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Physical characterisation and chemical composition of densified biomass fuels with regard to their combustion behaviour

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Abstract

With respect to the use of densified biomass fuels in fully automatic heating systems for the residential sector a high quality of these fuels is required. Several European countries already have implemented standards for such fuels. In other countries such standards are in preparation or planned. Furthermore, in some countries also standards from associations are existing (e.g. from the Austrian Pellets Association). In addition to these national standards, European standards for solid biomass fuels are under development. For producers of densified biomass fuels, especially for pellet producers, it is therefore very important to produce high-quality fuels keeping the limiting values of the standards addressed. However, in this context it has to be considered that as a high fuel quality as is necessary for the combustion of densified biomass fuels in automatic small-scale furnaces is not necessary if these fuels are used in larger industrial furnaces as they are equipped with more sophisticated flue gas cleaning, combustion and process control systems. Two pellet qualities, one for industrial and one for small-scale consumers seem to be more meaningful.

Within the framework of the EU-ALTENER-project "An Integrated European Market for Densified Biomass Fuels (INDEBIF)" a questionnaire survey of European producers of densified biomass fuels was performed. In this connection the possibility was offered to the producers to participate in an analysis programme with their fuels. An overview was obtained of the qualities of densified biomass fuels offered in the European market, covering pellets and briquettes from Austria, Italy, Sweden, Spain, Norway and the Czech Republic.

The parameters analysed were the dimensions of the fuels, the bulk and the particle density, the water and the ash content, the gross and the net calorific value, the abrasion, the content of starch (as an indication for the use of biological binding agents), the concentrations of C, H, N, S, Cl, K as well as of the heavy metals Cd, Pb, Zn, Cr, Cu, As and Hg. These parameters have been chosen following the Austrian, German, Swiss and Swedish standards for densified biomass fuels.

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The results showed that a majority of the participating producers produce fuels of high quality. However, wood pellets of some producers show a high abrasion, one of the most important quality parameters for pellets. An increased amount of fines often causes failures in the feeding systems used in the residential heating sector. In order to decrease abrasion, the addition of small amounts of biological binding agents (e.g. maize or rye) is possible. This kind of additive is most common in Austria.

Moreover, some producers obviously use not only chemically untreated raw materials or additives, which increase the content of pollutants. Such fuels cause problems regarding emissions, deposit formation and corrosion. Emission problems are expected due to increased contents of N, Cl, S as well as heavy metals. Increased concentrations of heavy metals additionally contaminate the ash, increased Cl concentrations raise the risk of corrosion. Moreover, an increased content of K has a negative effect on the ash melting behaviour and causes higher aerosol formation, which enhances deposit formation and particulate emissions.

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Keywords: Pellets; Biomass; Combustion; Fuel quality; Physical characterisation; Chemical composition

1. Introduction and objectives

An analysis programme of densified biomass fuels has been carried out within the framework of the EU-ALTENER-project "An Integrated European Market for Densified Biomass Fuels (IN-DEBIF)" [1]. The parameters tested within the framework of this analysis programme (see Table 1) are diameter, length, bulk and particle density, water content, ash content, gross and net calorific value, energy density, starch content, abrasion, the contents of C, H, N, S, Cl, K and the contents of the heavy metals Cd, Pb, Zn, Cr, Cu, As and Hg. The parameters have been chosen following the Austrian, German, Swiss and Swedish standards for densified biomass fuels and are discussed in detail in Section 3.

The starch content in wood pellets is not regulated by the standards mentioned above. There are regulations regarding the use of biological additives for the production of densified biomass fuels, however. The kind and amount of such biological additives used must usually be stated in an appropriate and comprehensible way. The starch content is a good indication for the use of biological additives and has therefore been analysed. The method used is described in Section 2.12.

In total 38 samples of densified biomass fuels were obtained within the framework of the analysis programme. The main share of densified biomass fuels formed wood pellets (60%), followed by wood briquettes (30%). Furthermore, Table 1

Parameters tested within the framework of the analysis programme and corresponding regulations in several European standards

Parameter Re	Regulation in					
Ö! M	NORM 7135	PVA	SS 187120	DIN 51731/SN 1666000		
Diameter ×		х	×	×		
Length ×		×	×	x		
Bulk density ^a			×			
Particle density ×		×		х		
Water content ×		×	×	×		
Ash content ×		×	×	×		
GCV						
NCV ×		×	×	×		
Energy density ^a						
Starch content ^a ×		×	×			
Abrasion ^a ×		×	×			
C (GCV calculation)						
H (GCV calculation)						
N (GCV calculation) \times		×		х		
S ×		×	×	х		
Cl ×		×	×	х		
K						
Cd		×		х		
Pb		×		х		
Zn		×		х		
Cr		×		х		
Cu		×		×		
As		×		×		
Hg		х		×		

Explanations: PVA, standard of the Austrian Pellets Association (Pelletsverband Austria); GCV, gross calorific value; NCV, net calorific value.

^aOnly for pellets; data source [2-5,18].

two samples each of bark briquettes and straw pellets were obtained. The samples have been collected from 30 different producers of densified biomass fuels located in six European countries, i.e. Austria, Spain, Sweden, Italy, the Czech Republic and Norway. Six of these participating producers took part with more than one sample, e.g. wood pellets and wood briquettes or different qualities of wood pellets. The project partner countries (Austria, Sweden and Spain) provided 79% of the samples.

The objectives of the work were to get an overview about the physical characterisation and the chemical composition of densified biomass fuels from different European countries and producers. Moreover, the possible variation of the quality of densified biomass fuels as well as a comparison between the analyses results achieved and the limiting values defined in the different national standards for densified biomass fuels have been indicated. Finally, possible relations between different parameters investigated as well as the relevance/influence of these parameters on the combustion process are discussed.

2. Methods

Most measurements and analyses were performed from the Institute of Chemical Engineering Fundamentals and Plant Engineering, Graz University of Technology in co-operation with the Institute of Chemistry, University of Graz. The gross calorific values were measured by the Federal Institute of Agricultural Engineering in Wieselburg (Austria). The contents of the elements C, H and N were determined by the Institute of Chemistry, University of Graz.

2.1. Determination of dimensions

The dimensions of the pellets were determined according to ÖNORM M 7135 by measuring the length and the diameter of 20 randomly selected individual pellets per sample. From these 20 values the average values for the diameter and the length as well as the ratio length/diameter were calculated. The dimensions of the briquettes were determined by measuring diameter and length of one briquette from each sample.

2.2. Determination of bulk density

The volume and the weight of a pellet sample have to be measured in order to be able to determine the bulk density. Weight measurements were performed with a normal laboratory balance, the volume of the samples was determined by using a graduated cylinder. The average bulk density was calculated from three measurement series per sample.

2.3. Determination of particle density

The particle density of pellets was determined according to ÖNORM M 7135 by measuring dimensions and weight of at least 15 randomly selected individual pellets per sample. The calculated particle density represents the average value of these measurements. The particle density of the briquettes was determined by measuring dimensions and weight of one briquette per sample.

2.4. Determination of water content

The water content was measured by weighing a fuel sample (about 100 g (w.b.)) before and after drying at 105° C according to ÖNORM G 1074.

2.5. Determination of ash content

The ash content was measured by applying two different methods (loss of ignition at 550°C according to SS 187171 and at 815°C according to DIN 51719) in order to be able to evaluate the differences between them.

2.6. Determination of K, Cu, Zn, As, Cr, Pb and Cd

- Digestion: Multi-step pressurised digestion with HNO₃ (65%)/HF/H₃BO₃.
- Detection: K with flame atomic adsorption spectrometry (FAAS), all other elements with inductively coupled plasma mass spectrometry (ICP-MS).

2.7. Determination of Hg

- Digestion: Pressurised digestion with HNO₃ (analytically pure) in a crystal vessel.
- Detection: ICP-MS.

2.8. Determination of S and Cl

- Digestion: Bomb combustion in oxygen; absorption in NaOH (0.05 M).
- Detection: High-performance liquid chromatography (HPLC).

2.9. Determination of C, H and N

Combustion and following separation by a gas chromatograph, detection of the elements by an elementary analyser (Erba EA 1108). Two determinations per sample were performed according to ÖNORM G 1071, G 1072 and G 1073.

2.10. Determination of the gross calorific value

Combustion of the sample under specific conditions in a bomb calorimeter according to DIN 51900.

2.11. Detection of the abrasion

The Ligno Tester LT II (manufacturer: Borregaard Lignotech) or an equivalent tester must be used for the determination of the abrasion according to ÖNORM M 7135. In this tester about 100 g pellets are exposed to an airstream of 70 mbar for 60 s. Before the test the pellets must be free of fines and therefore the share of fines must be separated from the pellets by manual sieving using a 3.15 mm sieve. After the abrasion test the pellets are weighed and the share of fines (abrasion) can be calculated. The share of fines of a sample is calculated as an average value from five single abrasion measurements performed.

2.12. Detection of the starch content

An unvalidated method is available to determine starch in pellets (by iodine/starch complex formation and photometry). The verification was made by analysing both, raw material and pellets (with and without addition of starch), with the result that an unambiguous qualitative detection and also a quantitative determination of starch in pellets are possible. As a next step it is recommended to validate the method applied.

3. Results for different parameters analysed and their influence on the combustion behaviour

3.1. Overview

The results of the analyses of wood pellets and wood briquettes are shown as average, minimum and maximum values including the standard deviation in Tables 2 and 3. Moreover, the guiding values according to the Austrian standard ÖNORM M 7135 (compressed wood or compressed bark in natural state—pellets and briquettes) [2], the German standard DIN 51731 (testing of solid fuels—compressed untreated wood) [3], the Swiss standard SN 166000 (testing of solid fuels—compressed untreated wood) [4], the Swedish standard SS 187120 (biofuels and peat—fuel pellets—classification) [5] and the standard of the Austrian Pellets Association are also shown in these tables.

Due to the small number of samples of straw pellets and bark briquettes, these respective results are shown as single values for each sample (see Table 4).

3.2. Dimensions of pellets

The main part of the pellet samples obtained is pellets with a diameter of about 6 mm. These pellets are mainly from Austria, Italy and Spain. Pellets with a diameter of 8 mm are most common in Sweden. In addition, pellets with a diameter of about 10 mm have also been obtained.

An important parameter in this context is the ratio length/diameter, which is in particular of great relevance, if pneumatic feeding systems are used, due to the fact that even a single long pellet is

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Table 2Results of the analyses of wood pellets

Parameter	Unit	Average value	Minimum	Maximum	Standard deviation	Guiding value
Diameter D	Mm	7.1	5.9	10.2	1.2	4–10 ^a
Length	Mm	16.1	8.6	29.6	5.2	$< 5 \times D^{a,b}$
Bulk density	kg/m ³	591	520	640	39	$> 600^{\circ}$
Particle density	kg/dm ³	1.18	1.03	1.30	0.06	$> 1.12^{a}$
Water content	wt% (w.b.)	7.7	5.7	9.0	0.9	<10 ^{a,b,c}
Ash content	wt% (d.b.)	0.51	0.17	1.61	0.30	< 0.5 ^{a,b}
Abrasion	wt% (w.b.)	4.05	0.58	12.53	3.52	<2.3 ^{a,b}
Starch content	wt% (d.b.)	0.22	0.00	1.25	0.42	
GCV	MJ/kg (d.b.)	20.3	19.8	20.7	0.2	
NCV	MJ/kg (d.b.)	19.0	18.6	19.4	0.2	>18.0 ^{a,b}
С	wt% (d.b.)	50.3	49.5	51.9	0.6	
H	wt% (d.b.)	5.7	5.5	6.1	0.2	
Ν	wt% (d.b.)	0.22	0.20	0.64	0.10	< 0.3 ^{a,d}
S	mg/kg (d.b.)	278	52	1922	454	<400 ^{a,b}
Cl	mg/kg (d.b.)	48	10	126	27	<200 ^{a,b}
К	mg/kg (d.b.)	493	302	1180	252	
Cd	mg/kg (d.b.)	0.14	0.06	0.20	0.03	< 0.5 ^{d,b}
Pb	mg/kg (d.b.)	0.43	0.07	2.19	0.44	<10 ^{d,b}
Zn	mg/kg (d.b.)	13.2	9.3	25.4	3.6	<100 ^{d,b}
Cr	mg/kg (d.b.)	0.6	0.1	3.3	0.8	<8 ^{d,b}
Cu	mg/kg (d.b.)	1.1	0.7	2.7	0.5	<5 ^{d,b}

Note: The results are based on the analyses of 21 wood pellet samples.

^aAccording to ÖNORM M 7135.

^bAccording to the standard of the Austrian Pellets Association.

^cAccording to SS 187120.

^dAccording to DIN 51731 and SN 166000.

able to block the transport pipe of such systems. According to ÖNORM M 7135 and the standard of the Austrian Pellets Association this ratio should be less than 5. As shown in Fig. 1, all samples analysed meet this guiding value. Italian samples show the highest and Swedish samples the lowest ratio.

The particle size and the range of variation in the particle size influence the selection of the conveying systems as well as the combustion behaviour of pellets. Pellets are very homogeneous, e.g. compared to wood chips, and available in the size recommended by smallscale pellet furnace manufacturers. The development of automatic biomass heating systems with a comfort similar to oil or gas fired heating systems was only possible because of the market introduction of pellets with uniform size and shape.

3.3. Bulk density of pellets

The bulk density of wood pellets varies in a range between 520 and 640 kg/m^3 (see Table 2). A guiding value for the bulk density of wood pellets is specified in the Swedish standard SS 187120 at 600 kg/m^3 . The results show that many of samples analysed have a bulk density below this value. For the samples from Italy, Sweden and Spain even the average value is below this guiding value. The highest bulk density (660 kg/m^3) was reached by a straw pellet sample (see Table 4).

In this context it is important to point out that none of the wood pellet samples reached a bulk density of 650 kg/m^3 , which is often stated as a design value by wood pellet producers. A low bulk density has a negative effect on the energy density and therefore also on the transportation costs and

Table 3			
Results of	the analyse	s of wood	briquettes

Parameter	Unit	Average value	Minimum	Maximum	Standard deviation	Guiding value
Diameter D	mm	79	52	93	18	40–120 ^a
Length	mm	250	74	313	77	$< 400^{a}$
Particle density	kg/dm ³	1.06	0.81	1.26	0.14	> 1.0
Water content	wt% (w.b.)	8.0	6.4	10.4	1.2	<10 ^a
Ash content	wt% (d.b.)	0.88	0.21	1.88	0.63	$< 0.5^{a}$
GCV	MJ/kg (d.b.)	20.2	19.9	20.5	0.2	
NCV	MJ/kg (d.b.)	18.9	18.3	19.4	0.3	$> 18.0^{a}$
С	wt% (d.b.)	50.5	49.4	52.2	1.0	
Н	wt% (d.b.)	5.6	5.5	5.8	0.1	
Ν	wt% (d.b.)	0.33	0.20	0.95	0.27	< 0.3 ^{a,b}
S	mg/kg (d.b.)	134	35	271	76	$< 400^{a}$
Cl	mg/kg (d.b.)	153	37	384	120	$< 200^{a}$
K	mg/kg (d.b.)	643	84	1380	419	
Cd	mg/kg (d.b.)	0.13	0.01	0.43	0.12	< 0.5 ^b
Pb	mg/kg (d.b.)	1.81	0.16	7.69	2.28	<10 ^b
Zn	mg/kg (d.b.)	12.4	1.7	23.2	6.1	<100 ^b
Cr	mg/kg (d.b.)	0.7	0.1	2.2	0.7	<8 ^b
Cu	mg/kg (d.b.)	1.4	0.7	2.8	0.7	<5 ^b

Note: The results are based on the analyses of 10 wood briquette samples.

^aAccording to ÖNORM M 7135.

^bAccording to DIN 51731 and SN 166000.

the storage capacity both for the pellet producer and the end user.

3.4. Particle density

The wood pellet samples meet the guiding value of 1.12 kg/dm^3 according to the Austrian standard, except for one sample from Italy. The variation in the particle density of wood pellets was between 1.03 and 1.30 kg/dm^3 (see Table 2). The Italian wood pellet samples show a very high variation in particle density but also have the highest average value. Regarding the other countries, none of the pellet samples were below the guiding value. No guiding value exists for straw pellets. Two wood briquette samples from Spain were below the Austrian guiding value of 1.0 kg/dm^3 and one bark briquette sample was below the Austrian guiding value of 1.1 kg/dm^3 (see Tables 3 and 4).

The particle density influences the bulk density and the combustion behaviour, as dense particles show a longer burnout time.

3.5. Abrasion of pellets

The results of the abrasion tests according to ÖNORM M 7135 for all wood pellet samples, separated according to the countries of origin, as well as the results for the straw pellet samples are shown in Fig. 2.

All wood pellet samples from Italy and Spain exceeded the guiding value of 2.3 wt% (w.b.) specified in the Austrian standard. Three of the seven Austrian wood pellet samples investigated showed very high abrasion values. These samples are from small pellet producers and are mainly used for industrial purposes (e.g. internal energy production). The four Austrian wood pellet samples from large producers showed an abrasion of 1.19 + 0.25%, thus keeping the guiding value defined. According to the results, less than 1% of the total amount of pellets produced in Austria exceed the guiding value. Regarding the Swedish wood pellet samples, four samples exceeded the guiding value slightly. One of the straw pellet samples showed an abrasion value close to the guiding value and one sample had a quite high abrasion value.

Table 4 Results of the analyses of straw pellets and bark briquettes

Parameter	Unit	Straw pellets		Bark briquettes		Guiding value ^a
		Sample 1	Sample 2	Sample 1	Sample 2	
Diameter D	Mm	8.1	8.2	96	b	40-120 ^c
Length	Mm	16.6	12.3	277	2.5	$< 400^{\circ}$
Bulk density	kg/m^3	660	540			$> 1.10^{\circ}$
Particle density	kg/dm ³	1.08	0.95	1.02	1.11	<18 ^c
Water content	wt% (w.b.)	7.2	5.6	7.4	9.6	< 6.0 ^c
Ash content	wt% (d.b.)	5.97	4.82	3.31	6.39	
Abrasion	wt% (w.b.)	2.37	19.89			
GCV	MJ/kg (d.b.)	18.6	19.0	19.9	19.9	
NCV	MJ/kg (d.b.)	17.4	17.7	18.8	18.7	$> 18.0^{\circ}$
С	wt% (d.b.)	46.1	47.2	50.5	50.3	
Н	wt% (w.b.)	5.4	5.7	5.3	5.2	
Ν	wt% (d.b.)	0.55	0.53	0.26	0.43	< 0.6 ^c
S	mg/kg (d.b.)	745	788	307	499	$< 800^{\circ}$
Cl	mg/kg (d.b.)	1158	1141	68	375	$< 400^{\circ}$
Κ	mg/kg (d.b.)	8680	7200	1090	2410	
Cd	mg/kg (d.b.)	0.11	0.06	1.33	1.16	< 0.5 ^d
Pb	mg/kg (d.b.)	0.71	0.46	1.32	10.14	< 10 ^d
Zn	mg/kg (d.b.)	10.0	6.0	120.0	170.0	$< 100^{d}$
Cr	mg/kg (d.b.)	2.3	1.9	1.0	10.8	< 8 ^d
Cu	mg/kg (d.b.)	2.8	2.2	3.5	7.1	< 5 ^d

^aValid for bark briquettes.

^bSpecial form.

^cAccording to ÖNORM M 7135. ^dAccording to DIN 51731 and SN 166000 (not valid for bark, therefore only used for a comparison).



Fig. 1. Length/diameter ratio of wood pellets.



Fig. 2. Abrasion of pellets. *<2.3 wt% (w.b.) according to ÖNORM M 7135 and the standard of the Austrian Pellets Association.



Fig. 3. Correlation between abrasion and particle density of wood pellets. Coefficient of correlation $r^2 = 3.9 \times 10^{-5}$.

Abrasion is one of the most important pellet parameters. The main reason for this is the fact that a high share of fines in the storage system of the end user can cause failures in the feeding system. A low share of fines in the storage system is therefore essential for a high reliability of the equipment and for end user convenience. In addition a high share of fines also increases dust emissions during combustion. On the basis of statements of several pellet producers, the expectation has been raised that a correlation between abrasion and particle density exists, which has not been confirmed by the analysis results achieved. The scatter diagram of abrasion and particle density is shown in Fig. 3. The calculated coefficient of correlation amounts to 3.9×10^{-5} .

An explanation for this is most likely the fact that the samples have been produced on different equipment with different raw materials under different conditions. In spite of these results, it could be expected that a correlation between abrasion and particle density exists for pellets produced under similar conditions. Moreover, other parameters such as water content or the use of binding agents probably have an influence on abrasion characteristics. Further investigations would be necessary, however, in order to clarify this question.

Another expectation was that there should be a relationship between abrasion and the water content. According to [6], the water acts as a binding agent, and a correlation between the water content and abrasion has been detected. This assumption could also not be confirmed, as shown in Fig. 4. The coefficient of correlation amounts to 0.06.

The different results may be explained by the fact that all the pellets analysed in [6] were produced in the same plant, on the same pellet mill and under conditions as similar as possible. The pellets investigated within this study were obtained from different pellet producers and were produced on different pellet mills and even with different technologies for densification. Furthermore, different raw materials were used (different wood species, wood chips, sawdust, dry, wet, etc.). The results according to [6] are partly confirmed by pellet producers stating that the production of high-quality pellets is only possible in a specific range of water content (usually between 8 and 12 wt% (w.b.)). A water content above and below this range leads to pellets with a lower quality. It can therefore be concluded that the water content has an influence on the abrasion characteristics of pellets above and below the range specified, but this influence disappears if the water content is within this range.

3.6. Starch content

Starch was detected in seven pellet samples (six from Austria and one from Sweden), 16 samples were free of starch. The starch content in the pellet samples which contained starch varied between 0.16 and 1.25 wt% (w.b.) with an average value of 0.67 wt% (w.b.). Since the starch content in pellets is a good indicator of the use of biological additives, it can be concluded that these producers use such binding agents. The results achieved are also in good agreement with the indications of the pellet producers in a questionnaire survey carried out in Austria, which confirms the conclusions derived.

Furthermore, these results show that the use of biological additives (e.g. maize, rye flour) in wood pellet production is most common in Austria and



Fig. 4. Correlation between abrasion and water content of wood pellets. Coefficient of correlation $r^2 = 0.06$.

is also allowed (up to 2 wt% (w.b.)) according to the Austrian pellet standard.

The reasons for the use of additives are to reduce the operating costs of pelletisation and to achieve a higher abrasion resistance. The real effect of use of additives can only be investigated if different pellets from one specific producer are investigated.

3.7. Water content

Except for one wood briquette sample from Spain, all samples meet the guiding values specified in the Austrian standard (10 wt% (w.b.) for wood pellets and wood briquettes, 18 wt% (w.b.) for bark briquettes). The water content of wood pellets was in a range between 5.7 and 9.0 wt% (w.b.) with an average value of 7.7 wt% (w.b.). No guiding value exists for straw pellets. However, the water content of the straw pellet samples was very low (between 5.6 and 7.2 wt% (w.b.)).

The water content has an influence on the net calorific value, the combustion efficiency and the temperature of combustion.

3.8. Ash content

The high ash content of densified biomass fuels produced from bark and straw in a range between 3.3 and 6.4 wt% (d.b.) is due to the high ash content of the raw materials. Looking at the ash content of densified woody biomass fuels (shown in Fig. 5), which was measured in a range between 0.17 and 1.88 wt% (d.b.), it is noticeable that some values clearly exceed the usual ash content of wood (between 0.4 and 0.8 wt% (d.b.) for softwood and between 1.0 and 1.3 wt% (d.b.) for hardwood). These numbers also show that the use of hardwood for the production of briquettes or pellets would result in a fuel ash content exceeding the limit specified in ÖNORM M 7135 at 0.5 wt% (d.b.). Nevertheless, the very high ash contents of some samples could possibly be an indication that the producers have not exclusively used chemically untreated raw material or have problems with mineral contamination of the raw material used (e.g. by sand or earth dragged in during the storage and manipulation of sawdust) [7].

The ash contents of all samples from Italy, Spain, the Czech Republic and Norway exceed the guiding value according to ÖNORM M 7135. All Austrian samples meet this guiding value. Regarding the Swedish samples, two exceed the guiding value slightly, which could possibly result from the ash content of the wood used. Two Italian samples are known to be produced from hardwood, which is the explanation for the higher ash content in these samples.



Fig. 5. Ash content of densified biomass fuels produced from wood. *<0.5 wt% (d.b.) according to ÖNORM M 7135 and the standard of the Austrian Pellets Association; ash content determined at 550 °C according to SS 187171.

In order to maintain a high operating comfort for end users in the residential heating sector, a high ash content must be avoided. On the one hand because of the demand of emptying the ash box at periodical intervals, on the other hand because of the increasing danger of slag and deposit formation in the furnace as well as due to rising dust emissions. The ash content of pellets could be higher, however, if the pellets are used in medium and large-scale applications due to the higher robustness as well as to the more sophisticated combustion and process control technology applied for such plants.

As described in Section 2.5, two different methods have been applied to determine the ash content (loss of ignition at 550 °C according to SS 187171 and at 815 °C according to DIN 51719). The relative differences between the ash contents determined have been calculated according to Eq. (1). The results are shown in Fig. 6. The comparison of the results indicates the ash content determined at 815 °C to be lower than that determined at 550 °C in general. However, the relative differences in the ash contents determined also showed a dependence on the different biomass fuels investigated. The relative difference for wood pellets varied between 15% and 32% with a median value of 23%, for wood briquettes the median value was slightly higher (25%). The difference between the ash contents of the two bark briquettes investigated is also within the same range as that for wood pellets. Regarding straw pellets, the relative difference between the ash contents determined was only about 5%.

$$\Delta_{\rm ash} = 100 - \frac{X_{\rm ash,550}}{X_{\rm ash,815}} \times 100 \tag{1}$$

where Δ_{ash} is the relative difference between the ash content (%); $X_{ash,550}$ the ash content determined at 550 °C according to SS 187171; $X_{ash,815}$ the ash content determined at 815 °C according to DIN 51719.

The lower ash content determined at $815 \,^{\circ}$ C can be explained by the increasing decomposition of carbonates as well as by the partial vaporisation of alkali metals (especially K) and chlorides with rising temperature. A direct dependence of the difference between the ash contents on the K concentration in the biomass samples could not be determined.



Fig. 6. Relative differences between the ash contents of densified biomass fuels determined by applying different methods. Relative difference between the ash content determined at 550 °C according to SS 187171 and 815 °C according to DIN 51719; calculation of the relative difference in the ash content according to Eq. (1); numbers in brackets indicate the number of samples investigated; the boxplots for wood pellets and wood briquettes show the median value, the 25% and the 75% quantile as well as the maximum and minimum values detected.

Following the results achieved within this work and also by other studies [8], the ash content of biomass fuels should be determined at $550 \,^{\circ}$ C.

3.9. Gross calorific value, net calorific value and energy density of pellets

The gross calorific values for the different raw material species are within the typical ranges, between 19.8 and 20.7 MJ/kg (d.b.) for wood and bark and between 18.6 and 19.0 MJ/kg (d.b.) for straw.

Eq. (2) is an empirical equation from the International Energy Agency (IEA) for the calculation of the gross calorific value [9]. The parameters needed to calculate the gross calorific value according to this equation are the contents of C, H, S, N, O and ash in the fuel. All these parameters were measured, except for the content of O, which is, in good approximation, the difference between the sum of the C, H, S, N and ash concentrations (in wt% (d.b.)) and 100%.

$$GCV = 0.3491X_{C} + 1.1783X_{H} + 0.1005X_{S}$$
$$- 0.0151X_{N} - 0.1034X_{O} - 0.0211X_{ash},$$

where GCV is the gross calorific value [MJ/kg (d.b.)]; *X* the content of C, H, S, N, O and ash in wt% (d.b.); source [9].

The correlation between the gross calorific values measured and calculated is shown in Fig. 7. This scatter diagram shows a clear relationship between the measured and calculated values, which is confirmed by the coefficient of correlation of 0.73. This means that a good estimation of the gross calorific value of densified biomass fuels is possible by using Eq. (2). However, it must be considered that the calculated gross calorific value is on average about 1.8% below the measured one.

NCV = GCV
$$\left(1 - \frac{X_{W}}{100}\right) - 2.447 \times \frac{X_{W}}{100}$$

 $-\frac{X_{H}}{200} \times 18.02 \times 2.447 \left(1 - \frac{X_{H}}{100}\right)$ (3)

where NCV is the net calorific value [MJ/kg (w.b.)]; GCV the gross calorific value [MJ/kg (d.b.)]; X_W the water content [wt% (w.b.)]; X_H the H content [wt% (d.b.)]; data source [1].

The energy density of pellets was derived from the net calorific value (calculation according to Eq. (3)) and the bulk density.

The results shown in Fig. 8 revealed that the energy densities of the wood pellets investigated



(2)

Fig. 7. Correlation between the calculated and measured gross calorific value of densified biomass fuels. Calculation of the gross calorific value according to Eq. (2); coefficient of correlation $r^2 = 0.73$.



Fig. 8. Energy density of pellets.

show a high variation between 8.9 and 11.5 GJ/m³. The wood pellet samples from Italy and Spain had, on an average, a lower energy density than the samples from manufacturers in Austria and Sweden. The lower energy density of straw pellets can be explained by the lower gross calorific value of straw. However, one straw pellet sample showed a rather high energy density due to a very high bulk density of this sample.

The energy density is of importance with regard to pellet storage and transport, because the necessary storage and transport capacity decreases with an increasing energy density, and storage and transport thus become more efficient and cheaper.

3.10. Content of C, H, N and volatiles

The C, H and N contents of the different kinds of raw materials (not only wood, bark and straw but also tropical wood and eucalyptus) and also most of the N values determined comply very well with the natural ranges of such materials according to data from the literature [10–14]. However, three wood samples show an increased N content. These increased values could be explained by the use of forestry wastes including bark or the assumption that the producers have not exclusively used untreated raw materials. A high N content could be an indication for the use of prohibited substances in the pelletisation process and results in increased NO_x emissions.

C, H and O are the main components of biomass fuels. C and H become oxidised during combustion by exothermic reactions (formation of CO_2 and water) and therefore influence the gross calorific value of the fuel. The organically bound O provides a part of the O necessary for the combustion process, additional O must be supplied by air injection. The C content in woody biomass (including bark) is higher than in herbaceous biomass fuels, explaining the slightly higher gross calorific value of woody biomass.

The quantity of volatiles in biomass fuels is high and usually varies between 76 and 86 wt% (d.b.) in woody biomass [9]. This high proportion of volatiles results the major part of the biomass fuel being vaporised before homogeneous gas-phase combustion reactions take place. The remaining char undergoes heterogeneous combustion reactions. Char oxidation lasts considerably longer than the oxidation of combustible gases during the combustion process. The amount of volatile matter therefore strongly influences the thermal decomposition and combustion behaviour of solid fuels.

3.11. Content of S and Cl

The S content of the wood samples investigated has been detected to vary between 35 and 1922 mg/ kg (d.b.), that of the bark samples between 307 and 499 mg/kg (d.b.) and that of the straw samples between 745 and 788 mg/kg (d.b.). Bark and straw have a higher natural S content than wood, which is in good agreement with the results achieved during this study (except three wood samples). The average S content of wood amounts to about 70 mg/kg (d.b.), that of bark to about 350 mg/kg(d.b.) and that of straw to about 700 mg/kg (d.b.) [10]. Furthermore, the results of the S content of densified biomass fuels produced from tropical wood and eucalyptus are within the natural range of such materials compared to data from the literature [11,12].

Three wood samples exceeded the guiding value for S according to ÖNORM M 7135. However, it must be pointed out that all samples from Austria, Spain, Norway and the Czech Republic keep the guiding value, while two Italian samples and one Swedish sample exceed it. An explanation for the high concentrations of S in some samples could be the use of lignosulphonate as additive. Such materials can be used as binding agents and two samples exceeding the guiding value for S are known to have been treated with such binding agents during pellet production. For the third sample exceeding the guiding value it is also most likely that lignosulphonate has been used in the production process, but this has not been confirmed.

Typical concentrations of Cl in wood amount to about 60 mg/kg (d.b.), to about 220 mg/kg (d.b.) in bark and to about 4900 mg/kg (d.b.) in straw [10]. The Cl contents of the wood and bark samples investigated were close to these typical values. The Cl content of the straw samples was measured at about 1150 mg/kg (d.b.) and that of the densified biomass fuels produced from tropical wood and eucalyptus is within the range of the usual content of such materials [11,12]. Regarding the wood samples, only one sample exceeded the guiding value of 200 mg/kg (d.b.) according to ÖNORM M 7135. Several samples containing tropical wood or eucalyptus exceeded this guiding value. Increased concentrations of the elements Cl and S can be the result of a chemical contamination by, e.g. insecticides, adhesives, glues, lacquer, dyestuff or wood preservatives in the raw material or a result of the use of additives. The concentrations of these elements should be limited due to their negative influences on the combustion process. High amounts of S and Cl can cause problems regarding emissions (HCl and SO_x), deposit formation and corrosion. Furthermore, Cl influences the formation of dioxins and furans (PCDD/F).

According to [9], an indicator for the corrosion potential of fuels, in particular biomass fuels, is the molar ratio of $2 \times$ sulphur to available alkali and chlorine in the fuel. This ratio should exceed 10 for low corrosion potential. The reason is that increased sulphur concentrations in the flue gas reduce the formation of alkali chlorides and enhance that of alkali sulphates, which are more stable and less corrosive.

For the calculation of this ratio, the available Cl, K and Na had to be determined. Regarding Cl, the total Cl in the fuel is assumed to be available. Concerning alkali metals, the fractions of K and Na bound in the bottom ash are not available in the flue gas. According to [15], the fractions of alkali metals bound in the bottom ash amount to about 40% for K and to about 30% for Na. Following about 60% of the total K and about 70% of the total Na content in the biomass fuel can be considered as available fraction.

Taking these findings as a basis, the molar ratios of $2 \times$ sulphur to available alkali and chlorine of the densified biomass fuels investigated in the framework of this study have been calculated and are shown in Fig. 9. As it can be expected for straw pellets (if straw is used as a fuel, usually corrosion problems occur in biomass fired boilers), their ratios are very low (0.25-0.30), due to their high natural chlorine content. The two bark briquette samples investigated show ratios between 0.6 and 0.9. Three wood pellet samples show quite high ratios between 4.9 and 18.6. These samples contain quite high sulphur concentrations, most probably caused by the use of lignosulphonate as a binding agent (as described above in this section). Wood pellets show a ratio between 0.42 and 1.46, wood



Fig. 9. Calculated molar ratios of sulphur to available alkali and chlorine for different densified biomass fuels as an indicator for the corrosion potential during combustion. *Wood pellets including lignosulphonate as a binding agent; numbers in brackets indicate the number of samples investigated; calculation of molar ratio according to [9]; the boxplots for wood pellets and wood briquettes show the median value, the 25% and the 75% quantile as well as the maximum and minimum values detected.

briquettes a ratio between 0.37 and 1.45. However, the median of the ratios calculated for wood briquettes is clearly lower than the median for wood pellets, most probably due to a higher amount of bark in these samples.

As shown in Fig. 9, all woody biomass fuels investigated (except the three samples of wood pellets containing lignosulphonate as a binding agent) show quite low $2 \times$ sulphur to available alkali and chlorine molar ratios, which indicate a certain corrosion potential. The very low ratios detected for the straw pellets indicating high corrosion potential comply with experiences already achieved in straw combustion plants [16,17].

3.12. K content

The K concentrations in the samples analysed are in good agreement with the usual K concentrations of the raw material used, which are, on an average, about 500 mg/kg (d.b.) for wood, 1500 mg/kg (d.b.) for bark and 10,000 mg/kg (d.b.) for straw. However, the results determined for some wood samples clearly exceed these typical values given.

The importance of the K content is due to its influence on the ash melting behaviour and on

aerosol formation. An increased K content leads to a decreased ash melting point, which can cause slag and hard deposit formation in the furnace and boiler. Moreover, increased K concentrations rise the amount of aerosols formed during combustion and thus fouling in boilers and fine particulate emissions. Pellets made from straw, in particular, are therefore not recommended for the use in small-scale applications.

3.13. Contents of heavy metals

The content of several heavy metals (Cd, Pb, Zn, Cr, Cu, As and Hg) has been determined within the framework of this analysis programme due to their limitation according to the standard of the Austrian Pellets Association, DIN 51731 and SN 166000.

It may be stated as a general rule that the content of all heavy metals is higher in bark compared to the other biomass fuels investigated, which can be explained by the natural accumulation of heavy metals in the bark. The heavy metal content of densified biomass fuels made from straw, tropical wood and eucalyptus usually is below or in about the same range as that of densified woody biomass fuels (except for the Cu content of tropical wood in one case).

All samples investigated made from wood, straw, tropical wood and eucalyptus meet the guiding values according to the standard of the Austrian Pellets Association. DIN 51731 and SN 166000 (again except for the Cu content of tropical wood in one case). However, some wood samples indicate increased heavy metal contents (especially Cd, Pb, Zn, Cr and Cu). In this context it is noticeable that many of the increased heavy metal concentrations were determined in the same samples, which is probably an indication that partly chemically treated raw materials (e.g. recycled wood) have been used. However, this assumption is not confirmed, due to lack of information about the raw materials used from the companies supplying these samples.

Heavy metals have a strong impact on the ash quality as well as on particulate emissions. Therefore, with regard to ecological reasons, the heavy metal content in biomass fuels should be limited, especially concerning their utilisation in smallscale systems, which are usually not equipped with dust precipitation devices.

This aspect underlines the high importance of appropriate quality control procedures in order to secure clean densified biomass fuels for small-scale applications. Such a quality control and quality assurance system has recently been introduced by the Austrian Pellets Association [18]. The system is based on a codification which makes it possible to find out both the producer and the date of production of the pellets as well as the way of the pellets from the producer to the end user (complete retraceability). For this purpose, small amounts of coloured pieces of wood containing a code are added to the pellets.

4. Conclusions

The homogeneity of pellets regarding size, water content and particle density is of great relevance to achieve fully automatic operation and complete combustion in small-scale furnaces.

A surprising result that contradicted several statements from pellet producers as well as a

publication [6] was that no correlation at all could be detected between abrasion and particle density as well as between abrasion and water content. The most likely explanation for this is the fact that the pellets analysed within this analysis programme were produced in different plants, on different pellet mills, even with different densification technologies, under different conditions and from different raw materials. This shows, that abrasion of pellets depends on several parameters.

The use of biological additives is most common in Austria. Only in one sample from Sweden starch could be detected. An interesting fact in this context is that the use of binding agents does not always result in low abrasion values and therefore high-quality pellets. This is again an indication that more than one relevant influencing parameter exist.

In general, it can be stated that small amounts of biological additives (up to 2 wt% (w.b.), as is allowed according to ÖNORM M 7135) do not influence the pellet quality negatively. But the use of chemicals or waste as additives as well as mixtures of natural and chemically treated biomass as raw materials should be generally avoided if the pellets are to be used in small-scale plants. Therefore, the strict control of the quality criteria stated in the national standards for pellets is of great relevance.

All wood and straw samples investigated meet the guiding values for heavy metals according to Austrian, German and Swiss standards (except for the Cu content in one case). In order to ensure a high quality of densified biomass fuels, appropriate quality control and quality assurance systems, in particular for densified biomass fuels used in small-scale applications, are recommended. A good example for a well working quality control and quality assurance system is the system recently introduced by the Austrian Pellets Association, which is based on a codification of the pellets.

Increased ash, N, S, Cl, Cd, Pb, Zn, Cr and Cu contents were detected in some samples. The increased values of several elements were usually detected in the same samples, which could be an indication that the producers have not exclusively used chemically untreated raw materials.

Producers using lignosulphonate as additive could also be detected on the basis of high S contents and the information available from these companies. The S content is therefore a good indicator for the use of such binding agents.

The use of straw pellets in the residential heating sector is not recommended at present. State-ofthe-art small-scale pellet furnaces are not designed and suitable for this kind of fuel. Due to the high ash content of straw, the ash box of the furnace would have to be emptied more frequently and therefore the use of straw pellets would have a negative effect on user convenience. Moreover, the increased concentrations of N. Cl and K in straw could cause problems regarding emissions, deposit formation and corrosion. Furthermore, the straw pellets analysed showed high abrasion values which could cause problems with the conveying systems. Straw pellets could, however, be utilised in medium and large-scale plants due to the higher robustness and the more sophisticated combustion technology of these systems.

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