INFLUENCE OF WOOD SPECIES AND ADDITIVES ON THE COMBUSTION BEHAVIOR OF WOOD PELLETS IN STOVES AND BOILERS

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ABSTRACT: Wood pellets typically consist of sprucebut also other wood species are added during pellet production. Moreover, additives such as binders and kaolin are added in small ratios to improve fuel quality and combustion behavior. In this study eight different wood species were used for pellet production as well as three binders and one type of kaolin were added to sprucewood without bark, using two additivation levels. All wood pellet assortments were extensively analyzed regarding their physical and chemical properties and combusted in a 6 kW pellet stove and a 15 kW pellet boiler. The results confirmed a significant impact of the wood species on emissions for the pellet stove ranging between 85 (douglas fir) and 4,189 mg/Nm3 (apple wood) for CO and 23 (pine wood) and 263 mg/Nm3 (apple wood)for total particulate matter(TPM)-emission. Similar combustion behaviour was observed for the pellet boiler test but at a lower emission level. For wood species with a high molar Si/K-ratio, lower emissions but also serious slagging was observed. Binders may affect emissions when high additivation levels are used depending on the kind of binder. Kaolin reduced CO and TPMemissions as well as slag formation, when the additivation level was according to appropriate stoichiometric calculations. When kaolin is overdosed it may have negative impact on TPM emissions due to entrainment or problems caused by the higher ash content. Additionally, slagging is increased by high additivation levels of kaolin. Generally, at the current state of the art the pellet stove technology shows larger susceptibility towards variable pellet qualities compared to automatic pellet boilers. Keywords: wood pellet, fuel characterization, additives, combustion, emissions

1 INTRODUCTION

Wood pellets are widely used in pellet stoves and pellet boilers all over Europe. A high proportion of the pellets sold on the market are already labelled with ENplus A1 and should therefore be a very homogeneous fuel causing comparable and low emissions at low variability, when combusted in the same combustion unit.

A pellet screening of 42 samples from certified wood pellets from all over Europe had shown a noticeable variability in some physical (i. e. pellet length, bulk density, particle density) and chemical parameters (i. e. content of potassium, calcium and magnesium) even within the EN*plus* A1 quality class of wood pellets [1]. Eight pellet assortments from the screening were selected and combusted in a pellet stove and had caused highly variable emissions in the study [2].

Moreover, common sources of raw materials for wood pellet production like spruce and pine residues from sawmills will become less important, since these wood species are sensitive to climate changes and bark beetles. But other wood species such as douglas fir, larch, beech and oak may become more important for wood pellet production. This may cause a higher variability in chemical composition due to the broader source of raw materials which is expected to reduce the current dominance of spruce wood.

Therefore, this study aims at a systematic assessment of pellet properties and their impact on emission behaviour during combustion in pellet stoves and boilers. Consequently, 26 pellet assortments with well-defined properties were produced at TFZ and extensively tested regarding their physical and chemical properties as well as their emission and slagging behavior during combustion in one pellet stove and one pellet boiler. In particular, the focus of this paper is set on wood species, binders and additives, which represent a certain extract of a more comprehensive work done in the project.

2 MATERIAL AND METHODS

2.1 Pellet production

For the present study in total 17pellet assortments were produced at TFZ using a Amandus Kahl 33-390 flat die pellet mill and 2 assortments were provided by the project partner DEPI using an industrial ring die pellet mill. For the investigation of the influence of wood species 5 assortments of soft-wood pellets and 6 assortments of hardwood pellets were produced without binders. The softwood pellets were represented by spruce without bark (reference), spruce with a high share of bark, pine without bark, douglas fir without bark and larch without bark. Hardwood pellets were presented by 6 assortments containingoak without bark, oak with a low share of bark, beech without bark, beech with a low share of bark, mixture of tropical hardwood (sipo, meranti,...)and apple from orchard. Thus, the most important wood species for present and future pellet production in Europe are covered.

Most of the pellets from the German and European market are produced from spruce wood with a small share of other softwood and various binders. Therefore, 9 pellet assortments using the same spruce material without bark (same as for reference) were produced while adding various binders or additives. The potential influence of binding agent on emissions was investigated using wheat starch and two qualities of potato starch in a low (0.5 w-%) and a high (1.8 w-%) dosage. The potential of emission reducing additives for wood pellets was investigated using kaolin in a low (0.17 w-%) and a high (0.34 w-%) dosage. The effect ofkaolin regarding its reduction potential for particle emissions by imbedding potassium into the ash was already investigated in former projects [3][4]. The low dosage was calculated using the guidelines for advanced biofuels and boilers from the BIOFLEX! project [5] and the high dosage was the calculated maximum of kaolin by which the threshold regarding the ash content for A1 pellets can still be met when wood with low ash content is used.

2.2 Fuel analyses

All pellet assortments were analysed regarding physical and chemical parameters as listed in ISO 17225-2, but also additional fuel parameters such as particle size distribution of the raw material, pellet hardness, particle density, ash and aerosol forming elementswere assessed, following the respective ISO standardmethod (see Table I).

Furthermore, the mass fraction of starch in the additivated pellets was analyzed using a starch testing kit and the mass fraction of hardwood was determined by UNIVPM using NIR-Analysis[6][7]. The mass fraction of carbonates in the fuel ash determined at 550 °C was detected by using a LECO RC 612 multiphase carbon and hydrogen determinator. The energy density ED (equation 1) and the bulk porosity BP (equation 2) were calculated using the parameters analyzed.

Equation 1: ED, a.r. = H_u , a.r. × BD, a.r. Equation 2: BP = 1 – (BD, a.r. / PD, a.r.)

Table I: Analysis methods for fuel properties of solid biofuels (a.r. = as received, d.b. = dry basis)

Fuel quality parameter	Unit	ISO
Moisture content MC	w-%	18134-2
Ash content AC	w-%,d.b.	18122
Net calorific value H _u	MJ/kg,d.b.	18125
Bulk density BD	kg/m³,a.r.17828	
Durability DU	w-%	17831-1
Fines F	w-%	18846
Particle density PD	kg/m³	18847
Particle distribution raw mat	. w-%	17830
C-,H-,N-content	w-%,d.b.	16948
Cl-,S-content	w-%,d.b.	16994
Minor elements	mg/kg,d.b.	16967
Heavy metals	mg/kg,d.b.	16968

No standard method is available for determination of pellet hardness. Here the "Amandus Kahl GmbH Co. KG, Pressling Härteprüfer" was used, according to the manufacturer's method description.

2.3 Combustion units and methodology

All pellet assortments were combusted in an ordinary pellet stove (drop down burner) having a nominal heat power output of 6 kW. The pellet stove had an hourly cyclic cleaning of the burner pot by temporary increasing the speed of the flue gas fan for approximately 1 min to blow out loose ash and residues from the burner pot. The combustion tests were carried out by applying full load condition for 2 h, whereas 1 h was a preheating phase followed by a 1 h testing phase at steady state operation. For the pellet stove only 4 repetitions for TPM and gaseous emissions each lasting over 14 minutes were evaluated because during the fifth measurement period the cyclic cleaning of the burner pot always took place. These measurements were only executed to investigate the influence of such cyclic cleaning intervals, but the results are not considered in this investigation. All wood species and a selection of the additivated pellets were combusted in a state-of-the-art pellet boiler with a nominal heat power output of 15 kW equipped with a dumping grate and a sidewise fuel insertion. The tests with the boiler were carried out at nominal load with a 1 h preheating phase to achieve stable combustion conditions. Five TPM measurements each lasting over

30 minutes were performed with each tested fuel during steady state boiler operation.

To determine the fuel consumption during combustion, theappliance was placed on a platform scale (Mettler-Toledo GmbH, MT KD600) with a resolution of 0.005 kg.

Figure 1 shows a schematic drawing of the test stand and the arrangement of the measurement devices. The gaseous components CO, CO₂, and O₂ were determined using a single component analyzer (ABB Automation GmbH ABB AO2020), NO_X by a chemiluminescence detector (Eco Physics GmbH CLD 822 MhrAnalysator) and for water vapour contentan FTIR-analyser (Ansyco GmbH FTIR DX4000N) was used. The recording interval for the continuous measurement was set to 10 seconds. The total particulate matter (TPM) was isokinetically sampled following VDI 2066-1 applying a filtration temperature during sampling of 160 °C and the filter pre- and post-treatment temperature of 180 °C and 160 °C, respectively. Only quartz plane filters were used and the particle deposition in the sampling line was also considered by rinsing the probe and determining the solid residue after evaporation of the liquid. The pellet stove was operated at a constant flue gas draught of -12±2 Pa and the pellet boiler was operated at -15±2 Pa as it is suggested by the manufacturers. The diameter of the flue gas duct and the connection pipe was 100 mm (stove)and130 mm (boiler). The flue gas velocity was continuously measured using a vane anemometer (Höntzsch GmbH, ZS25/25-ZG4) positioned in a narrowed stretch of the measurement section with a diameter of 64 mm (stove) resp. 100 mm (boiler) (see Figure 1). The heat consumption during the boiler tests was permanently regulated to a nominal load of 15 kW (± 8 %) following DIN EN 303-5.



Figure 1: Schematic drawing of the test stand used for stove and boiler tests (position of measurement points: T = temperature, p = pressure, v = velocity).

In addition, a carbon content determination of the collected flue gas particles was performed and the share of elemental and organic carbon was analyzed using a LECO RC 612 multiphase carbon and hydrogen determinator. The residual mass (TPM minus inorganic carbon(as CO_2) minusorganic carbon minus elemental

carbon) was summarized and supposed to represent the fraction or salts and oxides of particulate matter emissions. Furthermore, slag formation was also assessed by sieving all combustion residues using a 2 mm and a 1 mm mesh with a Retsch AS 200 control sieving machine performing a 3D sieving over 5 minutes applying an amplitude of 0.5.

2.4 Statistical analyses

If differences between mean values are declared as being "significant" the data was always previously tested to normality (Shapiro-Wilk) and significance was tested using a one-way ANOVA combined with the comparison of mean values via Tukey-test. The level of significance was always set to p<0,05.

3 RESULTS AND DISCUSSION

3.1 Fuel analyses

Table II to Table V summarize the results from fuel analyses.

3.1.1 Wood species (Table II and Table IV)

The ash content of the wood species used varies between 0.27w-% (oak 1) and 2.39 w-% (apple wood). Spruce with a high share of bark (0.83 w-%) and apple exceeded the threshold for A1 class pellets, while tropical hardwood mixture is straight at the limit for the ash content of 0.7 m-%. The fraction of carbonates which was analyzed from the 550 °C ashes rangesbetween 0.03 w-% for douglas fir and 0.69 w-% for apple wood. Spruce without bark, pine, douglasfir and oak are showing a rather low mass fraction of carbonates in the fuel ashes whereas spruce with a high share of bark, larch, beech, tropical hardwood mixture and especially apple wood showing higher mass fraction of carbonates in the fuel ash. The energy density varies between 11.1 GJ/m³ (oak 1) and 12.4 GJ/m³ (spruce 2), whereas softwood showed slightly higher energy densities than hardwood excepting beech 1. The fines from the pellets produced were rather low between 0 w-% (spruce 2, larch, beech 2 and apple) and 0.3 w-% (pine). The mean pellet length varies between 11.2 mm (tropical hardwood mixture) and 21.8 (beech 2) and is obviously also depending on the wood species.

The mass fraction of nitrogen in the fuel is an indicator for NO_x formation since it mainly results from the fuel nitrogen (N) [8]. According to the fuel N the lowest NO_Xemissions can be expected for larch wood pellets were N is below the here given detection limit of 0.05 w-% and the highest for apple wood having a nitrogen content of 0.19 w-%. Generally, softwood species with low share of bark showed slightly lower mass fraction of N than hardwood species except beech 1 where the fuel N is at 0.10 w.-%. The sum of aerosol forming elements (K+Na+Zn+Pb) ranges between 451 mg/kg, d (larch) and 4,323 mg/kg (apple wood), mainly dominated by potassium (K). Pine, douglas fir, beech 2, and especially apple and tropical hardwood mixturehad a considerablecontent of sodium (Na), too. According to the sum of aerosol forming parameters, higher TPM emissions could be assumed for hardwood pellets, especially beech, apple and also for spruce 2, due to it's the higher share of bark [8]. The presence of higher mass fractions of silicon(Si) in the fuel can reduce the release of potassium to the gas phasesince K is embedded in the ash during combustion [9]. This effect can be

estimated using the molar Si/Kratio [8] which indicates embedding of potassium when the value is above 2.5. For the tested wood species, the Si/Kratio lies between 0.19 (beech 1) and 2.61 (pine). According to this ratio low potassium release could be assumed for pine and douglas fir. The slagging behavior can be estimated by using the molar (Si+P+K)/(Ca+Mg+Al)-ratio [9] if the fuel is additivated with aluminumbased additives or if the biomass contains significant aluminum concentrations such as pine, beech 2 or apple wood. According to this index which should be below 2.5, lower ash melting temperatures can be estimated for pine (1.33), douglas fir (1.71), oak 1 (1.22), oak 2 (1.64) and beech 2 (1.48) compared to the other wood species were the index is below 1 and no slag formation is expected.

3.1.2 Additives (Table III and Table V)

The ash content of the reference material without binder was at 0.41 w-% whereas the additivated pellets using the same raw material had an ash content between 0.33 m-% (spruce 0.5 w-% potato starch (mod.)) and 0.63 m-% (spruce 0.34 w-% kaolin). The results regarding the binders reveal, that the raw material was not very homogeneous for all pellet batches. The differences in the ash content among the assortments with different binders could therefore not be onlycaused by the binder. The additivation with kaolin results in a significant increase of the ash content. The mass fraction of carbonates in the ash residue after ash determination is between 0.08 and 0.09 w-% and thus not affected by the binding agents, but it was reduced to 0.05 w-% for both kaolin additivation levels. The energy density was between 11.0 GJ/m³ (spruce with 1.8 w-% potato starch (nat.+mod.) and 12.3 GJ/m³ (spruce with 1.8 w-% wheat starch). Energy density is mainly influenced by the mean pellet length and particle density. The fines are for all assortments as low as 0.1 w-%. The mean pellet length differs between 17 mm (spruce without bark, reference) and 23 mm (spruce with 1.8 w-% potato starch (nat.)) and it seems that the dosage of the binding agent can affect the pellet length (wheat starch and potato starch (nat.)).

The addition of binders and additives caused an increase in the mass fraction of N in the fuel.Since kaolin typically contains no significant amounts of nitrogen the increase of N in the kaolin additivated fuels should be due to analyses error. Therefore, slightly higher NO_X emissions can be expected when binders are added. The sum of aerosol forming elements varies between 491 mg/kg (spruce with 0.17 w-% kaolin) and 610 mg/kg (spruce with 1.8 w-% wheat starch) and may be caused by natural variations of the raw material and cannot clearly be attributed to a binder or to kaolin. Nevertheless, it seems that high shares of binders or kaolin might slightly increase the content of aerosol forming elements. This is especially obvious for spruce with 1.8 w-% potato starch (mod.) were sodium is raised from detection limit of 10 to 22 mg/kg by adding the binder. With a high uncertainty slightly higher TPM emissions can be assumed when high shares of binders are used. The Si/K-ratio is between 0.25 and 0.29 for spruce with and without binders. When kaolin is added the Si/K-ratio rises to 0.44 for the low and 1.15 for the high additivation level. Regarding the slagging index, all assortments are on a very low level having an index of below 0.73, thereforeno slagging problems are expected. For the additivation with kaolin the slagging index (molar(Si+P+K)/(Ca+Mg+Al)-ratio) is reduced.

Parameter	Ash content	Fraction of carbonates in the fuel ash*	Bulk density	Particle density	Energy densitiy	Fines	Mean pellet length
Unit	w-%, d	w-%, d	kg/m ³ , a.r	kg/m ³ , a.r	GJ/m ³ , a.r.	w-%, a.r.	mm, a.r.
Spruce 1 without bark (reference)	0.41	0.09	692	1312	12.1	0.1	17.0
Spruce 2 with a high share of bark	0.83	0.24	720	1312	12.4	0.0	15.0
Pine without bark	0.61	0.05	671	1287	11.8	0.3	11.3
Larch without bark	0.39	0.10	701	1319	11.8	0.0	18.8
Douglas fir without bark	0.38	0.03	690	1341	11.7	0.1	18.7
Oak 1 without bark	0.27	0.07	666	1275	11.1	0.2	16.5
Oak 2 with a low share of bark	0.36	0.07	675	1268	11.4	0.1	16.9
Beech 1 without bark	0.63	0.15	691	1310	11.8	0.1	21.0
Beech 2 with a low share of bark	0.56	0.15	680	1301	11.5	0.0	21.8
Tropical hardwood mixture	0.70	0.18	650	1247	11.2	0.2	11.2
Apple wood from orchard	2.39	0.69	696	1307	11.6	0.0	20.7

Table II: Fuel properties of the produced test pellets from different wood species (a.r. = as received, d = dry basis)

* in residue after ash determination at 550 °C

Table III: Fuel properties of the produced test pellets from spruce using various additives (a.r. = as received, d = dry basis)

Parameter	Ash content	Fraction of carbonates in the fuel ash*	Bulk density	Particle density	Energy density	Fines	Mean pellet length
Unit	w-%, d	w-%, d	kg/m3, a.r	kg/m3, a.r	GJ/m ³ , a.r.	w-%, a.r.	mm, a.r.
Spruce 1 without bark (reference)	0.41	0.09	692	1312	12.1	0.1	17.0
Spruce with 0.5 w-% wheat starch	0.44	0.09	680	1310	11.8	0.1	17.3
Spruce with 1.8 w-% wheat starch	0.44	0.08	709	1319	12.3	0.1	18.3
Spruce with 0.5 w-% potato starch (nat.)	0.39	0.09	670	1292	11.5	0.1	21.9
Spruce with 1.8 w-% potato starch (nat.)	0.39	0.09	646	1283	11.0	0.1	23.0
Spruce with 0.5 w-% potato starch (mod.)	0.33	0.09	651	1291	11.1	0.1	21.7
Spruce with 1.8 w-% potato starch (mod.)	0.39	0.09	643	1268	11.0	0.1	21.4
Spruce with 0.17 w-% kaolin	0.48	0.05	678	1296	11.6	0.1	21.1
Spruce with 0.34 w-% kaolin	0.63	0.05	671	1294	11.5	0.1	21.8

nat. = native; mod. = lower quality starch with a high share of modified starch; * in residue after ash determination at 550 °C

Table IV: Relevant element concentrations and fuel indices of the produced test pellets from different wood species (d = dry basis)

Parameter	Ν	K	Na	Si	Al	Ca	Mg	K+Na+Pb+Zn	Molar Si/K	Molar (Si+P+K)/ (Ca+Mg+Al))
Unit	w-%, d	w-%, d	w-%, d	mg/kg, d	mg/kg, d	mg/kg, d	mg/kg, d	w-%, d	-mol/mol	mol/mol
Spruce 1 without bark (reference)	0.09	562	< 10	117	14	859	163	578	0.29	0.73
Spruce 2 with a high share of bark	0.11	698	13	529	99	2,429	221	743	1.05	0.54
Pine without bark	0.08	443	39	832	182	732	164	492	2.61	1.33
Larch without bark	< 0.05	447	< 10	165	14	840	303	451	0.51	0.60
Douglas fir without bark	0.09	411	22	604	87	536	84	438	2.05	1.71
Oak 1 without bark	0.14	612	16	< 100	41	513	46	630	0.23	1.22
Oak 2 wih a low share of bark	0.15	833	12	279	23	672	78	847	0.47	1.64
Beech 1 without bark	0.10	1,130	< 10	155	18	1,260	358	1,137	0.19	0.78
Beech 2 with a low share of bark	0.14	1,449	33	519	163	909	269	1,493	0.50	1.48
Tropical hardwood mixture	0.17	607	93	402	79	1,710	223	711	0.92	0.57
Apple wood from orchard	0.19	4250	56	1,570	163	7,850	878	4,323	0.51	0.75

Table V:Relevant element concentrations and fuel indices of the produced test pellets from spruce using various additives (d = dry basis)

Parameter	N	К	Na	Si	Al	Ca	Mg	K+Na+Pb+Zn	Molar Si/K	Molar (Si+P+K)/ (Ca+Mg+Al))
Unit	w-%, d	w-%, d	w-%, d	mg/kg, d	mg/kg, d	mg/kg, d	mg/kg, d	w-%, d	mol/mol-	mol/mol
Spruce 1 without bark (reference)	0.09	562	< 10	117	14	859	163	578	0.29	0.73
Spruce with 0.5 w-% wheat starch	0.10	549	< 10	< 100	18	818	159	568	0.25	0.73
Spruce with 1.8 w-% wheat starch	0.12	593	< 10	111	16	929	172	610	0.26	0.73
Spruce with 0.5 w-% potato starch (nat.)	0.16	489	< 10	< 100	35	865	160	504	0.28	0.63
Spruce with 1.8 w-% potato starch (nat.)	0.15	526	< 10	< 100	23	854	162	540	0.26	0.69
Spruce with 0.5 w-% potato starch	0.14	506	< 10	< 100	36	894	165	522	0.28	0.62
Spruce with 1.8 w-% potato starch	0.15	542	22	< 100	25	918	171	578	0.26	0.67
Spruce with 0.17 w-% kaolin	0.14	477	< 10	150	318	855	156	491	0.44	0.49
Spruce with 0.34 w-% kaolin	0.14	487	< 10	404	632	845	156	503	1.15	0.56

nat. = native; mod. = lower quality starch with a high share of modified starch

3.2 Results from combustion tests

3.2.1 Wood species

The COemissions (Figure 2) for the tested wood species applied in the pellet stove are between 78 mg/Nm³ for pine and 4,189 mg/Nm³ for apple wood due to the very high ash content. If apple is excluded the highest COemissions from the pellet stove were measured for beech 1 at a level of 461 mg/Nm3. Typically, softwood pellets with low share of bark caused lower CO emissions than hardwood pellets.The combustion of spruce with a high share of bark shows an increase of COemissions compared to spruce without bark but the difference is not significant. The German CO threshold value from the 1. BImSchV for such pellet stoves without water jacket is at 250 kg/Nm³; it is exceeded by beech 1 and beech 2, tropical hardwood mixtureand apple wood. But when looking to the minimum and maximum values, even oak 2 and spruce 2 could be problematic. For most wood species, the pellet boiler shows very low COemissions between 1 (pine) and 13 mg/Nm³ (beech 1) without a significant difference. In contrast to that the CO emissions for apple wood combusted in the boiler were as high as 247 mg/Nm³.



Figure 2:Influence of wood species on COemissions from a pellet stove and a pellet boiler

If apple wood is excluded the COemissions from the pellet stove show a significant correlation (R^2 =0.80) with the aerosol forming elements and a slight but not significant correlation (R^2 =0.39) with the carbonates in the fuel ashafter ash determination at 550 °C (see Figure 3). No similar correlation was found for the pellet boiler.



Figure 3: Linear regression of CO with carbonates in ash after determination at 550 °C, resp. with aerosol forming elements for the pellet stove excluding apple wood pellets.

The OGCemissions (Figure 4) for both, stove (93 mg/Nm³) and boiler (3 mg/Nm³) are the highest for apple wood which is obviously due to the high ash content which eventually inhibits a uniform access of combustion air to the ember. Regarding all other fuels used in the pellet stove, OGCemissions are generally slightly higher (not significant) for hardwood compared softwood pellets. For the pellet stove the to OGCemissions are showing a strongly significant correlation with $CO(R^2=0.99, not)$ illustrated). OGCemissions from the pellet boiler (excluding apple wood) are very low and close to the detection limit of the FID.



Figure 4:Influence of wood species on OGCemissions from a pellet stove and the pellet boiler

The NO_xemissions as illustrated in Figure 5 are ranging between 90 mg/Nm³ (larch) and 212 mg/Nm³ (tropical hardwood mixture) for the stove and between 93 mg/Nm³ (larch) and 364 mg/Nm³ (apple) for the boiler.NO_x emissions are higher for the pellet boiler due to the higher combustion temperature compared to the stove. Apparently,hardwood pellets (except oak 1) cause higher NO_xemissions than most softwood pellets, when comparing pellets with low share of bark. Also, the bark content seems to have an impact on NO_xemissions when comparing spruce 1 and spruce 2. This effect seems to be more pronounced for the boiler.



Figure 5:Influence of wood species on NO_xemissions from a pellet stove and a pellet boiler

Both appliances show a significant correlation between the NO_xemissions and the N-content in the fuel, both at a similar correlation coefficient of R^2 = 0.79 (see Figure 6). It seems that the boiler leads to a higherN to NO conversion ratioat higher nitrogen levels than the stove.



Figure 6: Linear regression of NO_xemission and Ncontent in the fuel, for pellet stove and pellet boiler

TPM emissions varied between 23 and 24 mg/Nm³ (pine and douglas fir) and between 63 and 65 mg/Nm³ (beech 1 and beech 2) for the pellet stove. For the pellet boiler they were between 5 and 6 mg/Nm³ (douglas fir and pine) and between 50 and 55 mg/Nm³ (beech 2 and beech 1). The highest TPM emissions were observed for apple wood with 109 mg/Nm3 for the boiler and 263 mg/Nm³ for the stove which are caused by the high mass concentration of aerosol forming elements on the one hand and by incomplete combustion due to the high ash content causing a quick buildup of the fuel bed in the pellet stove which lead to a blockage of the secondary air nozzles during test runs. The biggest differences between boiler and stove can be seen for pine, douglas fir and tropical hardwood mixture where the boiler emits less than half of the TPM emissions of the stove. For both appliances probably the bark content has a significant influence on TPMemissions when comparing e.g. spruce 1 and spruce 2 which should be caused by a higher mass fraction of aerosol forming elements in bark compared to stemwood[10].

Comparing pellets without or with a low bark content, for both appliances higher TPMemissions are found for hardwood compared to softwood. For pine and douglas fir the high Si/Kratio of >2 has obviously a significant positive impact on the formation of TPMemissions of the boiler, but not for the stove, whereobviously other PM formation mechanisms have been more effective.



Figure 7:Influence of wood species on TPMemissions from a pellet stove and a pellet boiler

The composition of the particles from TPMsampling for the pellet stove and boiler is given in Table VI. For spruce 2 and beech 2 combusted in the pellet boiler no analyses of the TPMfilters were performed. The first value in the table is always representing the stove and the second is for the boiler. The mass fraction of inorganic carbon (IC) can range between 0 and 13 m-% for both appliances and is the same for tropical hardwood mixture, spruce 1, and very similar for pine, douglas fir, oak 2 and beech 1. The mass fraction of IC should be mainly caused by entrainment of carbonate rich coarse particles (ash).

The main differences between the TPMemissions from stove and boiler are due to the formation of elemental carbon (EC). While for the pellet stove the mass fraction of EC on TPMemissions is between 0 m-% (oak 2, beech 1 and beech 2) and 54 m-% (pine) for the boiler it is only between 0 m-% (spruce, larch, oak 1, oak 2 and beech 1) and 2 m-% (pine). EC indicates soot formation from incomplete combustion and possibly also a slight entrainment of unburned charcoal particles. The mass fraction of organic carbon (OC)is substantially lower for the boiler (1 - 6 m-%) compared to the stove (6-14m-%). TPM emissions from the boiler mainly consist of salts and oxides for all wood species (81-99 m-%) whereas TPM emissions from the pellet stoveconsist of only 35 to 85 w-% salts and oxides. This lower share of salts and oxides explains the higher differences of the TPMemissions between boiler and stove when burning pine, douglas fir or tropical hardwood mixture.

While the Si/K-ratio effects the embedding of potassium into the fuel ash [8] [9] and therefore the salt and oxide fraction of TPM it does not affect the EC and OC fractions which are a main part of the TPMemissions from the pellet stove for these wood species.

Table VI:Composition of TPMemissions from the investigated wood species in the pellet stove / boiler, divided in IC (calculated as CO_2), EC, OC (adjusted by factor 1.4),salts and oxides (d = dry basis, n. d. = not determined)

Wood species	IC (CO ₂) w-%, d	EC w-%, d	OC w-%, d	Salts and oxides w-%, d
Spruce 1	2 / 2	6 / 0	12/2	80 / 96
Spruce 2	13 / n. d.	14 / n. d.	11 / n. d.	63 / n. d.
Pine	0 / 1	54 / 2	11/3	35 / 94
Larch	4 / 10	5 / 0	10 / 5	80 / 84
Douglas fir	2 / 1	40 / 1	10 / 4	49 / 95
Oak 1	7 / 4	12 / 0	14/3	67 / 94
Oak 2	1 / 0	0 / 0	14 / 1	85 / 99
Beech 1	11 / 13	0 / 0	12 / 6	77 / 81
Beech 2	4 / n. d.	0 / n. d.	6 / n. d.	90 / n. d.
Tropical hardwood mixture	1 / 1	16 / 0	14 / 1	69 / 99
Apple	2/11	40 / 0	12/3	46 / 86

The salt and oxide fraction of TPMemissions correlates well with the sum of aerosol forming elements (see Figure 8) for both the boiler (R^2 =0.87) and the stove (R^2 =0.89). Pine and douglas fir do not fit into this correlation which could be explained by the higher Si/Kratio of above 2 [8]. ECemissions from the pellet stove showed a significant correlation (R^2 =0.52, not illustrated) with the Si/Kratio which could probably be due to a negative impact of the higher Si content to the ash melting behavior, causing higher soot emissions by local lack of oxygen or channeling effects in the fuel bed for instance.



Figure 8: Linear regression of salt and oxide particles with the mass fraction of aerosol forming elements (K, Na, Pb, Zn) in the fuel for the investigated wood species (apple wood was here excluded).

3.2.2Additives

All additivated spruce assortments were tested regarding their emission behavior in the pellet stove but only the reference fuel (spruce 1), spruce with 1.8w-% modified potato starch and the two kaolin levels were combusted in the pellet boiler. In the following illustrations reference means always spruce 1 (without bark), for the other assortments the raw material was always the same as for spruce 1 and only the used additive and additivation level is given in the axis labeling.

The COemissions (Figure 9) for the tested are between 73 mg/Nm³ assortments (0.34 m-% kaolin)and 178 mg/Nm³ (1.8 m-% wheat starch) when combusted in the pellet stove. For the boiler the COemissions rangemuch lower, between 2 (0.34 m-% kaolin) and 5 mg/Nm3 (1.8 m-% potato starch mod.). For the pellet stove only 1.8 w-% wheat starch and 0.34 w-% kaolin showed significant differences. In trend (not significant) high additivation levels of binders seem to have a negative impact on COemissions while the high level of kaolin reduces the COemissions at the pellet stove. These effects (not significant) can also be seen for the pellet boiler but only on a very low level compared to the pellet stove.



Figure 9:Influence of additives on COemissions from a pellet stove and a pellet boiler

The OGCemissions (Figure 10) range between 2.4 mg/Nm³ (0.5 w-% wheat starch and 0,34 w-% kaolin) and 5.8 mg/Nm³ (0.17 w-% kaolin) for the pellet stove and between 0.5 and 0.6 mg/Nm³ for the boiler. While the boiler shows no significant impact of the additives on OGCemissions, the stove shows a significant increase of OGCemissions when a binder (except 0.5 w-% wheat starch) or the low kaolin level is added, compared to the reference.



Figure 10:Influence of additives on OGCemissions from a pellet stove and a pellet boiler

Regarding NO_xemissions (Figure 11) the stove shows a significant increase for fuel with wheat starch and native potato starch additive, compared to the reference. This is particularly true for the high additivation level while modified potato starch reduces NO_Xemissions especially for 1.8 w-%, this is also true for kaolin. This effect contrasts with the higher mass fraction of N for the additivated pellets which would rather indicate a NO_X increase. There is a weak correlation between NO_X and pellet length, indicating a depressing effect of the pellet length on NO_xemissions for the pellet stove, but this correlation is not statistically significant. The boiler shows an exactly opposite behavior with significantly increasing NO_xemissions for 1.8 w-% modified potato starch compared to the reference but slightly lower NO_X for kaolin additivation, as it was the case for the stove.



Figure 11:Influence of additives onNO_xemissions from a pellet stove and a pellet boiler

The TPMemissions for the additivated pellets in the pellet stove and boiler are illustrated in Figure 12. For the pellet stove, TPMemissions are only increased significantly when the fuel with 1.8 w-% modified potato starch was used, but in trend (not significant) all binders which are added in the high additivation level of 1.8 w.-% led to an increase of TPMemissions. Also, the variation regarding the single measurements was thus increased by trend. For the boiler, by trend (not significant) the same effect could be observed for modified potato starch. For kaolin, both appliances showed a reducing effect on TPMemissions for the low additivation level of 0.17 w-%, but this effect was only significant for the boiler (compared to the reference fuel). For the high additivation level the stove shows a significant reduction of TPMemissions compared to the reference fuel while for the boiler the TPMemissions are at thelevel of the reference fuel.



Figure 12:Influence of additives on TPMemissions from a pellet stove and a pellet boiler

The composition of the TPMemissions using different additives in the pellet stove and boiler is given in Table VII.The main difference between pellet stove and boiler can be found again in the elevated fraction on EC and OC on the collected particles for pellet stoves. The main component of the TPM emissions released from boilers are salts and oxides ranging between 93 and 98 w-% whereas the fraction of salts and oxides released from pellet stoves varies at a lower level, between 50 and 85 w-%. Consequently, the fraction of OC and EC is higher from pellet stove combustion. According to the composition of TPM for the stove, the increase of TPM by high additivation levels with wheat starch and modified potato starch is mainly due to elevated soot formation as indicated bythe EC content.

The same behavior was observed for kaolin addition for the stove. The mass fraction of EC rises with the amount of kaolin added whilethe salts and oxides are reduced by embedding of potassium into the ash bed. For the boiler it seems that the optimum regarding embedding of potassium is already reached at the low kaolin level while and at the highlevel kaolin can rather be entrained with the flue gas resulting in higher shares of salts and oxides in TPM emission. Differences between stove and boiler regarding the effect of the high and low kaolin level can probably be explained by different combustion temperatures and flow rates in the fuel bed which may have an effect on the potassium release [10][9] and also the entrainment of kaolin.

Table VII:Composition of TPMemissions from the investigated additivated pellets in the pellet stove / boiler, divided in IC (calculated as CO_2), EC, OC (adjusted by factor 1.4), salts and oxides (d = dry basis)

Spruce additivated with	IC (CO ₂)	EC	OC	Salts and oxides
-	w-%, d	w-%, c	l w-%, d	w-%, d
Reference	2/2	6 / 0	12 / 2	80 / 96
0.5 wheat	2 / -	2 / -	11 / -	85 / -
1.8 wheat	3 / -	35 / -	12 / -	50 / -
0.5 potato nat.	1 / -	13 / -	11 / -	74 / -
1.8 potato nat.	4 / -	6 / -	11 / -	79 / -
0.5 potato mod.	3 / -	9 / -	11 / -	78 / -
1.8 potato mod.	0 / 1	36 / 0	8 / 2	55 / 98
0.17 kaolin	2 / 1	20 / 3	18 / 4	60 / 93
0.34 kaolin	0 / 2	45 / 0	13 / 2	42 / 96

3.3 Results on slagging behavior

3.3.1 Wood species

The results of the granulometric investigation of the ashes from the combustion tests are shown as mass fraction of particles > 2 mm and > 1 mm after combustion test in Table VIII. Both appliances showed high fractions of particles > 2 and > 1 mm for pine, douglas fir, oak 2 and apple. The stove shows also higher values for beech 1 and beech 2 whereas the pellet boiler shows relatively low values for beech 1 and high values for particles > 1 mm for larch and oak 1. The share of particles > 2 mm on total ashes from combustion tests shows a significant correlation (see Figure 13) with the molar (Si+P+K)/(Ca+Mg+Al)-ratio for the stove $(R^2=0.88)$ and the boiler $(R^2=0.64)$. The results also confirm that the fuels with a high Si/Kratio like pine anddouglas fir have a higher risk of slag formation.

In contrast to the results regarding emission behavior it could not be confirmed that hardwood would also be disadvantageous in terms of slagging; this rather depends on mass fraction of Si, K, Ca, Mg and Al which depends on the wood species (e. g. K) but also on the growing region of the tree (e. g. Si and Al). Furthermore, the pellet stove and pellet boiler react differently on some wood species like beech 1, oak 1 and larch, this isprobably due to different temperature levels during combustion.

Table VIII:Share of particles > 2 mm and > 1 mm on the residues (ashes) of combustion tests for different wood species in pellet stove and boiler (a.r. = as received)

Wood species	> 2 mm stove	> 1 mm stove	> 2 mm boiler	> 1 mm boiler
_	w-%, a.r.	w-%, a.r.	w-%, a.r.	w-%, a.r.
Spruce 1	16	28	12	22
Spruce 2	3	5	2	4
Pine	32	47	33	49
Larch	5	9	9	30
Douglas fir	57	65	57	66
Oak 1	15	27	29	39
Oak 2	40	48	43	51
Beech 1	43	51	14	25
Beech 2 Tropical hardwood	35	43	25	43
mixture	3	13	2	8
Apple	18	49	33	52



Figure 13: Linear regression between share of particles > 2 mm on total ashes from combustion tests for the investigated wood species in the pellet stove and boiler. 3.3.2Additives

In Table IX the results of the granulometric investigation of the ashes from the combustion tests are shown as mass fraction of particles > 2 mm and > 1 mm in the combustion residues.For the pellet stove, no negative effect of the binders on slagging could be observed within the precision of the measurement, the same is true for the boiler trials. For the low kaolin level particles > 2 and > 1 mm are clearly reduced, for the boiler only the fraction > 2 mm is reduced. For both appliances especially the high level for kaolin has a negative effect on slag formation as determined in this granulometric assessment.

Table IX: Share of ash particles > 2 mm and > 1 mm on the ashes from combustion tests regarding additives in pellet stove and boiler (a.r. = as received, n.c. = not combusted in boiler)

Spruce additivated with	> 2 mm stove	> 1 mm stove	> 2 mm boiler	> 1 mm boiler
	w-%, a.r.	w-%, a.r.	w-%, a.r.	w-%, a.r.
Reference	14	22	12	22
0.5 wheat	12	20	n.c.	n. c.
1.8 wheat	18	25	n.c.	n. c.
0.5 potato nat.	12	22	n.c.	n. c.
1.8 potato nat.	7	15	n.c.	n. c.
0.5 potato mod.	9	17	n.c.	n. c.
1.8 potato mod.	14	22	17	28
0.17 kaolin	0	3	9	20
0.34 kaolin	21	32	33	48

4 CONCLUSIONS

The use of various wood species for pellet production may have a significant impact (double to quadruple) on the emissions during the combustion, mainly in pellet stoves but also in boilers. Generally, it was shown that the combustion of hardwood caused significantly higher gaseous and particulate emissions, especially when the ash content is above 0.5 w-% (e.g. beech and tropical hardwood mixture). This is mainly due to thehigher amount of aerosol forming elements (K, Na, Pb, Zn) in hardwood. Additionally, a slight impact of the carbonate content in the ash (i.e. residue at 550 °C) was found which was also typically higher in hardwoods. Therefore, it seems helpful to limit the ash content for A1 pellets to ≤ 0.5 w-%, particularly for use in pellet stoves. This will have the effect, that alsomixtures of softwood and high shares of hardwood and high share of bark would become unsuitable. The German emission threshold (1. BImSchV) for particle emissions was reached for all softwood pellets when the bark content was low, for a higher bark content and for all hardwood pellets the threshold was exceeded in the pellet stove but also in the boiler. For the stove even the COthreshold could be a challenge for most pellets from hardwood and for softwood with a high share of bark.

Wood species which are high in Si suchas pine or douglas fir (depending on growing region) showedvery low particulate and gaseous emissions due to a higher Si/Kratio, but over longer combustion phases they can also cause serious slag formation, which is usually not assessed during a regular type test of a stove or boiler.For realistic type testing results it could thus be helpful to demand a limit of the Si content in the fuel in order to keep the Si/K ratio on a more typical level and thus to prevent any unrealistically reduced PM emissions which do not reflect real life operations.

Binders may have a slight impact on emissions when the additivation level is high (e. g. 1.8 w-%),anddepending on the kind of binder used. This could be addressed in future by lowering the maximum allowed mass fraction of binders from today 2 w-% [12] to only 1 w-%, for example.

For the pellet stove and pellet boiler particulate matter and COemissions were reduced by fuel additivation with kaolin. But the appliances showed a different behavior on TPMemissions when the calculated additivationwas raised to double of the stoichiometrically calculated level. While the stove showed further reductions, the TPMemissions for the boiler increased.

From the analysis of the composition of TPMemissions it can be concluded that an overdosage of kaolin should be avoided for both furnace types. The boiler reacts with flue gas entrainment of kaolin. For the stove, however, potassium embedding is further improved by kaolin and thus the TPMemissions are reduced, but particle formation from incomplete combustion may be increased.

For both appliances the slagging behavior is improved at low fuel additivationlevel, butslag formation increases at the higheradditivation level. Therefore, when kaolin is used the dosing of the kaolin should be done strictly according to a stochiometric calculation which includes only a small safety margin (e.g.by following guidelines [3])to avoid entrainment of kaolin or slag formation.

Generally, the emissions from the pellet stove were clearly higher than those from the pellet boiler; this indicates that there is still considerablepotential for improvements of pellet stove technology. These improvements could be achieved by a consequent regulation of the fuel feeding rate via temperature measurement or lambda control which is still missing for most pellet stoves on the market.Furthermore, a more uniform fuel feeding may lead to lower emissions since the fuel bed is not disturbed so often. Additionally, it was observed during the test runs that a cyclic cleaning of the burner pot, simply by increasing the speed of the flue gas fan to blow out residues from the burner pot, will increase TPM emissions significantly and will also force sintering of the ashes below the char bed. Test runs without cyclic cleaning showed that the time frame between cleaning sequences could be extended by several hours before the fuel bed will eventually rise to a problematic level.

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7 LOGO SPACE



