The Increase of Silver Grass Ash Melting Temperature Using Additives

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Received: 11.12.2014 Accepted: 11.01.2015

Abstract- Some types of biomass have a high content of alkali oxides and salts and, thanks to it they feature a low ash melting temperature which may result in various combustion problems. Slag and sintered particles that are formed prevent the fuel from being supplied, restrict the access of combustion air, heat transfer in the heat exchanger and can cause corrosions. One possibility of increasing the ash melting temperature is to add additives. Some additives can change chemical composition of fuel ash, thus changing also the ash melting temperature. The paper deals with the adding of 2 % additives – kaolin, talc, lime, limestone, dolomite, bentonite to silver grass. The tested additives increased the ash content but they also changed its chemical composition, which resulted in the increase of the ash melting temperature. Best positive results were achieved with kaolin and lime which increased the ash melting temperature by approx. 300 °C.

Keywords- Biomass, Silver grass, Additives, Ash melting temperature, Chemical composition of ash, Ash content

1. Introduction

Solid fuels, including biomass, consist of combustible matter, ash and water. Combustible is that part of fuel which, when oxidized, releases heat, i.e. chemically bound energy in fuel. One part of combustible consists of carbon (C), hydrogen (H) or sulphur (S). These are so called active substances of combustibles which during oxidation give rise to heat [1]. Another part of the combustible consists of so called passive substances which do not supply any heat but which are bound to organic mass. Passive substances of combustibles are oxygen (O_2) and nitrogen (N_2) which, during a chemical reaction, do not release any heat or heat has to be supplied for the reaction to take place [2].

Fuel ballast consists of ash and water. Ballast is an undesirable fuel component. As the ballast ratio in the fuel increases the combustible fuel heat, or fuel efficiency decreases and the fuel price is lowered. When the ballast ratio in fuel is large, the fuel is not suitable for usage because transportation costs may be higher than its useful value [3].

Ash develops as the result of reaction of mineral substances present in biomass and oxygen. It is a solid residue resulting from the complete laboratory fuel combustion. It comprises mineral substances that are found in fuel. From the chemical point of view, biomass ash consists mostly of a mixture of oxides of inorganic elements K_2O, Na_2O, CaO, MgO, Fe_2O_3, Al_2O_3, SiO_2, P_2O_5, while their distribution in biomass is very variable (Fig. 1) [4]. Fuel quantity depends on conditions under which the combustion process takes place [5].

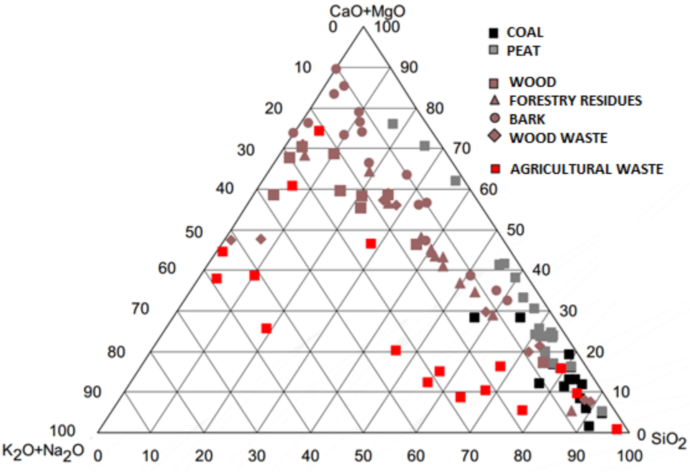

Figure 1. Main compounds present in biomass ash [6].

During the combustion of biomass or fuel in general, individual ash-forming elements react with flowing gas components and among themselves. In this way, the resulting ash becomes a mixture of complex compounds. Ash-forming elements are, e.g.: silicon, aluminium, iron, potassium, sodium, calcium, magnesium, sulphur, chlorine and phosphorus [7].

The concentration ranges of inorganic elements of biomass ash are variable in dependence on the biomass origin. From this reason, it is advantageous to classify them into groups or subgroups according to prediction of ash behaviour during combustion and during other energy utilization of biomass. The quantity of oxides in ashes or their distribution can vary in dependence on the ash temperature. When temperatures are high, elementary changes in biomass ash occur [8].

From ecological point of view, biofuels contain negligible amounts of heavy metals as cadmium and zinc. Particles containing these metals are most often parts of the finest fractions and are emitted into the environment. Larger heat sources therefore need a fine fraction filter. Unlike other fractions, fine particle fractions must not be used as fertilizers; they should be dumped in landfill sites [9].

In the process of fuel combustion, crude ash forms solid residue – ash which can be in the following forms [10]:

- Slag (cinder, sinters) – mineral substances contained in fuel that passed through the melting process during fuel combustion and created comprehensive glasslike mass,
- Scoria – mineral substances contained in fuel that went soft during combustion, sintered and formed a porous material,
- Ash – mineral substances found in fuel that remain in the form of a loose material during fuel combustion and fall through the grid to an ashbin,
- Fly ash – fine particles of solid residue (ash) that are, during the process of fuel combustion torn down by combustion gases from the fire place or from the fire chamber. They are captured in fly-ash separators or they are taken away by exhaust gases through a chimney to the atmosphere creating the emission fall of solid pollutants on the environment.

In order to ensure reliable operation of the combustion unit it is necessary to know the melting temperature of crude ash found in the fuel. According to its melting temperature, ashes are classified into [11]:

- low-melting ashes, the interval of ash melting temperatures is from 1000 to 1200 °C,
- medium-melting ashes, the interval ranges from 1200 to 1450 °C,
- high-melting ashes, the interval of ash melting temperatures is > 1450 °C.

Some biomass types have a high content of alkali oxides and salts. Thanks to it they feature a low value of ash meting temperature which might result in various problems during combustion or other energy utilization of biomass. Owing to the fact that fuel ash contains mixtures of various inorganic compounds, it does not have the exactly given melting value. Ash melting takes place in a wide temperature range, beginning with the deformation temperature, continuing with the softening and melting temperatures and ending with the temperature of flowing [12].

It is relatively complicated to maintain the temperature in the combustion chamber in such a range as to avoid the formation of sinters and aggregates. Though, it is possible to control the combustion temperature at least within a certain range so that the formation of sinters and aggregates will be significantly reduced [13].

Due to low values of the deformation temperature or ash melting temperature of some types of biomass, in the case of imperfect control in the combustion unit, the combustion temperature can exceed the critical value and, consequently ash sinters and sticks are formed on the heat exchanger surface and other components of the combustion unit, which reduces thermal efficiency of the device. Sinters, slags and sticks heat the combustion chamber with radiation, which results in the temperature increase of released exhaust gases [4]. Large pieces of sintered ash can mechanically damage parts of the combustion unit. The problems with ash behavior during fuel combustion are mostly connected with the use of phytomass [14].

Table 1 shows ash melting temperatures of some types of biomass obtained from numerous foreign papers, mainly from [15], [16], and [17].

Table 1. Phytomass ash melting temperatures in a dry state [4]

Ash	deformation temperature [°C]	softening temperature [°C]	melting temperature [°C]	flowing temperature [°C]
Phytomass – grasses and flowers				
Rapeseed	1005			above 1400
Artichokes – stem	1060		1130	1180
Grass	953	1023	1090	1165
Silver grass	940	990	1168	1240
Reed canary grass	1453	1513	1549	above 1586
Safflower	700		1430	above 1430
Thistle	970	1127	1308	1314
Average	973	1163	1318	1319

Ash can cause various problems in the combustion devices, e.g. it can prevent from heat transfer in heat exchangers, cause corrosion, preclude cleaning and ash removing, throttle the supply of fuel and combustion air and many others. The content of potassium, sodium, sulphur, chlorine and their compounds is monitored when using biofuels because during combustion the mentioned elements form a molten phase in which ash particles become sticky and adhere to the heat exchanger surfaces [18]. From the published sources [19], [20], [21] we can see that silicon, aluminium and iron achieve the highest variety of concentration from the elements forming biomass ash.

The causes of variable quality of fuel are quite unknown. Quantities of individual elements forming biomass ash differ in dependence on:

- type of biofuel (wood, plant, animal biomass),
- treatment of biofuel (decortication, refinement, etc.),
- parts of biofuel (trunk, branches, underground parts, etc.)
- way of biofuel cultivation (without or with human intervention, fertilization, spraying, etc.),
- places of biofuel cultivation (type of soil, altitude above sea level, incidence of solar radiation, etc.),
- other factors.

The presence of elements forming biomass ash is the result of chemical processes, intake of minerals from the soil and ways of biomass transportation [22]. Some of these elements are necessary for the plant growth [23]. The elements forming biomass ash are classified into macronutrients (potassium, calcium, magnesium, phosphorus and sulphur) and into micronutrients (iron, manganese and chlorine). Silicon, aluminum and sodium are not fundamental for the plant growth [18].

To avoid problems arising from the low melting temperature of biomass ash the following solutions can be implemented for its combustion:

- The use of special combustion units designed for the combustion of biomass with the low ash melting temperature.
- Adjustment of existing combustion units for combustion of biomass with low temperature of ash melting.
- Combustion of biomass having the low ash melting temperature with another fuel.
- Adding additives to biomass having the low ash melting temperature.

The article deals with the reduction of difficulties arising from the combustion of some biomass types when additives are added to increase the melting temperature. The experiments focused on the influence of six additives on the ash melting temperatures and ash-melting-temperature-related characteristics.

2. Materials Used and Methodology of Experimental Preparation of Samples

Silver grass was used in the experiment as an input raw material.

Silver grass has the low ash melting temperature; the deformation temperature reaches the value 940 °C. Relative moisture of the used raw material was 10%, heating value 17.6 $MJ.kg^{-1}$. Chemical composition of the raw material consists of 47.5 % carbon, 6.2 % hydrogen, 41.7 % oxygen, 0.73 % nitrogen, 0.15 % sulphur, 0.22 % chlorine. Ash of the used silver grass consisted of 54.4 % SiO_2, 4.7 % CaO, 2.3 % MgO, 0.1 % Al_2O_3, and 20.4 % K_2O.

The supplied raw material was crushed into fractions up to 10 cm. As the size was great for experiments, the fractions had to be made smaller. They were crushed in a hammer mill having the output approx. 20 $kg.h^{-1}$. After crushing, the fraction size did not exceed 4 mm. Crushed silver grass was then prepared to be mixed with various additives.

Additive is an addition agent (ingredient) added to some substance (product) with the objective to improve some of its properties. In practice, it is usual that with improving the properties new drawbacks begin to appear. Addition of additives can positively or negatively influence the increase of ash melting temperature or other properties or parameters of fuel. The influence of concrete additives can be only predicted before the experiment. For every used additive it is necessary to analyse its influences on all properties.

On the basis of knowledge and experience gathered by authors of scientific works concerning the chemical composition and possibilities of getting the particular additive, we used the following additives in our experiments:

- Kaolin – china clay kaolin Sedlec Ia was used as an additive for applications above 1300 °C. The used kaolin was in powder fractions (0-0.18 mm). The content of kaolinite ($Si_2Al_2O_5(OH_4)$) in the used raw material is 90.6 %. The composition of raw material is as follows: 45.7 % SiO_2, 35.8 % Al_2O_3, 0.1 % CaO, 0.3 % MgO, 1.3 % K_2O, 0.1 % Na_2O and 0.7 % Fe_2O_3.
- Talc – the talc we used is from the extraction site Horní Slavkov in the Czech Republic. It was crushed to the size up to 0.1 mm. The composition of raw material is as follows: 60.9 % SiO_2, 2.0 % Al_2O_3, 31.2 % MgO, 0.8 % Fe_2O_3 and 5.0 % H_2O+.
- Lime – originates from the processing of dolomitic limestone in the lime-pit Kosová Stráňavy Žilina district. It was crushed to the size up to 0.5 mm. Its composition is as follows: min. 86 % CaO, min. 5.0 % MgO, max. 7.0 % CO_2 and max. 2.0 % SO_3.
- Limestone – used as an additive was white crushed limestone of fraction particulates ranging from 0 – 0.5 mm. It is dolomitic limestone from the lime-pit Kosová Stráňavy in Žilina district. Its composition is as follows: min. 89 % $CaCO_3$, min. 7.0 % $MgCO_3$, max. 2.0 % SiO_2 and max. 2.0 % Al_2O_3 + Fe_2O_3.
- Dolomite – used as an additive was crushed white dolomite of fraction particulates ranging from 0 – 0.5 mm. It was supplied from the lime-pit Kosová Stráňavy in Žilina district. Its composition is as follows: 53 - 57 % $CaCO_3$, 42 - 46 % $MgCO_3$, 1.0 % SiO_2, 0.5 % Al_2O_3 and 0.4 % Fe_2O_3.

- Bentonite – used as additive was crushed grey bentonite with fraction particulates ranging from 0 – 0.5 mm. It is supplied from the lime-pit Jelšovský potok Kremnica district. Its composition is as follows: 57 - 61 % SiO_2, 18 – 21 % Al_2O_3, 2 – 3 %, 3.0 – 5.0 % MgO, 1.9 – 2.6 % CaO, 0.4 – 1.0 % K_2O, 0.2 – 0.7 % Na_2O, Fe_2O_3, 0.1 – 0.5 % FeO, and 0.2 – 0.3 % TiO_2.

In general, following key reactions among additives and compound containing potassium were expected, whereby following complex compound should be formed:

$$Al_2O_3.xSiO_2 + 2KCl + H_2O \rightarrow K_2O.Al_2O_3..xSiO_2 + 2HCl_{(p)} \qquad (1)$$

$$2KCl + H_2O_{(g)} + Al_2O_3 \rightarrow 2KAlO_2O + HCl_{(p)} \qquad (2)$$

$$2KCl + SiO_2 + 2H_2O \rightarrow K_2O.SiO_3 + 4HCl_{(p)} \qquad (3)$$

$$SO_{2(p)} + {}^{1}/_{2}\,O_{2(p)} \rightarrow SO_{3(p)} \qquad (4)$$

$$(NH_4)_2SO_{2(k)} \rightarrow 2NH_{3(p)} + SO_{3(p)} + H_2O_{(p)} \qquad (5)$$

$$Fe_2(SO_4)_{3(k)} \rightarrow SO_{3(p)} + Fe_2O_3 \qquad (6)$$

$$Al_2(SO_4)_{3(k)} \rightarrow SO_{3(p)} + Al_2O_3 \qquad (7)$$

$$SO_{3(g)} + 2KCl_{(p)} + H_2O_{(p)} \rightarrow 2HCl_{(p)} + K_2SO_4 \qquad (8)$$

$$KCl_{(p)} + CaHPO_42H_2O \rightarrow CaKPO_4 + 2H_2O + HCl_{(p)} \qquad (9)$$

$$K_2O.SiO_2 + 2CaCO_3 \rightarrow CaO.K_2O.SiO_2 + 2CO_{2(p)} \qquad (10)$$

Individual additives were added in the amount of 2 % with regard to the mass of the input raw material – silver grass.

3. Methodology of Experiments

During experiments and examinations of the influence of additive on biomass ash, the ash content, ash melting point and ash chemical composition were specified.

Ash content of the made pellet samples was determined in compliance with the standard STN EN 14775 [24]. Before the content was determined, the pellet samples were dried in a drying room at the temperature of 105 °C ± 2 °C. After drying, they were temporarily placed to a desiccator. The empty alumina bowl weighed with accuracy of 0.1 mg had been heated up to 550 °C ± 10 °C, and then cooled to the ambient temperature. Approximately 5 g of mixed sample was put into the bowl and spread on its surface. Again, the bowl was weighed with accuracy of 0.1 mg. The bowl was put into a cold furnace where it was heated according to the following procedure (Fig. 2):

- For 30 minutes the furnace temperature was evenly increased to 250 °C at the speed of 7.5 °C.min^{-1}.
- The temperature of 250 °C was maintained for 60 minutes to release volatile sample components before combustion.
- For 30 minutes the even increase of temperature continued up to 550 °C at the speed of 10 °C.min^{-1}.
- The temperature of 550 °C was maintained for minimally 120 minutes.

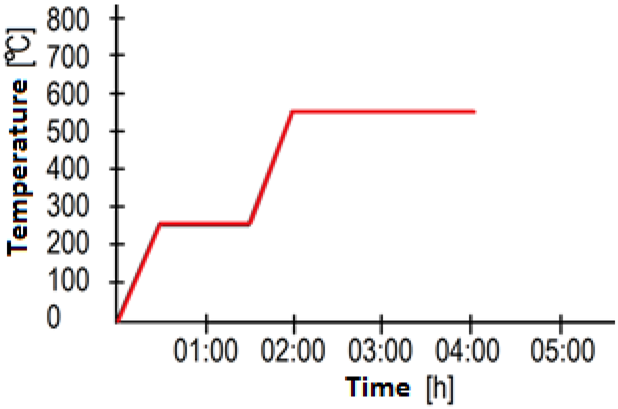

Figure 2. Course of temperature in furnace when biomass ash content was determined

The bowl with its content was taken from the furnace and after cooling it was weighed with accuracy of 0.1 mg. The ash content was determined from equation:

$$A_d = \frac{(m_3 - m_1)}{(m_2 - m_1)}.100\ [\%], \qquad (11)$$

where m_1 is the mass of the empty bowl in grams, m_2 is the mass of the bowl with the sample in grams, and m_3 is the mass of the bowl with ash in grams.

The ash melting temperature is a characteristic physical state of ash which takes place in its heating under precisely stipulated conditions [4]. When determining the ash melting temperatures in compliance with the standard STN ISO 540 [25], following temperatures were observed (Fig. 3):

- Ash deformation temperature (DT) – the temperature at which first signs of edge or border rounding due to melting appear.
- Ash softening temperature (ST) – the temperature at which, in the case that the height of the test specimen having the shape of a vertical pyramid or cone is identical to the width of its base or in the case that the test specimen having the shape of a cube or cylinder has its edges totally rounded without any change in the height.
- Ash melting temperature (HT) – the temperature at which the test specimen forms a semi sphere whose height is equal to the half of its base.
- Ash flowing temperature (FT) – the temperature at which ash spills over the pad in a layer whose height is approximately a third of the test specimen at the melting temperature.

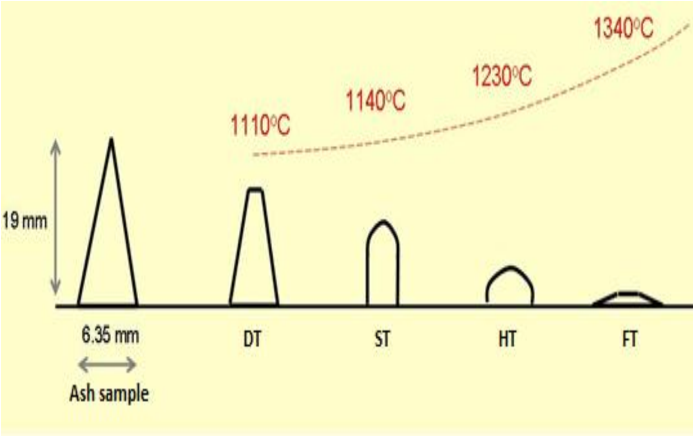

Figure 3. Change of ash specimen shape at determining ash melting temperatures

Chemical composition of ash was determined in a contract laboratory of a company by the emission spectrometric method with inductively coupled plasma. The percentage share of following compounds was determined: silicon oxide (SiO_2), lime oxide (CaO), manganese oxide (MgO), aluminum oxide (Al_2O_3) and potassium oxide K_2O.

4. Results of Experiments

Figure 4 shows the influence of various types of additives in the amount of 2 % contained in silver grass chaff on the amount of ash. The increase in the ash quantity was approximately equal to the amount of additive or exceeded this amount. It was caused by the fact that mineral additives did not contain any combustible and formed a ballast component of fuel. The higher increase in the fuel amount than the amount of additive could have been caused by the fact that the particular additive absorbed part of some substances which otherwise could have been found in burnt gases.

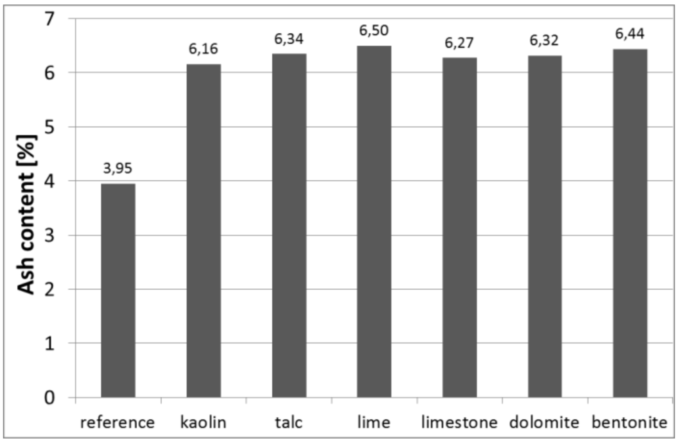

Figure 4. Influence of additives in silver grass chaff on the ash amount

Figure 5 displays the influence of various types of additives added in the amount of 2 % to silver grass chaff on the ash melting temperature.

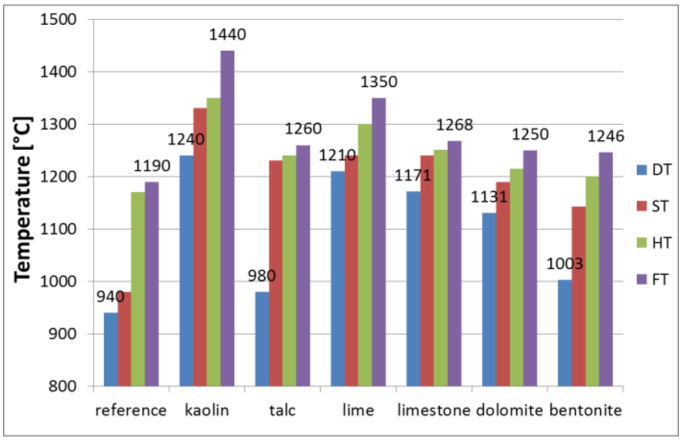

Figure 5. Influence of additives in silver grass chaff on the ash melting temperature

Figure 5 shows a relatively significant positive influence of all tested additives on all ash melting temperatures. After using mineral additives, individual melting temperatures of silver grass ash were higher by 2.6 – 35.7 % compared with the melting temperatures of the reference sample of silver grass without additives. The use of additives caused that easily molten ashes became medium molten ashes. Thanks to it, less problematic combustion in common heat sources would be possible. The biggest growth in the ash melting temperatures was seen when kaolin was used; the ash deformation temperature increased by 31.9 %, ash softening temperature increased by 35.7 %, ash melting temperature rose by 15.4 % and ash flowing temperature rose by 21 % in comparison to the individual ash melting temperatures of the reference sample. The most beneficial increase is the one by 300 °C in the deformation temperature whose value predicates most of possible problems during combustion due to the formation of slag and sinters.

Table 2 shows the influence of individual additives on chemical composition of silver grass ash.

Table 2. Influence of additives on chemical composition of silver grass ash

Sample	SiO2 content [%]	CaO content [%]	MgO content [%]	Al2O3 content [%]	K2O content [%]
Reference	54.4	4.7	2.72	0.22	20.37
Kaolin	33.8	9.3	2.1	19.92	8.4
Talc	56.9	4.2	9.4	0.29	11.8
Lime	17.3	14.9	4.3	0.2	5.1
Limestone	18.4	11.7	4.7	0.33	6.4
Dolomite	21.4	11.8	7.9	0.19	6.1
Bentonite	55.6	5.6	3.9	8.9	18.6

Tab. 2 shows the decrease in content of K_2O whose high content results in formation of eutectics with the low melting temperature. The greatest decrease in K_2O content was recorded when lime was used as additive; its content fell from 20.37 % to 5.1 %. In general, it can be said that 2 % addition of individual additive changed the content of individual compounds in dependence on chemical composition of the particular additive, e.g. addition of kaolin significantly increased Al_2O_3 as kaolin contains 35.8 % Al_2O_3. Similar situation was with the addition of lime which increased CaO content (from 4.7 % to 14.9 %) because lime contained 86 % CaO.

Simplifying the chemical composition of ash only to the content of SiO_2, CaO and K_2O, it is possible to plot their contents to the ternary phase diagram. From the chemical composition of silver grass ash with various additives in Table 2, relative amounts of SiO_2, CaO and K_2O were formed on the basis of following relations:

$$p_{SiO2} = \frac{\%SiO_2}{\%SiO_2 + \%CaO + \%K_2O} \tag{12}$$

$$p_{CaO} = \frac{\%CaO}{\%SiO_2 + \%CaO + \%K_2O} \tag{13}$$

$$p_{MgO} = \frac{\%K_2O}{\%SiO_2 + \%CaO + \%K_2O} \tag{14}$$

Relative quantities of SiO_2, CaO and K_2O are displayed in Table 3.

Table 3. Resultant relative quantities of SiO_2, CaO and K_2O in silver grass ash

Sample	p_{SiO2} [%]	p_{CaO} [%]	p_{K2O} [%]
Reference (1)	68.4535	5.914181	25.63231
Kaolin (2)	65.63107	18.05825	16.31068
Talc (3)	78.05213	5.761317	16.18656
Lime (4)	46.3807	39.94638	13.67292
Limestone (5)	50.41096	32.05479	17.53425
Dolomite (6)	54.45293	30.02545	15.52163
Bentonite (7)	69.67419	7.017544	23.30827

Having plotted the relative quantities of SiO_2, CaO and K_2O found in silver grass ash together with additives on the ternary phase diagram K_2O – CaO – SiO_2; it is possible to verify the influence of additives on the change of melting temperature of eutectic K_2O – CaO – SiO_2 (Fig. 6). It is a Gibbs triangle with the help of which properties of systems composed of three components are investigated. It uses the fact that the sum of relative representation of all three components is 100%. The mixture of any composition is displayed with a point inside the equal sided triangle. The concentrations of all three components are shown in mass percentage.

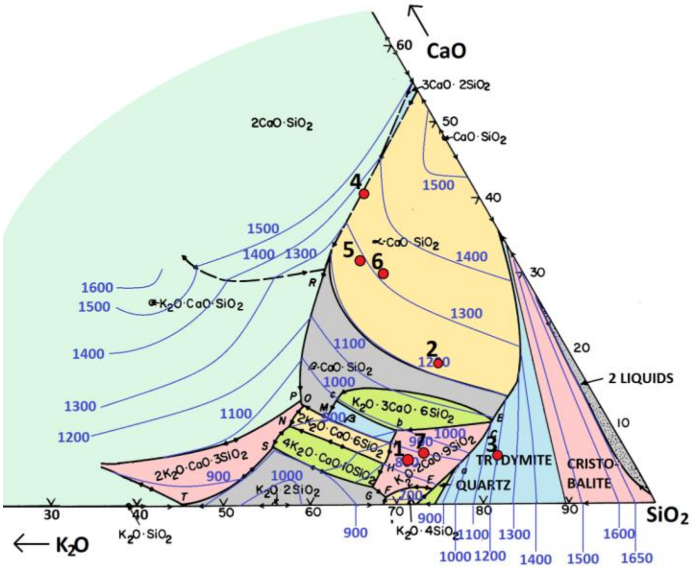

Figure 6. Influence of additive on the melting temperature of eutectic K_2O – CaO – SiO_2

In Fig. 6 the positive influence of additives on the melting temperature of eutectic K_2O – CaO – SiO_2, is obvious, which is related to the influence of additives on the ash melting temperature (Fig. 5). The highest increase in the melting temperature of eutectic K_2O – CaO – SiO_2 was recorded when lime was used when the position in the ternary diagram (and simultaneously the composition of eutectic) moved in the direction towards the upper angle of CaO. Thanks to it, the melting temperature of eutectic K_2O – CaO – SiO_2 increased approx. by 500 °C, which was significantly manifested in all ash melting temperatures (Fig. 5).

5. Conclusion

The adding of additives to biomass is one possibility of reducing combustion problems due to the low melting temperature of biomass ash. The article deals with the addition of six additives in the amount of 2 % to silver grass which is one of relatively common fast-growing energy crops with the low ash melting temperature. On the basis of the obtained results, the positive influence on ash melting temperature with all tested additives can be affirmed.

The highest increase of the ash melting temperature was recorded when kaolin and lime were used; they increased the ash deformation temperature by 300 °C or 270 °C. In the case of lime, it was caused mainly due to the change in the composition of eutectic K_2O – CaO – SiO_2, which resulted in the increase of the melting temperature of eutectic. In general, additives changed chemical processes in the ash during combustion, which changed chemical composition of ash; which was verified by experiments. The result is the higher ash content in all samples with additives in comparison to the reference sample.

Based on the results it can be confirmed that additives can be a highly efficient tool to reduce the problems caused by the low ash melting temperature of various biomass types. Though, to use additives in practice, it is necessary to verify the influence of additive on other properties of biofuels or elimination of additive shortage.

Acknowledgements

This paper has been elaborated in the framework of the project „Support research and development in the Moravian-Silesian Region 2013 DT 1 - International research teams“ (02613/2013/RRC) and the project APVV-0458-11 “Solving issues with low-melting ash during the biomass combustion”.. Financed from the budget of the Moravian-Silesian Region.

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