ORIGINAL

Effect of aluminosilicate powders on the applicability of innovative geopolymer binders for wood-based composites

Ali Shalbafan1 · Johannes Welling2 · Joachim Hasch3

Received: 13 March 2016 / Published online: 10 March 2017 © Springer-Verlag Berlin Heidelberg 2017

Abstract In this research, a new binder class for wood based composites, named geopolymer binder, was developed based on pozzolanic by-products (e.g. fly ash). Additionally, effects of different amounts of silica fume, as a replacement agent with other aluminosilicate components (e.g. fly ash and metakaolin), have been evaluated in the innovative binder. The Automated Bonding Evaluation System technique was used to characterize the bonding shear strength of the developed geopolymer binder. It was shown that the best shear strength for fly ash based binders was obtained by the lowest press temperature and longest pressing time. The addition of silica fume (from 20% up to 100%) significantly influenced the bonding shear strength in all binder types. Due to the chemical and mineralogical compositions, silica fume displays higher pozzolanic activity than metakaolin whereas fly ash shows lower strength in comparison to metakaolin. The silica fume (100%) based binder has also superior shear strength compared to those of conventional UF resin and other geopolymer binders. Bonding shear strength like that for UF resin was achieved by substituting only 20% silica fume in geopolymer binder compositions.

1 Introduction

Nowadays, wood based panels bonded with formaldehydebased adhesives are widely used in furniture manufacture and interior decoration. They release formaldehyde during the service life (Pizzi and Mittal [2003](#page-9-0)). Moreover, the sensitivity of the general public has grown with regard to environment and its protection, especially for formaldehyde which is harmful to people's health. Therefore, there is growing interest in finding alternatives for the synthetic binders currently used regarding reduction of formaldehyde emission from wood based panels. Inorganic binding materials can be used as an alternative approach (Sarmin et al. [2014](#page-9-1); Dukarska and Czarnecki [2016](#page-8-0)).

A form of inorganic material was discovered and further developed by Davidovits in the late 1970s (Shaikh [2013](#page-9-2)). The reaction of an aluminosilicate powder with a highly concentrated alkali solution produces a synthetic alkali aluminosilicate composition named "geopolymer" (Davidovits [2008](#page-8-1)) or broadly termed "inorganic polymer" (Provis and van Deventer [2009](#page-9-3)). The terminology and definitions of 'geopolymer' is more diverse and often conflicting depending on the raw material selection and processing parameters. Depending on the geopolymerisation process, it can be named geopolymer binder, geopolymer cement, geopolymer concrete, etc. From a terminological point of view, geopolymer cement is a binding system that hardens at room temperature, like regular portland cement. If a geopolymer compound requires heat setting, it may not be called geopolymer cement but rather geopolymer binder (Davidovits [2008](#page-8-1)). Geopolymer binder is an emerging class of mineral material that can be manufactured from the natural raw materials containing high amounts of silica (Si) and alumina (Al) mineral composition as well as industrial by-products, such as fly ash, silica fume (SF) etc., activated

 \boxtimes Ali Shalbafan ali.shalbafan@modares.ac.ir

¹ Department of Wood and Paper Science and Technology, Faculty of Natural Resources and Marine Sciences, Tarbiat Modares University, Noor, Iran

² Thünen-Institute (TI) of Wood Research, 21031 Hamburg, Germany

³ Kronopol Sp. Z o.o., ul. Serbska 56, 68-200 Zary, Poland

by the alkaline activator. Alkaline activator is an admixture from high alkaline solution (sodium or potassium silicate) with sodium or potassium hydroxide (Pacheco-Torgal et al. [2008](#page-9-4)).

Utilization of industrial wastes or by-products has been extensively investigated in geopolymer technology, due to the importance of sustainable technology developments. Among all of the waste products, fly ash and silica fume to be used as source materials for geopolymer has become a subject of growing interest (Shaikh [2013;](#page-9-2) Prud'homme et al. [2010a](#page-9-5)). Fly ash from power plants is available in large amounts and is relatively cheap compared to the other pozzolanic materials. Fly ash has two main classifications; low calcium (ASTM Class F) fly ash and high calcium (ASTM Class C) fly ash. Class F fly ash is more preferred as a source material than Class C, because the presence of calcium in high amounts may interfere with polymerization process and alter the geopolymer microstructure (Gourley [2003](#page-8-2)). Silica fume is also a by-product of the production of ferrosilicon industry and also of silicon metal. Silica fume is a highly effective pozzolanic material due to its screaming fineness and high silica content. Silica fume can be used in geopolymer admixture to improve its properties like compressive strength, bond strength and abrasion resistance (Thomas [2013\)](#page-9-6). Prud'homme et al. ([2010b\)](#page-9-7) studied the biocompatibility between wood and geomaterial compound as well as the mechanical performance of the resulted assemblies. They concluded that the interaction between geomaterial foam and wood leads to the formation of an interface with desirable mechanical properties. The mechanical performance of wood, geomaterial binder and two types of earth brick has been investigated by Gouny et al. [\(2013](#page-8-3)). They showed that geopolymer binder has a good adhesion between wood and earth brick. Brew and Mackenzie ([2007\)](#page-8-4) mentioned that the chemical and mineralogical composition of pozzolanic by-products, for example fly ash and silica fume, is far more complex than metakaolin and can be changed depending on the conditions of the manufacturing processes from which they were derived.

There is a lack of information about the curing characteristics of wood-based panels with geopolymer binders prepared under hot pressing conditions and concerning the panel's performance. The Automated Bonding Evaluation System (ABES) enables to scientifically explore the kinetics of adhesion and the bonding strength development of a diversity of wood adhesives (Humphrey [1993](#page-8-5)). Therefore, the ABES technique was used in this research to characterize the bonding shear strength of the geopolymer binder developed based on pozzolanic by-products (e.g. fly ash and silica fume).

In the present study, an attempt was made to produce geopolymer binder based on fly ash and to characterize its bonding shear strength by considering parameters such as different water glasses, the ratio of water glass solid content to the chemical base (W:B) and ratio of alkaline solution to aluminosilicate powder (S:P). Additionally, effects of different amounts of silica fume, as a replacement agent with other aluminosilicate components (fly ash and metakaolin), were evaluated on produced geopolymer binder. Finally, a comparative analysis was performed between geopolymer binders based on different aluminosilicate components and conventional urea formaldehyde (UF) resin to see their effectiveness to produce wood-based composites.

2 Materials and methods

2.1 Materials

Geopolymer was synthesized by alkaline activation of aluminosilicate materials obtained from natural minerals (e.g. metakaolin) or industrial by-products (e.g. fly ash and silica fume). Alkaline activator was an admixture of alkaline silicate (sodium silicate or potassium silicate) with chemical base [sodium hydroxide (NaOH) or potassium hydroxide (KOH)]. Shalbafan et al. ([2016\)](#page-9-8) concluded that the molar ratio of SiO_2 : M_2O (M = Na or K) has an important effect on the geopolymer binder characteristics. They found that the binder with a lower molar ratio of $SiO₂:M₂O$ resulted in higher bonding shear strength obtained by ABES. In this study, two types of water glass (one based on sodium silicate and the other based on potassium silicate) were used having low molar ratio of $SiO₂:M₂O$. The water glasses were supplied from Woellner GmbH (Germany). Detailed descriptions of the water glass used are presented in Table [1](#page-1-0).

Commercial potassium hydroxide flakes (KOH) supplied from BASF (Germany) were dissolved in water glass as the chemical base. The main aluminosilicate materials used in this study were fly ash and metakaolin. Silica fume was also used as substitute agent for fly ash and metakaolin. Class F fly ash according to EN 450-1 [\(2012\)](#page-8-6) was supplied by Holcim AG (Kiel, Germany). The metakaolin (Argical M1000) was supplied by Ferropem (France). Commercial silica fume (SIDISHIELD C25) was supplied by Elkem AS (Norway). The amount of amorphous $SiO₂$ and the specific

Table 1 Detailed description of water glasses

	Code Silicate type	Dry content (%)	Weight ratio Molar ratio	$(SiO_2:M_2O)^a$ $(SiO_2:M_2O)^a$
50T	Sodium silicate 44.5		2.5	2.6
42T	Potassium silicate	40	1.9	2.9

a Ratio A:B equal to 3.3 means 3.3 mol of A and 1 mol of B

surface area in this silica fume was more than 97.5% and $20 \text{ m}^2/\text{g}$, respectively.

2.2 Binder preparation

Firstly, potassium hydroxide was dissolved in an appropriate amount of water (depending on the final binder solid content). Then, the appropriate amount of water glass according to the recipe was added and mixed into the aqueous solution. Due to the exothermic reaction of KOH dissolution in water, the admixture should be cooled down to around room temperature (25 °C). Afterward, the desired aluminosilicate component was gradually added to the prepared alkaline solution and blended with a mixer (1000 rpm). Blending was performed until complete mixing of the ingredients had been achieved. The blending time was between 3 and 5 min, depending on the ratio of alkaline activator to aluminosilicate powder. The higher this ratio, the longer is the blending time.

2.3 Variable composition

Firstly, geopolymer binder was produced based on fly ash. Two different groups of weight ratios between binder compositions were tested in this binder; (1) weight ratio of solid content of water glass to the chemical base (W:B) which was varied between 0.66, 1, 1.33, (2) weight ratio of alkaline solution to the aluminosilicate powder (S:P) which was varied between 0.66, 1, 1.33, 1.66. In this series of tests, only one type of sodium silicate (50 T) and one type of potassium silicate (42 T) were used as water glass. The final solid content of the binder in each recipe was slightly changed to control and keep the specified ratio in the final binder composition. Developed geopolymer binder based on metakaolin has previously been tested and discussed in detail by Shalbafan et al. ([2016](#page-9-8)).

The substitution of fly ash and metakaolin with the silica fume was performed at the second series of tests. The amount of substituted silica fume was 0, 20, 40, 60, 80, and 100% (w/w) of its corresponding aluminosilicate powder (metakaolin or fly ash). Here, the ratios of W:B and S:P were kept constant at 1 and 1.33, respectively. A corresponding comparison was performed between geopolymer binders based on different aluminosilicate powder (metakaolin, fly ash and silica fume) within different ABES test protocols. Finally, the geopolymer binder (based on 50 and 42 T water glasses) was compared with conventional urea–formaldehyde resin with a similar ABES protocol. Table [2](#page-2-0) shows the compositions of the binders.

Table 2 Variable compositions of the geopolymer binder

a Ratio A:B equal to 3.3 means 3.3 mol of A and 1 mol of B

W water glass, *B* chemical base, *S* alkaline solution, *P* aluminosilicate powder

The mechanical shear strength of the samples was tested by means of ABES (Fig. [1\)](#page-3-0) on beech (*Fagus sylvatica*) veneer strips with a nominal thickness of 0.5 mm. Conditioned beech veneer strips $(25^{\circ}C, 65\% \text{ RH})$ with dimensions of $117 \times 20 \times 0.5$ mm³ were used for binder testing using ABES running different protocols. The veneer strips were glued together based on the standard configuration (5 mm overlapped strips) and mounted in the apparatus with an overlapping area of 100 mm². Then, adherent strips were pressed together at 1.2 N/mm² within the specified time. Bond strength evaluation was carried out almost instantly in shear mode (the ABES instrument was digitally controlled and pneumatically driven). It is believed that process parameters have significantly influenced the bonding shear strength. Therefore, the ABES tests were carried out at two pressing times (50 and 110 s) and two press temperatures (100 and 110° C). The adhesive amounts were kept constant in the entire test variations at 13.6 µl. Six replicates for each series of variations were conducted with ABES.

Corresponding comparison of the newly developed geopolymer binders with conventional urea–formaldehyde resin is an essential and determining factor for further developments. Conventional liquid urea–formaldehyde (UF) resin (Kaurit 350, BASF, Germany) was tested with ABES protocol similar to those for geopolymer binders. The UF resin was tested as received from the company (without any additives, e.g. hardener and water) with a solid content of 66%. Detailed protocols of ABES tests for binders are presented in Table [3](#page-3-1). It has to be mentioned that the UF partial degradation was not observed within these measuring parameters range (press time and temperature). Addition of hardener can accelerate the curing of UF resin. It is well understood that longer press time for cured UF resins gives lower curing performance due to partial degradation of the bond. To compare the bonding shear strength of UF resin and geopolymer binder in similar press times (50 and 110 s), no hardener was added to control the partial degradation of UF resin.

2.4 Statistical analysis

A one-way analysis of variance (ANOVA) of the bonding shear strength was performed with the SPSS software. Statistical differences between variations were evaluated by multiple comparisons based on a Tukey's HSD test due to the homogeneity of variances. The statistical significance was set at $P < 0.05$. Different capital letters denote significant differences of ingredients ratios and SF% in each test code and different small letters denote significant differences among test codes for each ingredients ratio and SF%.

3 Results and discussion

3.1 Characterization of geopolymer binder based on fly ash

3.1.1 Ratio of alkaline solution to aluminosilicate powder (S:P)

Bonding shear strength for geopolymer binder based on fly ash with different alkaline solution ratio to aluminosilicate powder (S:P) is presented in Fig. [2](#page-4-0)a, b). Generally speaking, Tukey HSD multiple comparison test showed that increasing the S:P ratio from 0.66 up to 1.66 resulted in the decreasing of shear strength in both geopolymer binder groups (Na 50 and K 42 T). It has to be mentioned that the binder solid content was decreased from 78 to 65%, while the S:P ratio was increased from 0.66 to 1.66. In geopolymer binder, the aluminosilicate powder acted as a binding

Table 3 Different protocols of ABES tests

Code		Pressing Time (s) Temperature $({}^{\circ}C)$ Amount (µl)	
A	50	100	13.6
B		110	
C	110	100	
Ð		110	

Fig. 1 Schematic illustration of the automated bonding evaluation system (ABES) set-up

Fig. 2 Effect of variable binder composition on bonding shear strength of fly ash based binder; **a** binder based on sodium silicate (Na 50 T) with S:P ratio of 0.66, 1, 1.33 and 1.66, **b** binder based on potassium silicate (K 42 T) with S:P ratio of 0.66, 1, 1.33 and 1.66,

c binder based on sodium silicate (Na 50 T) with W:B ratio of 0.66, 1 and 1.33, **d** binder based on potassium silicate (K 42 T) with W:B ratio of 0.66, 1 and 1.33

agent and the alkaline solution acted as the activator solution for the binding agent. Increasing the S:P ratio means that the amount of aluminosilicate powder (as a binding agent) is decreased, thereby leading to the weakening of the bonding strength. On the other hand, the amount of water glass (Na₂O and/or K₂O) that existed in the final binder mixture was elevated in binders with an higher S:P ratio.

Although the appropriate content of $Na₂O$ and/or $K₂O$ is very important for improved solubility of aluminosilicate powder, increasing their content can also weaken the geopolymerization process (Heah et al. [2012](#page-8-7)). However, the highest value of bonding shear strength was obtained for the binder with S:P ratio of 0.66, but it has to be noticed that the workability of such binder $(S:P=0.66)$ was decreased due to its high solid content (78%). It has to be mentioned that such geopolymer binder (higher ratio of S:P) can be a good candidate to be used for engineered wood products like plywood and glued/cross laminated timbers. For wood-based panels like particleboard or fiberboards, the binder should have spraying ability which can be obtained by lower S:P ratio (like 1.33). Additionally, better coating of wood surfaces can be achieved by a lower binder solid content or higher S:P ratio.

Fly ash geopolymer binder based on sodium silicate (Na 50 T) has higher bonding shear strength compared to those of potassium silicate (K 42 T). This is attributed to the molar ratio of $SiO₂:M₂O$ of the water glass used. The sodium silicate has a lower molar ratio of $SiO₂:M₂O$ compared to that of potassium silicate. The lower the molar ratio of SiO_2 : M_2O , the better was the bonding shear strength (Shalbafan et al. [2016](#page-9-8)).

The temperature and duration of curing time were found to be critical factors affecting the bonding shear strength of fly ash geopolymer binder (denoted by small letters in Fig. [2](#page-4-0)). Changing the pressing parameters of ABES has significantly influenced the bonding strength of fly ash geopolymer binder. Elevating temperature from 100 to 110°C in this research led to significant decrease of shear strength

in all of the S:P ratios. Nagral et al. ([2014\)](#page-8-8) reported that elevating the curing temperature more than 80 °C, lowered the compressive strength of fly ash geopolymer. The binder based on fly ash is quite sensitive to the curing temperature. The lower the curing temperature, the higher was the shear strength of the binder. On the other hand, the pressing time had a vivid and significant influence on the binder strength. Increasing of pressing time from 50 to 110 s led to the doubling of bonding shear strength, especially for the sodium silicate-based binder in all of the S:P ratios. Hence, it can be said that the pressing parameters for fly ash based geopolymer binder should be set up with a low pressing temperature and accordingly longer pressing time (protocol C).

3.1.2 Ratio of water glass solid content to chemical base (W:B)

The solid content of water glass (W) was the sum of its SiO₂ and M₂O (M=Na or K). Chemical base (B) used in this study was potassium hydroxide (KOH). A mixture of these materials formed the alkaline activator/solution which was used for dissolving of the aluminosilicate powder. Therefore, the ratio of W:B was evaluated in three levels of 0.66, 1 and 1.33, while the S:P ratio was kept constant at 1.33. Bonding shear strengths for geopolymer based on fly ash with different W:B ratios are presented in Fig. [2c](#page-4-0), d) and their statistical differences are denoted with capital letters in Fig. [2](#page-4-0) columns. The lowest bonding shear strength was obtained for the W:B ratio of 0.66. Low alkaline concentration prevents dissolution of fly ash, so there would not be enough dissolved aluminum for the formation of alkaline aluminosilicate, meaning that silica will be free in the mixture (Pacheco-Torgal et al. [2008\)](#page-9-4). It is also shown that increment of W:B ratio leads to the enhancement of bonding strength. Expressive strength increase was observed for the increased W:B ratio from 0.66 to 1. Xu and van Deventer ([2000\)](#page-9-9) also showed that the use of water glass raises the dissolution of the prime aluminosilicate materials. Zhaohu and Yunping [\(2001](#page-9-10)) also reported that increasing the amount of sodium hydroxide leads to the increased mechanical strength of geopolymer based on fly ash, due to the crystallization of excess silicate in the mixture. On the other hand, by varying the W:B ratio from 1 to 1.33, the shear strength of geopolymer binder did not significantly change. Lee and van Deventer ([2002\)](#page-8-9) mentioned that although excess alkaline activator increased dissolubility, it also results in the formation of an aluminosilicate gel during early stages which leads to mechanical strength decrease. For the fly ash based geopolymer, Palomo et al. ([1999\)](#page-9-11) mentioned that the increase of alkaline activator concentration up to a certain level leads to better mechanical strength, while more increasing concentration has a negative influence on geopolymer characteristics. It is worth mentioning that the higher alkalinity of the binder can result in increasing deterioration of the wood structures (Pizzi and Mittal [2003](#page-9-0)). Mai and Militz [\(2004](#page-8-10)) also reported that high pH value of water glass leads to the strength loss of wood. Hence, for the fly ash geopolymer binder produced in this study, the W:B ratio of 1 can be selected for further analysis.

A corresponding comparison between geopolymer binders based on sodium or potassium silicate, with similar ABES protocols, showed higher bonding shear strength for binder based on sodium silicate. As mentioned earlier, this was attributed to the molar ratio of $SiO₂:M₂O$ of the water glass used (Shalbafan et al. [2016](#page-9-8)). Xu et al. ([2001\)](#page-9-12) confirmed that decreasing of the molar ratio of $SiO₂:M₂O$ leads to the higher dissolution of silica and aluminum and accordingly higher mechanical strength was achieved.

Important factors influencing the binder characteristics are pressing time and temperature. Therefore, geopolymer binder prepared with different W:B ratio was also tested with varying press parameters and their statistical differences are denoted with small letters in Fig. [2](#page-4-0) columns. As mentioned earlier, the geopolymer binder based on fly ash was quite sensitive to press temperature. Shear strength was significantly decreased while the temperature was elevated from 100 to 110° C (Nagral et al. [2014;](#page-8-8) Wang et al. [2004](#page-9-13)). A contrary trend was perceived when the pressing time was increased from 50 to 110 s. The longer the pressing time, the higher was the bonding shear strength. The best shear strength was obtained at the lowest press temperature and longest pressing time (Test code C) which is also statistically confirmed by Tukey HSD multiple comparison test (denoted with small letter of a).

3.2 Substitution of aluminosilicate powder by silica fume (SF)

Influence of substitution of aluminosilicate materials (e.g. fly ash and metakaolin) with silica fume in geopolymer binder composition has been investigated. The amount of substituted silica fume in binder mixture was 0, 20, 40, 60, 80 and 100% of its aluminosilicate component (e.g. fly ash or metakaolin). The geopolymer binder was produced with constant W:B and S:P ratios of 1 and 1.33, respectively, due to their better binder workability and bonding shear strength. Bonding shear strength for geopolymer based on fly ash and metakaolin with different amounts of substituted silica fume are presented in Fig. [3](#page-6-0) and their statistical differences are denoted with capital letters in figure columns. It is obvious that the addition of silica fume (from 20% up to 100%) effectively influenced the bonding shear strength in both binder types. The higher the replaced silica fume, the higher was the bonding shear strength in both geopolymer binders (based on

Fig. 3 Substitution of silica fume (SF) in geopolymer binder based on; **a** fly ash and sodium silicate (Na 50 T), **b** fly ash and potassium silicate (K 50 T), **c** metakaolin and sodium silicate (Na 50 T), **d** metakaolin and potassium silicate (K 50 T)

sodium or potassium silicate). Prud'homme et al. ([2010a\)](#page-9-5) mentioned that the addition of silica fume to the geopolymer composition involved the modification of the chemistry and the porosity of the sample. They mentioned that the generation of porosity was probably due to the H_2 produced by water reduction. This dehydrogenase production behaved as a foaming agent. It has to be noticed that to attain a remarkable porous material, the amount of silica fume must be at least 50 wt%. In summary, the formation of this foam played a major role in the increase of the viscosity, leading to the consolidation of the material and its strengths. Prud'homme et al. ([2010b](#page-9-7)) have observed a strong interface between wood and geopolymer binder based on silica fume. They mentioned that inter-diffusion of potassium of wood and geopolymer was responsible for such strong interface. In fact, potassium migrated as potassium hydrogen carbonate by hydration effects which can be promoted at the interface. Such phenomena seem to be also responsible for the enhancement of bonding shear strength of geopolymer binders used in this study.

The statistical comparison showed that metakaolin based binder has a significantly higher strength compared to fly ash based binder in both cases of sodium and potassium silicate. Several researchers mentioned that metakaolin was preferred due to its high dissolubility in alkaline activator and improved mechanical strength (Xu and van Daventer [2000](#page-9-9); Shaikh [2013](#page-9-2)). Additionally, the use of sodium silicate in alkaline activator led to the slight increase of bonding shear strength. In this regard, the molar ratio of SiO_2 : M_2O (M = Na or K) plays an important role. The lower the molar ratio the higher was the bonding shear strength. A closer look at Fig. [3](#page-6-0) revealed that the increased trend of binder shear strength was more pronounced at the 60–100% silica fume addition. It has to be noticed that below the substitution ratio of 50%, the silica fume acts as a reinforcing agent, while at the higher ratio of 50%, the silica fume is the base material and metakaolin or fly ash

were the replaced agent. Geopolymer binder performances were governed by its base aluminosilicate materials (Davidovits [2008\)](#page-8-1).

The geopolymer binder performance was also tested with different pressing parameters using ABES (statisti-cally denoted with small letters in Fig. [3](#page-6-0) columns). It is shown that pressing time has a positive influence on the binder strength. The longer the pressing time, the higher was the shear strength. On the other hand, changing pressing temperature from 100 to 110° C in this study, showed a slight decrease of shear strength. As mentioned earlier, the sensitivity of geopolymer binder to the pressing time was more pronounced compared to that of pressing temperature (Wang et al. [2004\)](#page-9-13).

3.3 Comparison between different aluminosilicate powders

Comparison of geopolymer binders using different aluminosilicate components (e.g. fly ash, metakaolin, silica fume) with constant W:B ratio of 1 and S:P ratio of 1.33 is presented in Fig. [4.](#page-7-0) It is visible that metakaolin based binders have utmost shear strength compared to fly ash based binder. The uttermost strength was obtained for the geopolymer binder based on silica fume. It has to be considered that the chemical composition of fly ash is far more complex than metakaolin and can be varied depending on the conditions of their origin. Therefore, the chemistry of geopolymer binder formation from fly ash materials remains a challenge (Brew and Mackenzie [2007](#page-8-4)). Appearance and performance of metakaolin powder are very consistent due to the controlled nature of its processing (Rashad [2013](#page-9-14)). Wild et al. [\(1996](#page-9-15)) and Olufemi ([2013\)](#page-8-11) mentioned that silica

fume displays higher pozzolanic activity than metakaolin in geopolymer composition whereas fly ash shows lower activity in comparison to that of metakaolin. This can be explained by their chemical and mineralogical compositions, pozzolanic activity and disparity in their fineness. Hence, it is necessary to find the optimum use of substituted silica fume in final geopolymer binder composition to achieve the desired strength as well as its workability requirements for wood based panel application.

3.4 Geopolymer binders versus urea formaldehyde resin

It is necessary to compare the shear strength of geopolymer binder developed with conventional urea–formaldehyde (UF) resin to see its effectiveness to be used for woodbased panels manufacturing. The bonding shear strength of geopolymer binder based on different aluminosilicate components is shown in Fig. [5.](#page-8-12) For such comparison, the processing parameters for all binder types were kept constant; press temperature of 110°C, press time of 110 s, binder amount of 13.6 µl. It can be seen that the geopolymer binder based on fly ash has the lowest shear strength value, followed by metakaolin. The silica fume based binder has superior shear strength compared to that of conventional UF resin as well as other geopolymer binders based on fly ash and metakaolin. Bonding shear strength of UF resin was 2.48 N/mm². It has to be noticed that the shear strength of the UF resin can also be more increased by elevating the press temperature above 110° C which was not within the scope of this study (Ferra et al. [2011](#page-8-13)). Additionally, the bonding performance of UF resin can be changed depending on the type and amount of hardener which was not used

Fig. 4 Effect of aluminosilicate powder types used in geopolymer binder based on; **a** sodium silicate (Na 50 T), **b** potassium silicate (K 42 T)

Fig. 5 Comparison between urea formaldehyde versus geopolymer binders

in this study (for preventing of UF degradation). It also has to be stated that the binder performance in a laboratory hot pressing system would be different from the one of ABES system.

Even though, the best binder performance was observed for the geopolymer binder based on silica fume, it still has to be considered that silica fume is more expensive than metakaolin and fly ash. On the other hand, as can be seen from Fig. [3](#page-6-0), bonding shear strength like for UF resin can be achieved by substituting around 20% silica fume in geopolymer binder compositions. Production of wood-based panels with the best geopolymer binder variations is planned for future work.

4 Conclusion

The performance of the developed geopolymer binder was evaluated using ABES technique (bonding shear strength). The results showed that sodium silicate-based binders have a higher shear strength compared to potassium silicate. Although the shear strength was increased by lowering the S:P ratio from 1.66 to 0.66, binder workability for wood-based panel productions has to be considered. In this regard, the S:P ratios of 1 and 1.33 have good workability. Bonding shear strength was also raised, while the W:B ratio was elevated from 0.66 to 1.33. Since, increasing W:B ratio means an increase in binder alkalinity and consequently more wood cell deterioration, the moderate ratio of 1 can be used for further analysis. The silica fume was also used as substitution component by fly ash and metakaolin based binder. It was shown that increasing the substituted amount of silica fume from 0 to 100% in both binders positively and significantly enhanced the bonding shear strength. The more the addition of silica fume, the higher the shear strength was achieved. The binder based on 100%

silica fume had superior shear strength compared to the other geopolymer binder as well as UF resin. Moreover, the shear strength of the same UF resin can be achieved by the addition of around 20% silica fume with an appropriate pressing protocol.

Hence, with an optimum use of substituted silica fume in final geopolymer binder composition, desired shear strength as well as good binder workability for wood-based panel application can be achieved. Finally, geopolymer binders based on different aluminosilicate types showed good potential to be used for wood-based panel manufacturing. Further research is needed to characterize the wood based panels produced from different geopolymer binders.

Acknowledgements The work presented was sponsored by Swiss Krono Group. The authors would also gratefully acknowledge the Wöllner GmbH and Ferropem companies for supplying materials.

References

- Brew DRM, Mackenzie KJD (2007) Geopolymer synthesis using silica fume and sodium aluminate. J Mater Sci 42(11):3930–3993
- Davidovits J (2008) Geopolymer chemistry and applications. 3rd edn, 2011: Institute Geopolymer, Saint Quentin, France
- Dukarska D, Czarnecki R (2016) Fumed silica as a filler for MUPF resin in the process of manufacturing water-resistant plywood. Eur J Wood Prod 74(1):5–14
- EN 450-1 (2012) Fly ash for concrete. Definition, specifications and conformity criteria. European Standard, Brussels, Belgium
- Ferra JMM, Ohlmeyer M, Mendes AM, Costa MRN, Carvalho LH, Magalhaes FD (2011) Evaluation of urea-formaldehyde adhesive performance by recently developed mechanical tests. Int J Adhes Adhes 31:127–134
- Gouny F, Fouchal F, Pop O, Maillard P, Rossignol S (2013) Mechanical behavior of an assembly of wood–geopolymer-earth bricks. Constr Build Mater 38:110–118
- Gourley JT (2003) Geopolymers; opportunities for environmentally friendly construction materials. Conference on Adaptive Materials for a Modern society, Sydney, Institute of Materials Engineering, Australia
- Heah CY, Kamarudin H, Mustafa Al Bakri, A M, Bnhussain M, Luqman M, Khairul Nizar I, Ruzaidi CM, Liew YM (2012) Study on solids-to-liquid and alkaline activator ratios on kaolin-based geopolymer. Constr Build Mater 35:912–922
- Humphrey PE (1993) Device to test adhesive bonds. United States Patent, US005176028A. The State of Oregon Acting by and through the Oregon State Board of Higher Education on behalf of Oregon State University, USA
- Lee WKW, van Deventer JSJ (2002) The effect of ionic contaminants on the early-age properties of alkali-activated fly ash-based cements. Cem Concr Res 32(4):577–584
- Mai C, Militz H (2004) Modification of wood with silicon compounds. Inorganic silicon compounds and sol-gel systems: a review. Wood Sci Technol 37(5):339–348
- Nagral MR, Ostwal T, Chitawadagi M (2014) Effect of curing temperature and curing hours on the properties of geo-polymer concrete. Int J Comput Eng Res 4(9):1–11
- Olufemi FS (2013) Reactivity of cement combinations containing Portland cement, fly ash, silica fume and metakaolin. Int J Eng Res Appl 3(3):582–587
- Pacheco-Torgal F, Castro-Gomes J, Jalali S (2008) Alkali-activated binders: a review. Part 2. About materials and binders manufacture. Constr Build Mater 22(7):1315–1322
- Palomo A, Grutzek MW, Blanco MT (1999) Alkali-activated fly ashes. A cement for the future. Cem Concr Res 29(8):1323–1329
- Pizzi A, Mittal KL (2003) Handbook of Adhesive Technology. Marcel Dekker AG, Basel
- Provis JL, van Deventer JSJ (2009) Geopolymers; structures, processing, properties and industrial applications. Woodhead Publishing Limited, New Delhi, India, 454 p
- Prud'homme E, Michaud P, Joussein E, Peyratout C, Smith A, Arrii-Clacens S, Clacens JM, Rossignol S (2010a) Silica fume as porogent agent in geo-materials at low temperature. J Eur Ceram Soc 30(7):1641–1648
- Prud'homme E, Michaud P, Peyratout C, Smith A, Rossignol S, Joussein E, Sauvat N (2010b) Geomaterial foam to reinforce wood. Ceram Eng Sci P 31(10):3–10
- Rashad AM (2013) Metakaolin as cementitious material: history, scours, production and composition—a comprehensive overview. Constr Build Mater 41:303–318
- Sarmin SN, Welling J, Krause A, Shalbafan A (2014) Investigating the possibility of geopolymer to produce inorganic-bonded wood composites for multifunctional construction material—a review. BioResources 9:7941–7950
- Shaikh FUA (2013) Review of mechanical properties of short fibre reinforced geopolymer composites. Constr Build Mater 43:37–49
- Shalbafan A, Welling J, Hasch J (2016) Geopolymers as potential new binder class for wood based composites industry. Holzforschung 70(8):755–761
- Thomas M (2013) Supplementary cementing materials in concrete. Taylor & Francis Group, CRC Press, Boca Raton
- Wang K, Shah SP, Mishulovich A (2004) Effects of curing temperature and NaOH addition on hydration and strength development of clinker-free CKD-fly ash binders. Cem Concr Res 34(2):299–309
- Wild S, Khatib JM, Jones A (1996) Relative strength, pozzolanic activity and cement hydration in superplasticised metakaolin concrete. Cem Concr Res 26(10):1537–1544
- Xu H, van Deventer JSJ (2000) The geopolymerisation of aluminosilicate minerals. Int J Miner Process 59(3):247–266
- Xu H, van Deventer JSJ, Lukey GC (2001) Effect of alkali metals on the preferential geopolymerization of stilbite/kaolinite mixtures. Ind Eng Chem Res 40(17):3749–3756
- Zhaohu X, Yunping X (2001) Hardening mechanisms of an alkaline activated class F fly ash. Cem Concr Res 31(9):1245–9