cl to K and Na is present, and sulfate and chloride components are very likely. Assuming that the entire sulfur reacts with the K and Na present, the potential of maximum aerosol emissions can be calculated. This estimation of aerosol emissions does not take into account losses caused by the condensation of ash-forming vapors on the walls and the formation of deposits in boilers in large plants. In addition, the particle losses caused by reaction of ash-forming vapors with coarse fly ash particles and gaseous emissions of S (e.g., SO_x) and Cl (e.g., HCl) are not taken into account.

In Figure 2 the estimation of the maximum aerosol emissions is summarized.

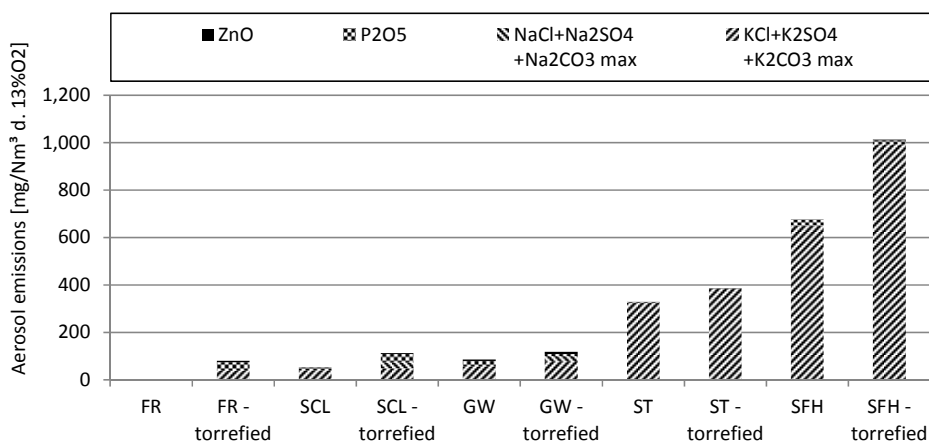


Figure2: Estimated aerosol emissions of the fuels tested

As shown in Figure 2, low aerosol emissions in the raw gas (<120 [mg/Nm³; 13 Vol.% O₂ dry flue gas]) can be expected for FR, SCL and GW for untreated and torrefied fuels. For ST (untreated and torrefied) the aerosol emissions are in the range of ~360 [mg/Nm³; 13 Vol.%O₂ dry flue gas]. High aerosol emissions (670 [mg/Nm³; 13 Vol.%O₂ dry flue gas]) are predicted for SFH - untreated, which are further elevated for the torrefied fuel. The high aerosol emissions for SFH are mainly caused by the high K content in the fuel and high evaporation rates of the alkali metal to the gas phase during combustion. As anticipated by the fuel index evaluation higher aerosol emissions have to be expected for torrefied fuels compared to the respective untreated fuel. Aerosol emissions are clearly dominated by K compounds which has also been reported in a former study by Sommersacher et al. (2013).

Based on the release data, an estimation of potential gaseous SO₂ and HCl emissions can be carried out. With the release data for S, Cl, K and Na and the assumption that the alkali metal compounds are formed 50% from sulfates and 50% from chlorides, the gaseous emissions of SO₂ and HCl can be assessed (S and Cl, which are not bonded to K and Na). This estimate does not take into account particle losses in boilers (e.g. reaction of ash forming vapors with coarse fly ash particles) as well as S and Cl, bound by Ca and Mg. Figure 3 shows the potential of the gaseous SO₂ and HCl emissions of all fuels tested.

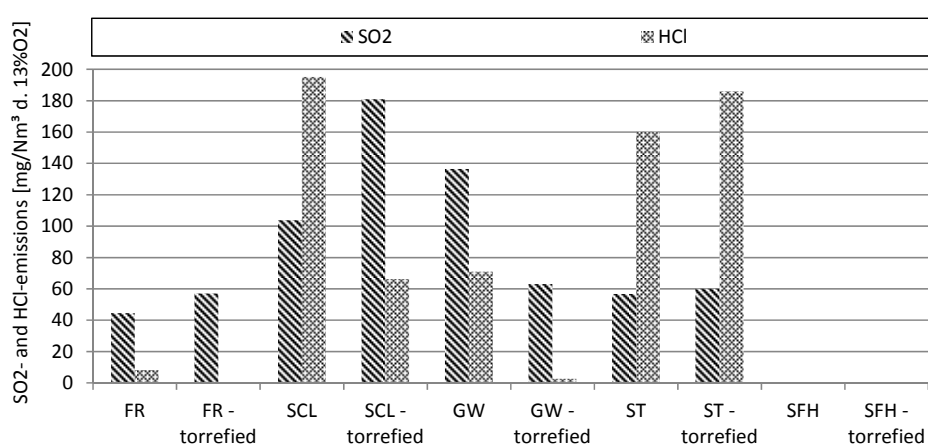


Figure 3: Estimated SO₂ and HCl emissions of fuels tested

According to the estimation of SO₂ and HCl emissions, summarized in Figure 3, elevated (<195 [mg/Nm³; 13 Vol.%O₂ dry flue gas]) SO_x and HCl emissions are very likely to appear for SCL (untreated and torrefied), GW (untreated) and ST (untreated and torrefied). In case of FR (untreated and torrefied) and GW (torrefied) low (<63 [mg/Nm³; 13 Vol.%O₂ dry flue gas]) and for SFH (untreated and torrefied) almost zero SO_x and HCl emissions can be expected. In case of SCL significantly higher S and lower Cl contents in the fuel (see Table 1) for the torrefied material compared to the thermally untreated material were determined. This causes the different SO_x and HCl emissions for thermally untreated in comparison to the torrefied material. For GW lower S and Cl concentrations were detected for torrefied material compared to the thermally untreated fuel which can explain the lower SO_x and HCl emissions for the torrefied material.

The ash residues after the combustion were loose with partly sintered particles for FR, SCL, GW (untreated and torrefied). For SFH a loose ash has been obtained for the untreated and torrefied fuel, whereas for ST already for the untreated fuel severe sintering and slagging occurred. The slagging tendency becomes even worse for the torrefied material in case of ST what is in good agreement with the respective fuel indices for prediction of the ash melting tendency.

4. Conclusions

Agricultural and waste-derived solid biomass fuels are potentially available in vast amounts for generation of sustainable energy. Their utilisation is widely hampered by their "difficult" combustion behaviour. Torrefaction (a mild form of pyrolysis) can improve the fuel properties of agricultural and waste-derived fuels, which are usually critical concerning a direct application in combustion plants.

In this study the combustion behaviour of five widely available untreated and torrefied biomass fuels - forest residues (FR), sugar cane leaves (SCL), green waste (GW), winter wheat straw (ST) and sunflower husks (SFH) - were investigated by the application of a multi-step laboratory approach (chemical analyses, fuel indices and fixed-bed lab-scale reactor combustion tests).

The combustion tests in the fixed-bed lab-scale reactor showed that higher durations of the release of volatiles and charcoal gasification for torrefied material compared to untreated material are needed to reach a complete burnout. The test results also indicate higher bed temperatures for torrefied fuels during the char burnout. The increased time to reach a complete burnout and the higher fuel bed temperatures have to be considered in the grate design (e.g. grate length, recommendation of flue gas recirculation - to keep the bed temperature below the ash melting starting temperature, etc.).

In general higher aerosol emissions have to be expected for torrefied fuels compared to thermally untreated biomass fuel. The aerosol emissions for SFH (untreated and torrefied) are (>670 [mg/Nm³; 13 Vol.% O₂ dry flue gas]) on a very high level. On the other side for this fuel very low SO_x- and HCl emissions can be expected. For ST (untreated and torrefied) the aerosol emissions are in the range of 360 [mg/Nm³; 13 Vol.% O₂ dry flue gas] and also relatively high HCl emissions (~170 [mg/Nm³; 13 Vol.% O₂ dry flue gas]) can occur. For FR, SCL and GW (untreated and torrefied) low aerosol emissions (<114 [mg/Nm³; 13 Vol.% O₂ dry flue gas]) can be expected. For these fuels only for SCL (untreated and torrefied) elevated SO_x and HCl emissions were predicted.

An improvement regarding high temperature corrosion can be expected for torrefied FR, SCL and SFH compared to untreated material. For FR, SCL, GW (untreated and torrefied) loose ash residues after the combustion with partly sintered particles were obtained. The ash residues were loose for SCL for the untreated and torrefied fuel, whereas for ST already for the untreated fuel serious sintering and slagging occurred. The slagging tendency becomes even worse for the torrefied material in case of ST.

The investigations revealed that SFH (untreated and torrefied) can be recommended as a fuel directly applicable in grate furnaces (also without flue gas recirculation) because of the low slagging tendency, but the high aerosol emission needs to be considered. Also FR, SCL and GW (untreated and torrefied) can be applied under consideration of the slagging tendency, whereas ST is not recommendable concerning the slagging tendency. However, for all fuels investigated the applicability in grate fired plants needs special considerations, since they do not behave like natural stem wood. Therefore, experimental investigations e.g. with a lab-scale combustion facility is suggested. Such combustion tests reveal the measures which must be taken to ensure an undisturbed and continuous operation in grate furnaces. According to such an investigation, the advantages of torrefied material (e.g., higher calorific value, better storage feasibility, etc.) can be fully exploited.

Acknowledgments

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Reference

- Bärenthaler G.; Zischka M.; Hanralsdsson C.; Obernberger I.; Determination of major and minor ash-forming elements in solid biofuels. *Biomass and Bioenergy* 2006, pp. 983-997
- Bergmann C.A. P., Kiel H.A. J., 2005, Torrefaction for biomass upgrading, 14th European Biomass Conference & Exhibition, Paris, France, 17-21 October 2005
- Bi X. T., Chen W-H., J. Peng, A state-of-the-art review of biomass torrefaction, densification and applications, 2015, *Renewable and Sustainable Energy Reviews* 44, 847-866.
- Brunner T., Biedermann F., Kanzian W., Evic N., Obernberger I., 2013, Advanced biomass fuel characterization based on tests with a specially designed lab-scale reactor. *Energy & Fuels* 27 (10), 5691-5698.
- Jones J., Ibrahim R. H.H., Darvella L. I., Williams A., 2013, Physicochemical characterisation of torrefied biomass, *Journal of Analytical and Applied Pyrolysis* 103, 21-30.
- Ptasinski K.J., van der Stelt M.J.C., Gerhauser H., Kiel J.H.A., 2011, Biomass upgrading by torrefaction for the production of biofuels: A review, *Biomass and Bioenergy* 35 (9), 3748-3762.
- Sommersacher P., Brunner T Obernberger I., 2012, Fuel indexes: A novel method for the evaluation of relevant combustion properties of new biomass fuels. *Energy & Fuels*, 26, 380-390.
- Sommersacher P, Brunner T, Obernberger I, Kienzl N, Kanzian W., 2013, Application of Novel and Advanced Fuel Characterization Tools for the Combustion Related Characterization of Different Wood/Kaolin and Straw/Kaolin Mixtures. *Energy Fuels* 27(9), 5192-5206.