LANDTECHNIK 70(2), 2015, 17–24
DOI:10.15150/lt.2015.2655

LANDTECHNIK
AGRICULTURAL ENGINEERING

Pelletizing of fuel blends mixed with lignin for energetic use in small scale combustion units

Frank Döhling, Nadja Weller

As part of future bio-economy concepts technical lignin, by-products derived from the pulp- and paper industry, may be recycled for further utilization. Via a specially developed process the liquid lignin lye can be converted into a solid state. The lignin granules were mixed with a blend of canola straw and peeled oat bran in different proportions (1–2 wt.-%) to adjust the characteristics of the mixtures for pelletizing. Combustion tests with the produced pellets were carried out in a small scale combustion unit (25 kW) showing that depending on the amount and kind of lignin CO-, SO_2- and particulate matter emissions are affected. Through further optimization of fuel mixtures and by means of secondary measures existing emission limits could be complied.

Keywords
Lignin, straw, pelletizing, combustion, alternative biofuel, energy related use, pellet boiler

Due to limited resources of fossil fuels such as mineral oil or coal the production of energy from renewable resources is becoming increasingly important. To ensure the aspired diversification of energy sources there is a growing demand for currently unused non-woody biomasses obtained as by-products of industrial processes. Technical lignin is a by-product of the pulp and paper industry and potentially eligible for an energetic utilization. The lignin is separated through the pulping of wood by adding various pulping chemicals. The lignin lyes, calcium ligninsulfonate (Ca-LS) from the sulfite pulping and kraft lignin (Kraft-L) from the sulfate pulping, were transformed into granulate through a special process (Narra et al. 2012a, Narra et al. 2012b).

For the energetic use in small scale combustion plants fuel in form of pellets is of great interest due to a potentially high added value for compacted fuels. However, since the pure lignin solid fuels show strong melting phenomena even at low temperatures occurring in a pellet press (usually 80 to 110 °C) their utilization as mono fuel for pellet-fired small scale combustion plants was not taken into consideration. Melting of the lignin granules in the auger system even before reaching the combustion chamber and thereby blocking or sticking to the screw conveyor is a possible risk. Both available lignin types were therefore added in different proportions to a blend of 80 wt.-% canola straw and 20 wt.-% peeled oat bran. The pelleting of the carrier material was improved by the use of lignin granules due to their binding qualities. The mixing ratio was determined according to the amounts of fuel available.

The use of solid biofuels is regulated by the German Federal Immission Control Act and the respective ordinance. The German First Ordinance on the Implementation of the Federal Immission Control Act (regulation on small and medium-sized combustion plants) (1st BImSchV 2010) applies

received 25 September 2014 | accepted 12 March 2015| published 27 March 2015

to the construction, design and operation of combustion systems that do not require approval under § 4th BImSchG. According to § 3 (1), 1st BImSchV lignin fuels could be classified as other sustainable resources, so called “no. 13 fuels”. However, it is unclear whether the responsible authority will follow this assignment, as technical lignin is the product of a chemical treatment of the raw material wood and may thus not be regarded as a renewable resource. If technical lignin is not classified as a “no. 13 fuel” it could only be utilized in combustion plants requiring a permit according to the 4th BImSchV (2013). The emission limits of the Technical Guidelines on Air Quality Control (TA Luft 2002) would then apply.

Fuel properties

For the practical use of solid biofuels a set of characteristics that influence the combustion process should be known. These include the chemical composition and the combustion-technical and physical-mechanical properties of the fuel. For non-woody pellets for the utilization in non-industrial small scale combustion plants (according to the 1st BImSchV) these properties are defined in the international standard DIN EN ISO 17225-6 (2014). By the use of lignin during pelleting additional bonds can be established whereby an improvement in the physical-mechanical properties of the pellets can be achieved (Berghel et al. 2013). Furthermore the two lignin granules have lower potassium (K), chlorine (Cl) and nitrogen (N) concentrations than canola straw and thus reduce the overall K, Cl and N concentrations in the fuel. The values listed in Table 1 were obtained according to the methods prescribed in DIN EN ISO 17225-1 (2014). The peeled oat bran (HSK) has good pelleting properties (Adapa et al. 2009) and due to low sulfur (S) and K contents an advantageous chemical composition regarding slagging behavior and SO_x emissions (Table 1). A basic blend (RS/HSK) consisting of 80 wt.-% canola straw (RS) and 20 wt.-% peeled oat bran (HSK) was prepared with both lignin types (Ca-LS and Kraft-L) added in proportions of 1 wt.-% and 2 wt.-%, respectively.

An increasing proportion of lignin alters the chemical composition of the fuels (Table 1). Compared to the basic blend (RS/HSK) the ash and S content is increased whereas the concentration of Cl, N and K is lowered. Hence reduction effects of HCl and NO_x emissions can be expected accompanied by increased SO_2 emissions. With respect to the reduced K content a partially positive impact on the ash melting behavior and slagging tendency as well as on the amount of particulate matter (PM) emissions is expected.

With regard to a normative classification of the produced blends, the specification of DIN EN ISO 17225-6 (2014) for herbaceous biomass (fruit biomass, aquatic biomass and blend and mixtures, class B) with respect to ash and water content, and N content could be complied. Contrary to expectations the calorific values of the individual raw materials show partly higher values than the lignin blended fuels. The interpretation of the results in terms of normative classification however remains unchanged. In one case and probably due to the inhomogeneity of the blend the Cl content is too high. The requirement for S is exceeded by all fuel blends due to the already high S content of the canola straw (RS) and in particular the high S content of the two lignin components.

Fuel-blend preparation and pelleting

The raw materials RS and HSK as well as the two lignin granules were ground in a first processing step. In order to prevent melting of the Ca-LS and gluing of the mill liquid nitrogen had to be applied during the grinding process to keep the temperature in the mill low. The materials were homogenized

Table 1: Selection of fuel characteristics of the raw materials

Parameter	Unit	RS	HSK	RS/HSK[3]	Kraft-L	Ca-LS	RS/HSK +1 % Kraft-L	RS/HSK +2 % Kraft-L	RS/HSK +1 % Ca-LS	RS/HSK +2 % Ca-LS	DIN EN ISO 17225-6[4]
Moisture	[wt.-%]	8.8	9.3	8.9	7.3	47.1	8.6	10.2	10.0	9.9	≤ 15
Net calorific value[1]	[MJ/kg dm]	16.6	16.8	16.7	15.8	8.6	17.1 (16.7)[5]	17.3 (16.7)	16.4 (16.6)	16.8 (16.5)	≥ 14.5
Ash content[2]	[wt.%/kg dm]	7.4	5.2	7.1	27.5	16.9	7.1	7.2	7.2	7.4	≤ 10
C	[wt.%/kg dm]	45	46.7	45.3	41.2	37.8	45.7	46.2	43.7	45.5	-
H	[wt.%/kg dm]	5.9	6.1	5.9	4.1	2.7	5.4	5.5	5.3	5.6	-
N	[wt.%/kg dm]	1.3	0.8	1.2	0.6	0.4	1.1	0.9	0.9	0.8	≤ 2.0
S	[wt.%/kg dm]	0.3	0.1	0.3	2.3	23.8	0.4	0.5	0.4	0.7	≤ 0.3
Cl	[wt.%/kg dm]	0.5	0.1	0.4	0.1	n.n.	0.3	0.4	0.3	0.3	≤ 0.3
K	[mg/kg dm]	19,000	8,657	16,931	8,650	852	15,400	14,600	13,700	13,000	-
Fines < 3,15 mm	[wt.-%]	-	-	-	-	-	6.6	5.6	0.8	3.7	≤ 3
Durability	[wt.-%]	-	-	-	-	-	92.2	89.4	91.4	93.9	≥ 96
Bulk density	[kg/m³]	-	-	-	-	-	505	445	476	531	≥ 600

[1] H, N, S, corrected..
[2] At 550 °C, except Kraft-L (815°C).
[3] 80 wt% RS + 20 wt% HSK, calculated values.
[4] herbaceous biomass, fruit biomass, aquatic biomass and blends and mixtures, class B.
[5] Data in brackets are calculated.
dm = dry matter
Ca-LS = Calciumligninsulfonat
Kraft-L = Kraftlignin
n.n. = below limit of detection
RS/HSK =Blend of canola straw and peeled oat bran (calculated values)
RS/HSK+1% Kraft-L resp. RS/HSK+2% Kraft-L = Blend of canola straw and peeled oat bran with 1 wt% Kraftlignin
RS/HSK+1% Ca-LS resp. RS/HSK+2% Ca-LS = Blend of canola straw and peeled oat bran with 1 or 2 wt% Ca-Ligninsulfonate

prior to pelleting and the water content was set to 16.5 wt.-%. The pelleting was performed by a commissioned company. The blended fuel pellets showed deviations from the normative specifications regarding their physical-mechanical properties while the physical-mechanical properties of RS and HSK could not be determined due to low fuel availability. Since the availability of the novel prepared lignin granules was scarce the pelleting was carried out using default parameter settings. A further optimization of the pelleting process could not be realized. With about 6 wt.-% the Kraft-L added pellet batches have significantly higher proportion of fines and the mechanical durability on average is slightly lower than that of Ca-LS batches. The pellets had a wide range of bulk densities scattering around 500 kg/m^3. No dependency of this pattern could be found with the amount of lignin present in the pellet. With regard to the normative specification of the physical-mechanical properties (bulk density, fines, mechanical durability) specific adjustments of the pelletizing process parameters are necessary to meet the standard requirements. The influence of both lignin granules to the required energy input for pelleting could not be measured at the commissioned company. In preliminary tests at a laboratory press Lu et al . (2014) showed a positive effect on the pelleting process by the addition of lignin sulfonate to wheat straw and wood. With an optimized setting of the pellet press it can be assumed that the effect is also observable for the fuels used in this study.

Combustion tests

The combustion tests were carried out with a pellet boiler suitable for pelletized herbaceous fuels with a nominal heat output of 25 kW at full load conditions. The relevant gas emissions (CO, NO_x, SO_2, HCl) were measured with an FTIR spectrometer.

The measurements of PM were carried out according to the German VDI 2066 Part 1 (2006). Figure 1 shows the total average of the gas concentrations recorded as 15 minute average values (VMW) throughout the combustion test. Maximum and minimum concentration values refer to the range of VMW. A VMW is based on 90 individual measurements with a measuring time of five seconds each. All measurements were made at steady state. In addition, a visual assessment of the combustion behavior was made. In general the amount of emissions depends not only on the fuel used but also on plant-specific factors such as combustion chamber geometry, air flow and grate technology. The present data therefore only apply to the one small scale combustion plant used for the measurements.

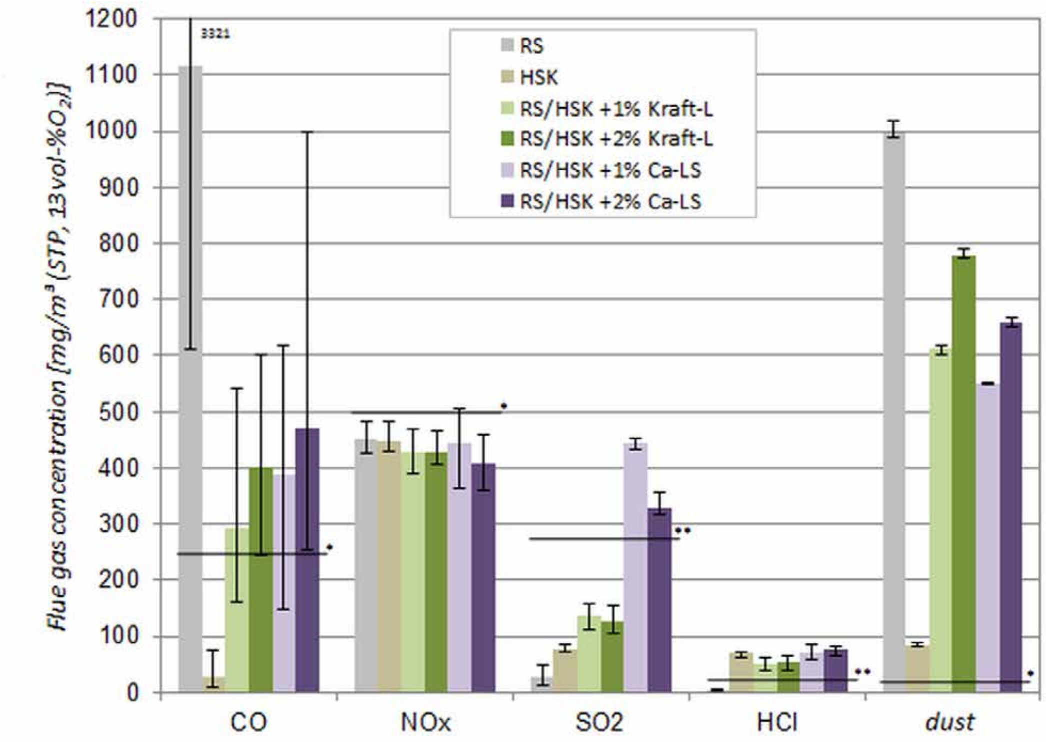

Figure 1: Gaseous and particulate emissions of four fuel blends, RS/HSK - Blend of canola straw and peeled oat bran, Kraft-L - Kraftlignin, Ca-LS - Calciumligninsulfonate, error bars represent minimum and maximum values,
* - emission thresholds according to the German First Ordinance on the Implementation of the Federal Immission Control Act (CO- and NOx-threshold according to type test, dust threshold according to recurrent measurements
** - emission thresholds according to the Technical Guidelines on Air Quality Control (TA Luft), thresholds according to the Technical Guidelines on Air Quality Control (TA Luft) were converted from a reference oxygen content of 11 vol.% to 13 vol.%

Nevertheless statements about the general suitability of the examined fuels and the applicability to larger systems can be derived.

Both reference fuels RS and HSK show significant differences in the concentration of the emitted pollutants except for NO_x (Figure 1) and their combustion behavior was very different. The canola straw pellets (RS) showed a very intense slagging whereas the slagging of the peeled oat bran pellets (HSK) was comparably small. Since K is also an important component of the mineral dust fraction PM emissions (dust) from RS pellets are remarkably high. Furthermore, the strong slagging prevents a complete gas burnout, hence CO emissions are high. Concerning HCl and SO_2 emissions pellets from RS show a lower emission level than the HSK pellets. The combustion and emission behavior of the RS/HSK blend could not be examined due to limited availability of the raw material.

For all fuel blends containing lignin a stable combustion with a pronounced flame pattern could be observed. The variants with higher lignin contents showed a stronger tendency to sintering and slagging of bottom ash. However, the agglomerates formed were friable, porous and of low mechanical strength, hence they were mostly destroyed by the grate movement or by falling into the ash pan. In particular the use of HSK seems to have a positive impact on sintering and slagging while the lignin increases the risk. Despite a visually satisfactory combustion process, the CO limit value for a type test (1^{st} BImSchV 2010) of any of the fuel blends containing lignin was exceeded. In general CO emissions are subjected to substantial fluctuations resulting from the break-up of slagged ash agglomerates. The fuels blended with Ca-LS have, depending on the lignin concentration, higher CO emissions than the fuel blended with Kraft-L (Figure 1). For both lignin blends an increased proportion of 2% lignin leads to an increase in CO emissions by 80 to 100 mg/m³. This increase is probably caused by an incremental slagging restraining the gas burnout. Interestingly, the slagging cannot be clearly assigned to the K content of the fuels as this decreases with increasing lignin proportion, therefore it can be assumed that slagging is influenced by other parameters. In contrast to the CO emissions the NO_x limit for a type test according to 1^{st} BImSchV (2010) for all four fuel blends is complied. The averaged values, 400 to 450 mg/m³ are at a similar level.

No difference in SO_2 emissions was observed between fuel blends with Kraft-L (137 and 126 mg/m³). The SO_2 emissions from Ca-LS fuel blends with values of 329 and 443 mg/m³ were significantly higher and a strong difference between the 1% Ca-LS and 2% Ca-LS variant was observed. This difference cannot be attributed to the S content of the fuels since the S content of 2% Ca-LS variant is higher than the S content of 1% Ca-LS. The release of S during combustion is not only dependent on the absolute content in the fuel but also the remaining mineral fuel composition. It can thus be assumed that S was primarily emitted in particulate form and additionally enriched in the bottom ash (Vainio et al. 2013). With regard to the SO_2 emissions from biomass combustion no limit in the 1^{st} BImSchV (2010) is determined. The TA Luft (2002), however, sets a limit of the equivalent of 280 mg/m³ (based on 13 vol.-% O_2 in the exhaust gas) which could be complied by the two Kraft-L enriched fuels and has been exceeded by the two Ca-LS enriched fuels.

The HCl emissions of all four blended fuels were more than double as high as the required 24 mg/m³ limit of TA Luft (2002) (based on 13 vol% O_2). The HCl emissions of Ca-LS blended fuels were about 20 mg/m³ higher than that of the Kraft-L variants. In contrast, the Cl content of the fuels had little variation so that the reduced HCl emissions of Kraft-L fuels can be explained by an increased release of Cl in a particulate form such as KCl (Sommersacher et al. 2012, Johansen et al. 2013). Also, the higher K contents of Kraft-L enriched fuels support this. The PM emissions ranging from 551 mg/m³ to 779 mg/m³ can be considered as very high. This was to be expected since the sum of the aerosol forming elements (S, Cl, K, Na, Pb, Zn) especially characterized by a high K content was an indication for that (Obernberger 2014). The emissions of fuels with Kraft-L are significantly higher than for fuels with Ca-LS when comparing the respective 1 wt% and 2 wt% blends. An increase in the lignin component to 2 wt% results in a sharp increase in dust emissions by more than 100 mg/m³. Hence an appropriate filtering technique for observing the limits should be installed.

Conclusions

Lignin, in the form of Kraft-L and Ca-LS, is not suitable as mono fuel for utilization in market available pellet boilers according to the 1st BImSchV (2010). The strong softening tendencies of the material at relatively low process temperatures do not allow compaction. Using it without compaction would bare the risk of melting and gluing to the conveyer screw before reaching the combustion chamber. The utilization as a non-pelletized mono fuel in an especially designed combustion system (not necessarily a small scale combustion unit) may be feasible, however, was not considered as part of the project. In general, combustion plants whose conceptual design is based on the 17th BImSchV (2013) (Ordinance on the incineration and co-incineration of waste) may be considered.

However, the use of lignin blended fuels in small scale combustion units can create a high added value. In the combustion experiments, both Kraft-L and Ca-LS could be made available for an energetic use by blending them in proportions of 1 or 2 wt.-% to a basis mixture of 80 wt.-% RS and 20 wt.-% HSK. The physical-mechanical requirements of DIN EN ISO 17225-6 (2014) could not be complied and further compaction tests would be necessary but not possible in the present work. However, the authors assume that with further targeted tests the production of standardized pellets containing lignin is possible. The calorific value and the ash content of the lignin enriched pellets were similar to those of straw fuels. With respect to critical emission related elements the high S content is particularly noteworthy. Furthermore, also the high content of K has a negative effect on the combustion and slagging behavior as well as the amount of PM emissions. Also in terms of chemical properties the requirements of DIN EN ISO 17225-6 (2014) were only partially met. This is also reflected in the results of the combustion tests. The SO_2 and PM emissions were strongly dependent on the type of lignin. The NO_x emissions were of no concern. From a legal perspective, it is questionable whether the licensing authority classifies an RS/HSK lignin blended fuel as a renewable raw material due to the chemical pre-treatment of the lignin. Alternatively a classification as an additive should be considered. Furthermore, the stringent requirements of the type test, especially with regard to the CO and PM emissions as well as the additional measurement of PCDD/F cannot be complied without further measures of fuel optimization and the use of a suitable deposition technique (Obernberger 2005). Therefore, the application of the considered lignin enriched fuels in small scale combustion plants without further pretreatment and without appropriate secondary reduction measures is questionable. The application in licensed installations (> 100 kW to < 50 MW) should be considered as an option since the use of specified equipment in accordance with powerful filtering technique (e.g. cyclone, electric or fabric filters) is mandatory.

References

1. BImSchV (2010): Erste Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes – Verordnung über kleine und mittlere Feuerungsanlagen vom 26. Januar 2010, BGBl. I S. 38

4. BImSchV (2013): Vierte Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes – Verordnung über genehmigungsbedürftige Anlagen vom 2. Mai 2013, BGBl. I S. 973, 3756

17. BImSchV (2013): Siebzehnte Verordnung zur Durchführung des Bundes-Immissionsschutzgesetzes (Verordnung über die Verbrennung und die Mitverbrennung von Abfällen – 17. BImSchV) vom 2. Mai 2013, BGBl. I S. 1021, 1044, 3754

Adapa, P.; Tabil, L.; Schoenau, G. (2009): Compaction characteristics of barley, canola, oat and wheat straw. Biosystems Engineering 104(3), pp. 335–344

Berghel, J.; Frodeson, S.; Grantström, K.; Renström, R.; Ståhl, M.; Nordgren, D.; Tomani, P. (2013): The effects of kraft lignin additives on wood fuel pellet quality, energy use and shelf life. Fuel Processing Technology 112, pp. 64–69

DIN EN ISO 17225-1 (2014): Biogene Festbrennstoffe – Brennstoffspezifikationen und -klassen – Teil 1: Allgemeine Anforderungen, Deutsche Fassung EN ISO 17225-1:2014, Beuth Verlag

DIN EN ISO 17225-6 (2014): Biogene Festbrennstoffe – Brennstoffspezifikationen und -klassen – Teil 6: Klassifizierung von nicht-holzartigen Pellets (ISO 17225-6:2014); Deutsche Fassung EN ISO 17225-6:2014, Beuth Verlag

Johansen, J. M.; Aho, M.; Paakkinen, K.; Taipale, R.; Egsgaard, H.; Jakobsen, J. G.; Frandsen, F. J.; Glarborg, P. (2013): Release of K, Cl, and S during combustion and co-combustion with wood of high-chlorine biomass in bench and pilot scale beds. Proceedings of the Combustion Institute Vol. 34, pp. 2363-2372

Lu, D.; Tabil, L. G.; Wang, D.; Wang, G.; Emami, S. M. (2014): Experimental trials to make wheat straw pellets with wood residue and binders. Biomass and Bioenergy 69, pp. 287–296

Narra, S.; Logsch, F.; Glaser, C.; Ay, P. (2012a): Sludge-pelleting of paper and pulp residue. XXVI International Mineral Processing Congress “Innovative Processing for Sustainable Growth”, New Delhi, India, 24-28 September 2012, pp. 03723-03739

Narra, S.; Ay, P.; Glaser, C.; Logsch, F. (2012b): Verfahren zur Herstellung von wasserunlöslichen Lignin-Agglomeraten; DE102012102327B3

Obernberger, I. (2005): Nutzung fester Biomasse in Verbrennungsanlagen unter besonderer Berücksichtigung des Verhaltens aschebildender Elemente, Schriftenreihe Thermische Biomassenutzung, Technische Universität Graz, Institut für Ressourcenschonende und Nachhaltige Systeme, Band 1, 4. Auflage

Obernberger, I. (2014): Strategy for the application of novel characterization methods for biomass fuels: Case study of straw. Energy & Fuels 28(2), pp. 1041-1052

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(1), pp. 380–390

TA Luft (2002): Erste Allgemeine Verwaltungsvorschrift zum Bundes-Immissionsschutzgesetz (Technische Anleitung zur Reinhaltung der Luft - TA Luft)

Vainio, E.; Yrjas, P.; Zevenhoven, M.; Brink, A.; Laurén, T.; Hupa, M. ; Kajolinna, T.; Vesala, H. (2013): The fate of chlorine, sulfur, and potassium during co-combustion of bark, sludge, and solid recovered fuel in an industrial scale BFB boiler. Fuel Processing Technology 105, pp. 59–68

VDI 2066, Blatt 1 (2006): Messen von Partikeln – Staubmessung in strömenden Gasen – Gravimetrische Bestimmung der Staubbeladung. In: VDI/DIN-Handbuch Reinhaltung der Luft, Band 4: Analysen- und Messverfahren, Hrsg.: Verein Deutscher Ingenieure

Authors

Dipl.-Ing. Nadja Weller and **Dipl.-Ing. Frank Döhling** are scientific staff in the workgroup Innovative Solid Fuels of the department Thermo-Chemical Conversion (head of department: Dr.-Ing. Volker Lenz) at the Deutsches Biomasseforschungszentrum, Torgauer Straße 116, 04347 Leipzig, E-Mail: Frank.Doehling@dbfz.de.

Acknowledgements

The presented results are in part the work of the joint project “LignoBioFuel - Entwicklung eines Verfahrens zur Herstellung von stofflich und energetisch nutzbaren Bioagglomeraten auf der Basis von Lignin” (FZK 03KB58) funded by the German Federal Ministry of Economic Affairs and Energy pursuant to a decision of the German Bundestag.