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Screening industrial and organic waste for potential use in polyurethane adhesives

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Abstract: This study evaluated the potential use of 15 industrial and organic waste materials - including fly ash, bottom ash, fluidized bed ash, slag, photovoltaic glass, sulfur, lignin, biochar, textile fibers, hemp fibers, sawdust, eggshells, bamboo fibers, fluidized bed sand - as fillers in two-component polyurethane (2C PU) adhesives. The materials were characterized for chemical composition, particle size distribution, moisture content, calorific value, FTIR spectra, and metal leachability. Fly ash from pulverized coal combustion demonstrated the highest compatibility with the PU matrix among all tested materials. Adhesive formulations with various fly ash-to-chalk ratios were synthesized and tested for shear strength. The highest value (4.50 MPa at 20 °C) was obtained with 10% fly ash and 90% chalk, indicating a favorable synergistic effect. In contrast, the formulation containing 100% fly ash showed a substantial drop in performance at elevated temperature (0.10 MPa at 100 °C), revealing a thermal limitation. These findings suggest that fly ash may serve as a sustainable and cost-effective partial filler in PU adhesives, contributing to circular economy goals. However, its limited thermal stability must be considered for products intended for high temperature applications.

Introduction

Polyurethane (PU) is a versatile polymer widely used in various industries, including construction, automotive, aerospace, and biomedical sectors (Sikdar et al. 2022). Its properties, such as mechanical strength, flexibility, and chemical resistance, can be tailored by adjusting the ratio of soft and hard segments during synthesis, making it suitable for a wide range of applications (Hinrichsen 1994). Recent studies have focused on enhancing thermal stability, adhesion, and durability, which are essential for high-performance applications such as coatings, foams, adhesives, and elastomers (Shafiq et al. 2024). The global polyurethane market, valued at $89.3 billion in 2022, is projected to reach $120.1 billion by 2030, driven by increasing demand for sustainable packaging, automotive composites, and high-performance adhesives (Global Polyurethane Market 2021).

Two-component polyurethane (2C PU) adhesives are one of a key applications of polyurethane (PU) technology. They consist of a reactive polyol resin and an isocyanate hardener, forming strong and durable bonds. their excellent adhesion makes them t suitable for a wide range of substrates, including metals, wood, aluminum, and composites (Prociak 2016). In this study, wood-to-wood joints as a baseline were selected as a baseline to ensure consistent and controlled testing conditions and to isolate the effect of filler type on bond performance.

`Traditionally, PU adhesives are produced from petroleum-based raw materials, contributing to environmental pollution through greenhouse gas emissions and resource depletion. To address these challenges, researchers have focused on bio-based precursors and recyclable PU formulations (Ciastowicz et al. 2024). The transition to a circular economy has further emphasized the need for sustainable materials in the PU industry. Initiatives such as the European Green Deal and the Renovation Wave promote energy efficiency and the use of sustainable materials in construction, aligning with global efforts to reduce carbon emissions and waste (European Commission 2020; Lesiuk et al. 2012; Ragossnig and Schneider 2019). In this context, industrial and organic waste materials are increasingly recognized as viable fillers to reduce reliance on fossil-based resources and enhance polymer sustainability. Incorporating waste materials into PU adhesives, however, presents challenges. Variability in chemical composition, high moisture content, inconsistent particle size, and limited compatibility with polyurethane chemistry can compromise

performance. Functional groups such as hydroxyl (–OH), carboxyl (–COOH), and amine (–NH_2) may interfere with isocyanate reactions, affecting network formation and mechanical performance. Moreover, excess water or high calorific values can destabilize polymerization and pose safety risks during processing (Petrie 2007).

Previous studies have examined the use of various waste materials such as fly ash, biochar, eggshells, wood sawdust, and lignin as fillers in polyurethane systems, particularly in rigid and flexible foams, coatings, hotmelt, and thermoplastic polyurethanes [(Bartczak et al. 2022; Onwubu et al. 2025; Pęczek et al. 2024; Sharma et al. 2024; Shivakumar et al. 2024; Tombarkiewicz et al. 2022). However, comprehensive research on the optimal selection and processing of these materials for 2C PU adhesives is lacking. Moreover, the environmental and economic impacts of using waste-derived fillers in large-scale production remain insufficiently explored. Most previous works focused on a limited number of waste types and rarely addressed critical parameters such as moisture content, particle size distribution, functional group composition, calorific value, or metal leachability. Additionally, 2C PU adhesives differ significantly from other polyurethane systems. in formulation, application conditions, and curing behavior

This study evaluates 15 industrial and organic waste materials as potential fillers for 2C PU adhesives. These include eggshells, textile fibers, hemp and bamboo fibers, lignin, biochar, various types of ash, slags, sulfur, and photovoltaic glass. These materials were characterized for water content, organic matter content, combustible fraction, ash content, high heating value, particle size, and elemental composition, including carbon (C), hydrogen (H), nitrogen (N), sulfur (S), and oxygen (O). Additionally, Attenuated Total Reflection Fourier Transform Infrared (ATR-FTIR) spectroscopy provided insights into chemical structures of the examined materials. The most promising materials were selected for further testing in PU adhesive formulations based on the obtained results. To assess their feasibility as fillers, an initial synthesis was carried out to evaluate their compatibility with the PU system and their impact on adhesion properties. The ultimate goal was to identify waste-derived fillers capable of replacing conventional materials in the production of polyurethane adhesives, thereby contributing to circular economy principles and sustainable PU adhesive development.

Materials and methods

Materials

In this study, 15 different waste materials available in Poland were analyzed as potential fillers for two-component polyurethane (2C PU) adhesives. The materials included both organic and inorganic waste, sourced from the chemical, energy, textile, and biomass processing industries. The primary selection criteria were large-scale availability and alignment with circular economy principles, which support the feasibility of industrial implementation.

Lignin (sodium lignosulfonate, 58% lignin content) was supplied by Pro-export Chemicals, and biochar derived from sunflower husks was obtained from Treeden Group Sp. z o.o. Bamboo and hemp fibers were provided by Rettenmaier Polska Sp. z o.o., while textile fibers (a mixture of natural and synthetic bristles) came from Polfibra Noto Sp. z o.o. Eggshells were sourced from Zakład Przetwórstwa Jaj EVEGS Sp. z o.o., and spruce sawdust from Produkcja Palet i Opakowań Drewnianych.

Among the inorganic fillers, sulfur was supplied by Siarkopol Tarnobrzeg Sp. z o.o. Despite its limited use in polyurethane systems and the known risk of thermal degradation, it was included due to its very low moisture content and wides availability in the Polish market (Housecroft 2018). Glass from photovoltaic panels, obtained from the Department of Applied Bioeconomy at the Wrocław University of Environmental and Life Sciences, was selected based on its low moisture content and assumed high silica (SiO_2) content, typical of soda-lime glass. This composition is expected to provide a thickening effect in the adhesive matrix, similar to conventional silica-based fillers (Varshneya 2019).

Inorganic waste materials such as fly ash from biomass combustion and bottom ash were obtained from Enea Elektrownia Połaniec. Conventional fly ash, slag, fluidized bed fly ash, and sand from fluidized beds were provided by Veolia EKOZEC Sp. z o.o.

To ensure consistency throughout the manuscript, the following terminology is used: “fly ash” refers to ash from conventional coal combustion, “fly ash BC” denotes ash from pulverized coal combustion, and “fly ash BCFBB” refers to ash from biomass combustion in a bubbling circulating fluidized bed boiler. These types differ in combustion conditions and are expected to exhibit distinct physicochemical characteristics.

Characterization

All supplied raw materials were tightly sealed and stored under standard laboratory conditions (T = 23 ± 1°C, RH = 50 ± 5%) to minimize the influence of external factors on their properties. Sample preparation was carried out under the same controlled conditions. Each measurement of water content, organic matter content, high heating value, particle size, and elemental composition, including oxygen (O) content, was performed in quintuplicate. ATR-FTIR analysis was performed on a single representative sample. The results are presented as mean values to, ensure accuracy and statistical significance.
Water content was determined using an OHAUS MB90 moisture analyzer. Organic matter (OM) content was measured according to PN-EN 15169:2011 standard, using a muffle furnace SNOL 8.2/1100 LSM01 at 550°C. The combustible fraction (CF) was determined following PN-Z-15008-04:1993 standard using the SNOL 8.2/1100 LSM01 muffle furnace at 815°C.

The high heating value (HHV) was measured in accordance with ISO 1928:2020 using an IKA C200 calorimeter equipped with an IKA C248 oxygen station and a C5010 calorimetric bomb. Measurements were performed at 17-25°C under an oxygen pressure of 30 bar.

The particle size was determined according to ISO 3310-1:2016 using a set of laboratory sieves (MULTISERW-Morek) with mesh sizes of 2, 1.0, 0.5, 0.25, 0.15, and 0.063 mm .

Prior to elemental (CHNS) analysis, all samples were oven-dried at 105 °C for 14 hours to remove physically bound moisture. Elemental composition (C, H, N, S) was determined following PN-EN ISO 16948:2015-07 using a PerkinElmer model 2400 Series analyzer (Waltham, MA, USA), as described by Stegenta-Dąbrowska et al. (2024).

ATR-FTIR spectra were recorded using a Perkin-Elmer Spectrum Two spectrometer equipped with a single-reflection UATR accessory. Spectra were collected in the range of 4000–450 cm-1, with a resolution of 4 cm^{-1}, using 4 scans and 3551 data points.

Leaching tests were performed in accordance with PN-EN 12457-4:2006 standard. Water extracts were analyzed for pollutant indicators following PN-EN ISO/IEC 17025:2018-02 standard. The test consisted of a one-step static leaching procedure with a total extraction time of 24 hours, including 6 hours of intensive stirring followed by and 18 hours of rest. After extraction, the eluates were filtered through 0.45 mm paper filters.

The filtered extracts were analyzed for leachable metals and other indicators of pollution, such as barium (Ba), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), molybdenum (Mo), nickel (Ni), lead (Pb), zinc (Zn), selenium (Se), antimony (Sb), arsenic (As), chlorides (Cl), fluorides (F), sulphates (S), dissolved organic carbon (DOC), and total dissolved solids (TDS). Zn, Cu, Ba, Pb, Ni, Mo, Cr, and Cd were qualified using ICP-OES (Perkin-Elmer Optima 7300 DV) in accordance with instrument following the PN-EN ISO 11885:2009 standard. As, Se, and Sb were analyzed by ICP-MS using a PerkinElmer NexIon 2000 instrument following the PN-EN ISO 17294-2:2016-11 standard. Hg was analyzed using the AAS-CV technique on a PerkinElmer PinAAcle 900T instrument following the PN-EN ISO 12846:2012 section 7 and PN-EN ISO 12846:2012/Ap1:2016-0 standards.

Shear strength was determined under compressive loading in accordance with ISO 6238:2018. Wood-to-wood joints were used as substrates for adhesion testing. Six adhesive formulations were prepared, each containing a different chalk-to-fly ash ratio, replacing 50% of the total filler content. Each formulation was synthesized once, and 10 wood-to-wood joints were bonded using the resulting adhesive. Priot to testing, samples were conditioned at +23°C ± 1°C and 50% ± 5% RH and until complete curing was achieved. Shear strength measurements were conducted using an Instron 3367 testing machine.

Synthesis

Component A was formulated using 46% plant-based triglycerides of fatty acids, 1% short-chain polyol (ethane-1,2-diol), 50% filler, 0.5% antifoaming agent, and 2.5% drying agent. The synthesis process began with drying the polyols and filler under vacuum conditions (50 bar) at 65°C for 2 hours. Subsequently, the antifoaming agent and drying agent were added, and the mixture was stirred for 15 minutes under the same conditions. Polymeric diphenylmethane diisocyanate (p-MDI) was used as component B, serving as the isocyanate component. The adhesive synthesis was carried out using a PC Labor system planetary disperser. Components A and B were mixed in a 100:20 ratio, corresponding to a reaction index of 1.07.

In the formulations, the 50% filler content was replaced with various combinations of chalk and fly ash: 100% chalk (standard filler), 100% fly ash instead of chalk, 75% fly ash and 25% chalk, 50% fly ash and 50% chalk, 25% fly ash and 75% chalk, 10% fly ash and 90% chalk. Technical chalk supplied by Trzuskawica S.A. was used as the reference filler in the adhesive formulation. According to the manufacturer's specification, the chalk consists predominantly of calcium carbonate ($CaCO_3$, ≥97.5%) with minimal impurities (Fe_2O_3 ≤0.1%). The particle size distribution is characterized by a residue of ≤0.1% on a 0.063 mm sieve.

Criteria of waste selection of waste for recycling

The selection of raw materials for further testing was based on prior experience and the specific requirements of 2C PU adhesive production. The aim was to identify waste materials that do not interfere with polyurethane polymerization, ensure safe processing, and contribute positively to final adhesive performance. The following criteria were applied:

1) Moisture content was required not to exceed 0.8%, as elevated water levels can disrupt the isocyanate-polyol reaction and reduce adhesive strength. Moreover, excess moisture would necessitate pre-drying, leading to additional processing costs and extended production time.

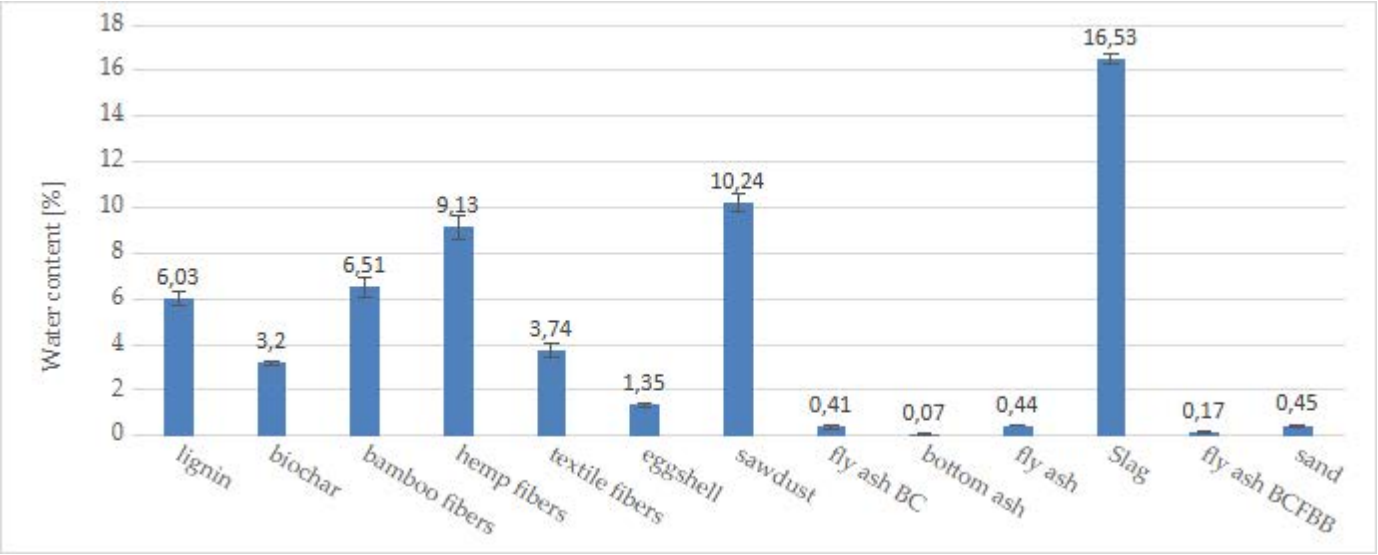

Fig 1. Water content (% by weight) in selected industrial and organic waste materials evaluated as potential fillers for polyurethane adhesive formulations.

2) Materials were selected if no more than 70% of their mass remained on a 0.25 mm mesh sieve, indicating that the dominant particle fraction was below 250 µm. This ensured good dispersion, minimized agglomeration, and eliminated the need for additional grinding.
3) FTIR spectra were analyzed to identify functional groups that could affect polymerization (Table 1). While hydroxyl (-OH) groups may support bonding, excessive amounts can lead to over-crosslinking and brittleness. Amino ($-NH_2$), carboxyl (-COOH), and thiol (-SH) groups were considered undesirable in excess, as they can destabilize the curing process.
4) A low heating value was preferred to minimize thermal risk during processing. High combustion energy can lead to exothermic reactions or gas release, compromising safety and material stability.
5) The presence of metals such as tin was avoided, as they may act as catalysts and induce uncontrolled reactions or the formation of undesirable products.
6) All materials were evaluated for pollutant leaching. Only those that did not exceed regulatory thresholds for heavy metals and other hazardous elements were considered acceptable.

Results

Chemical and energetic properties.

Materials with the lowest water content (Figure 1), such as glass and sulfur (<0.01%), are practically free of moisture due to their low surface polarity and non-porous structure, which limit physical adsorption (physisorption) of water molecules. This low affinity for moisture is typical for inorganic substances lacking polar functional groups and significant surface activity (Švábová et al. 2012). Since these values are below the detection limit of the measuring equipment used, moisture can be considered virtually absent in these materials, and their resistance to water absorption from the environment remains very high. Based on the data presented in Figure 1, slightly higher water contents were recorded for bottom ash (0.07%) and fly ash from biomass combustion in a fluidized bed boiler (fly ash BCFBB) (0.17%). Part of this moisture may originate from the environmental adsorption due to the porous structure of these materials. Similarly, fly ash from biomass combustion (fly ash BC), which represents the fine fraction of ash carried in the flue gas stream, contains a certain amount of water depending on fuel properties and combustion parameters. In this case, moisture may also result from steam condensation during transport and storage.

The highest moisture content among the analyzed materials was found in slag (16.53%). Such a high value indicates significant porosity, which promotes water retention. Slag, formed as a byproduct of high-temperature processes, may contain both chemically bound water and moisture retained within its structure through adsorption. This high-water content makes its potential use as a filler in polyurethane adhesives problematic, as intensive drying would be required to prevent interference with chemical reactions. For organic materials such as lignin, sawdust, bamboo fibers, hemp fibers, and textile fibers, the water content is significantly higher compared to inorganic materials (except for slag). This behavior results from their high hygroscopicity, which is directly related to

Table 1. Functional groups identified in tested waste materials and their predicted influence on polyurethane adhesive reactivity and performance.

Functional Group	Absorption Band [cm^{-1}]	Impact on PU Adhesive
Hydroxyl Groups (-OH)	3200–3600	The presence of OH groups in waste materials can improve bonding with isocyanate, but excess OH groups may lead to over-crosslinking of the adhesive, affecting its flexibility.
Amino Groups ($-NH_2$, -NHR)	N-H stretching 3300–3500	Amino groups can interfere with the polymerization reaction of isocyanate and polyol, leading to issues with the adhesive's quality. Materials containing these groups should be avoided.
Carboxyl Groups (-COOH)	O-H Stretching: 2500–3300 cm^{-1} C=O Stretching: 1700–1750 cm^{-1}	Carboxyl groups can affect the hardness of PU adhesives, but their excess will reduce flexibility, limiting the adhesive's use in applications requiring flexibility.
Thiol Groups (-SH)	2550–2600 cm^{-1}	The presence of thiol groups can introduce unwanted chemical reactions with isocyanate, compromising the adhesive's stability. These groups should be avoided in waste materials.
Isocyanate Groups (-NCO)	2250–2280 cm^{-1}	Isocyanate groups in waste materials can cause excessive crosslinking in the adhesive, resulting in increased stiffness and difficulty in processing.
Water (H_2O)	O-H Stretching: 3200–3600 cm^{-1}	Water in waste materials can influence the polymerization reaction, slowing it down and leading to reduced adhesive strength and durability.
Urethane Groups (-NHCOO-)	C=O Stretching: 1700–1730 cm^{-1}	Urethane groups can react with isocyanate, forming additional bonds in the adhesive structure. However, excess of these groups may make the adhesive too stiff, reducing its flexibility.

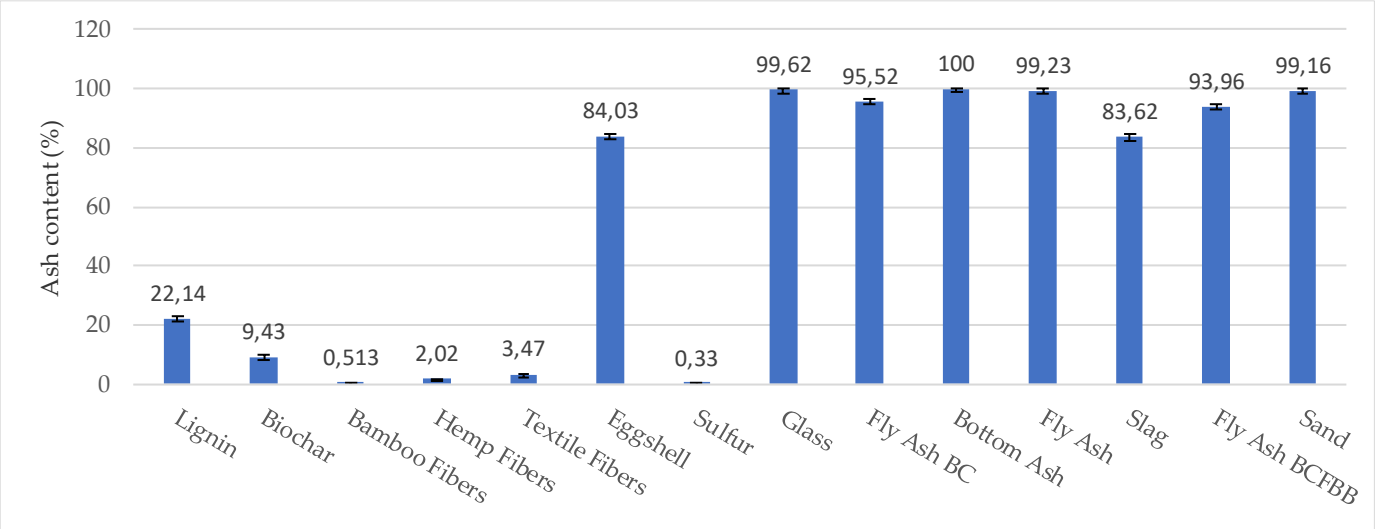

Fig 2. Ash content (% by weight) in organic and inorganic waste materials tested as potential fillers in PU adhesive formulations.

the presence of hydroxyl (-OH) groups in lignin, cellulose, and hemicellulose. These functional groups readily form hydrogen bonds with water molecules. The elevated moisture content in these materials presents a major challenge for their use as fillers in polyurethane adhesives, as excess water can disrupt the polyaddition reaction, leading to structural defects and undesirable porosity in the adhesive joint.

In summary, inorganic materials, especially glass and sulfur, exhibit minimal moisture content, making them suitable for use in polyurethane adhesive formulations. In contrast, slag exhibits exceptionally high-water content, significantly limiting its applicability without proper preparation. Although organic materials have lower moisture levels than slag, they are highly susceptible to moisture absorption, which necessitates prior drying before their introduction into polyurethane systems.

Analysis of ash content in the investigated waste materials (Figure 2) revealed clear differences between mineral and organic wastes. Mineral wastes exhibited consistently high ash content, ranging from 100% for bottom ash to 95.52% for fly ash generated from biomass combustion. In contrast, organic wastes showed significantly lower ash contents, with values below 0.01 % for sawdust and up to 22.15% for lignin. An exception was observed for eggshells, which exhibited a high ash content of approximately 84.03%, attributable to their chemical composition dominated by calcium carbonate ($CaCO_3$), an inorganic compound.

Table 2. Chemical and elemental composition of selected waste materials for potential use in polyurethane adhesives.
** combustible fraction [CF], organic matter [OM], *The measured value falls below the detection capability of the instrument

Name of the raw material waste	CF** [%]	OM** [%]	C [%]	H [%]	N [%]	S [%]
lignin	77.86 ± 0.26	77.02 ±0.36	41.52 ±0.28	4.46 ±0.36	0.68 ±0.05	6.71 ±0.26
biochar	90.75 ±0.30	90.56 ±0.40	72.00 ±0.17	3.50 ±0.27	0.90 ±0.01	0.06 ±0.01
bamboo fibers	99.47 ±0.26	99.47 ±0.26	42.45 ±0.41	6.30 ±0.39	≤ 0.01*	1.76 ±0.16
hemp fibers	97.98 ±0.36	97.48 ±0.32	44.21 ±0.36	6.05 ±0.36	0.35 ±0.01	1.37 ±0.20
textile fibers	96.53 ±0.28	96.23 ±0.28	58.51 ±0.26	6.74 ±0.25	12.02 ±0.20	1.82 ±0.08
eggshell	15.97 ±0.15	14.95 ±0.14	14.55 ±0.18	0.32 ±0.08	0.93 ±0.11	0.17 ±0.01
sawdust	100 ±0.30	100 ±0.20	49.39 ±0.13	6.14 ±0.28	≤ 0.01*	1.75 ±0.03
sulfur	99.66 ±0.24	99.66 ±0.23	0.09 ±0.02	≤ 0.01*	≤ 0.01*	91.96 ±0.95
glass	0.38 ±0.05	0.38 ±0.02	0.23 ±0.01	≤ 0.01*	≤ 0.01*	≤ 0.01
fly ash BC	4.48 ±0.04	4.47 ±0.02	3.43 ±0.05	0.06 ±0.02	≤ 0.01*	0.99 ±0.02
bottom ash	< 0.01*	< 0.01*	≤ 0.01*	≤ 0.01*	≤ 0.01*	≤ 0.01
fly ash	0.37 ±0.03	0.26 ±0.01	0.31 ±0.04	≤ 0.01*	≤ 0.01*	0.30 ±0.04
Slag	16.38 ±0.46	15.98 ±0.18	7.58 ±0.11	0.03 ±0.01	≤ 0.01*	0.26 ±0.06
fly ash BCFBB	6.04 ±0.18	5.9 ±0.12	6.04 ±0.36	≤ 0.01*	≤ 0.01*	≤ 0.01*
sand	0.83 ±0.06	0.83 ±0.07	0.03 ±0.01	≤ 0.01*	≤ 0.01*	0.13 ±0.02

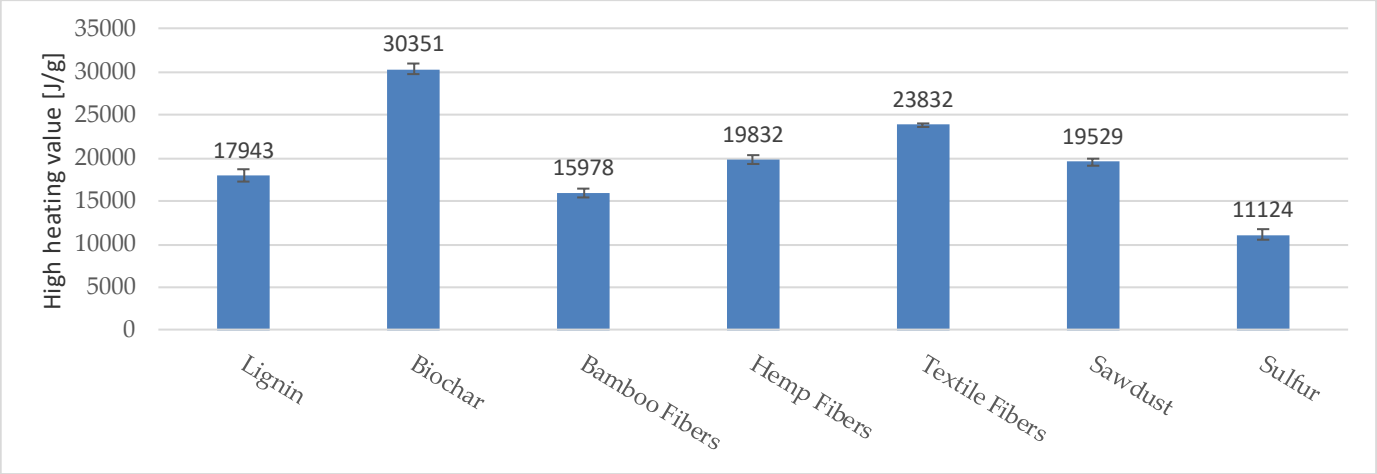

Fig 3. High heating value (HHV, J/g) of selected organic and waste-derived materials evaluated as potential fillers in polyurethane adhesives.

Mineral wastes such as glass, fly ash BC, bottom ash, conventional fly ash, slag, fly ash BCFBB, and sand displayed higher ash contents compared to organic materials primarily because they consist mainly of minerals and inorganic compounds that do not decompose during combustion and therefore remain as ash. In contrast, organic wastes contain a high proportion of combustible organic matter, which is largely oxidized during thermal treatment resulting in significantly lower ash residues. Additionally, many mineral wastes originate from industrial processes such as coal combustion, which results in high ash content.

Organic wastes such as lignin, biochar, bamboo fibers, hemp fibers, textile fibers, and sawdust originate from natural sources and contain relatively small amounts of inorganic constituents capable of forming ash. Sulfur, despite being classified as a mineral waste, exhibited a low ash content of 0.33%. This behavior can be explained by its chemical structure, high reactivity, and low level of inorganic impurities resulting in minimal residues after combustion. During combustion, sulfur is predominantly converted into gases such as sulfur dioxide (SO_2), leaving minimal solid residue in the form of ash.

Organic waste materials generally exhibit higher values of organic matter and combustible fraction (Table 2). Sawdust showed a total combustion value of 100% for both parameters, while other organic wastes ranged from 77.86% to 99.48%, with the exception of eggshells, which exhibited a low combustible fraction of approximately 14%. The reduced combustibility of eggshells can be attributed to their organic and mineral composition, which is less prone to thermal degradation. Elemental analysis of waste materials revealed the highest carbon (C) content in biochar, reaching 72%. This result is expected, as biochar is produced through the pyrolysis of biomass, a process involving the thermal decomposition of organic materials, which concentrates carbon. In contrast, the lowest carbon content (< 0.01 %) was observed in bottom ash, due to its mineral composition, consisting mainly of inorganic substances remaining after fuel combustion.

Organic waste materials showed carbon contents ranging from 14.55% (eggshells) to 58.51%, excluding biochar. The carbon content in organic materials is related to their natural structure, which contains large amounts of organic compounds such as cellulose, lignin, and other hydrocarbons. Eggshells again represent an exception due to their primary composition of calcium carbonate ($CaCO_3$), which accounts for their comparatively low carbon content. Mineral waste materials generally exhibited low carbon contents ranging from 0% to 7.58% (slag), which aligns with their inorganic nature. Materials such as glass, ash, and slag are largely devoid of carbon-based compounds, resulting in minimal or negligible carbon levels. Analysis of nitrogen (N) content showed that most materials contained levels between <0.01% and 1% nitrogen, which is typical for both organic and mineral materials. Textile fibers were an exception, likely due to the presence of proteins (e.g., keratin) or nitrogenous compounds used during their production processes. The highest sulfur (S) content was recorded in sulfur waste, reaching 91.96%, which reflects its composition as nearly pure elemental sulfur. In other materials, sulfur was present only in trace amounts, as is typical for both organic and mineral wastes unless sulfur is a primary constituent.

Mineral wastes exhibited a low higher heating value (HHV) of <1 J/g, except sulfur, which had an HHV of 11124 J/g [Figure 3]. The absence of a measurable heating value in most mineral wastes can be attributed to their inorganic composition, which lacks combustible materials. Mineral wastes such as glass, ash, and slag primarily consist of inert, non-flammable compounds such as silicates, oxides, and other minerals that do not release energy during combustion. These materials remain stable at high temperatures and therefore contribute no calorific value. Sulfur, on the other hand, represents an exception due to its unique chemical properties. As an elemental substance, sulfur undergoes oxidation during combustion, releasing significant energy in the form of heat, which accounts for its elevated HHV of 11.124 J/g. This makes sulfur one of the few mineral components with notable energetic potential, as it combusts to form sulfur dioxide (SO_2), generating considerable thermal energy. In contrast, organic wastes showed significantly higher HHV values, ranging from 15.978 J/g for bamboo fibers to 30.351 J/g for biochar. This substantial difference is due to the organic nature of these materials, which are rich in carbon-based compounds such as cellulose, hemicellulose, lignin, and

other hydrocarbons. These compounds possess high energy content because they readily undergo combustion to release heat.

The particle size distribution analysis for various materials, shown in Appendix 1, revealed significant differences in their suitability as potential fillers, particularly in terms of their ability to pass through sieves with specific mesh sizes. Materials such as lignin, biochar, fly ash, and glass exhibited the first retained residues on a 0.25 mm mesh sieve, indicating their suitability for filler applications. Sulfur and bamboo fibers showed residues on a 0.50 mm mesh sieve, suggesting potential use as fillers , although further processing may be required. In contrast, materials such as hemp fibers, textile fibers, eggshells, sawdust, slag, and bottom ash showed substantial residues on larger mesh sizes (2 mm), indicating that they are too coarse to be used as fillers. Specifically, hemp fibers and textile fibers, as well as eggshells, did not pass through 2 mm mesh sieve, rendering them unsuitable for use as fillers in many applications.

In summary, materials with smaller particle sizes, such as lignin, biochar, fly ash, glass, and sand, demonstrate greater potential as fillers, whereas coarser materials such as hemp fibers, textile fibers, and eggshells either require further processing or are unsuitable for this purpose.

Functional group analysis

The identification of functional groups in waste materials was conducted using ATR-FTIR spectroscopy. The results of the analysis are presented in Table 3, which includes the identified absorption bands and the corresponding functional groups. The ATR-FTIR spectra are provided in Appendix 2. Functional group assignments were based on established FTIR literature sources (Socrates 2004).

Hydroxyl groups (–OH), identified in lignin, bamboo fibers, hemp fibers, textile fibers, sawdust, and eggshells, can react with isocyanates, contributing to bond formation in the polyurethane structure. This reaction is central to the polymerization process, where strong bonds are formed that determine the hardness and stability of the final product. A high concentration of hydroxyl groups can enhance the reaction; however, an excess may lead to over-crosslinking, thereby reducing the adhesive's flexibility. For example, bamboo and hemp fibers show peaks in the 3200–3600 cm^{-1} range, corresponding to O–H stretching and indicating a significant presence of hydroxyl groups. This behavior is typical of lignocellulosic materials. However, the contribution of physically adsorbed water cannot be ruled out, as the samples were not dried before analysis. These bands can influence

Table 3. Analysis of ATR-FTIR Spectral Data for Waste Materials.

Name of the raw material waste	Absorption bands (cm^{-1})	Assigned vibrations
lignin	3304, 1595, 1508, 1420, 1270, 1134, 1031	OH (3400-3200); C=O (1700-1600); C=C aromatic (1510-1450); C-O (1300-1200); C-H (1120-1000)
biochar	2903, 2850, 1605, 1400, 1210	C-H (3000-2800); C=C (1620-1570); C-H (1420-1380); C-O (1250-1150)
bamboo fibers	3315, 2900, 1620, 1314, 1159, 1029, 557	OH (3350-3200); C–H (CH_2/CH_3 stretching) (2900 – 2850); C=C (1640-1590); CH_2 (1430-1380); C-O (1250-1150); Si-O (1100-1000); out-of-plane bending (700–500)
hemp fibers	3332, 2900, 1603, 1423, 1320, 1237, 1036, 558	OH (3350-3200); C–H (CH_2/CH_3 stretching) (2900 – 2850); C=C (1640-1590); CH_2 (1430-1380); C-O (1250-1150); Si-O (1100-1000); out-of-plane bending (700–500)
textile fibers	3299, 2918, 1635, 1449, 1234, 1032, 556	OH (3350-3200); C–H (CH_2/CH_3 stretching) (2900 – 2850); C=C (1640-1590); CH_2 (1430-1380); C-O (1250-1150); Si-O (1100-1000), out-of-plane bending (700–500)
eggshell	3550, 2918, 1638, 1401, 1020, 879, 712	OH (3600-3450); C-H (2950-2850); C=O (1650-1600); CO_3^{2-} (1500-1350); C-O (1080-950); $CaCO_3$ deformation (900-850); Lattice vibrations (720-700)
sawdust	3304, 2900, 1595, 1508, 1420, 1134, 1031	OH (3400-3200); C–H (CH_2/CH_3 stretching) (2900 – 2850); C=C (1630-1580); CH_3 (1410-1360); Si-O (1100-1000)
glass	2907, 2810, 780-1100	Si-O-Si (780 - 1100); C-H (2900-2700)
fly ash BC	1417, 995, 874, 774, 595	CO_3^{2-} (1500-1350); Si-O (1250-1150); Al-O (1100-900); Fe-O / Ca-O (600-500)
bottom ash, fly ash, slag, fly ash BCFBB	approx. 1022, 876	Si-O (1100-950); CO_3^{2-} (900-850)
sand from fluidized beds	1081, 778	Si-O-Si (1100-1000); Si-O bending (800-750)

adhesive performance, particularly through their reactivity with isocyanates.

Carbonyl groups (C=O), present in lignin and eggshells, may also affect PU adhesive properties. Peaks in the 1700–1600 cm^{-1} range, corresponding to C=O vibrations, indicate their presence. While these groups do not directly react with isocyanates, they can increase stiffness, potentially increasing hardness while reducing flexibility, which limits their use in applications requiring flexibility. Their overall effect depends on their relative abundance compared with other functional groups . C–H groups, found in sawdust and biochar, also affect interactions with isocyanates. In sawdust, a band near 2900 cm^{-1} corresponds to aliphatic $–CH_2–$ and $–CH_3$ stretching vibrations, characteristic of cellulose and hemicellulose. Peaks in the 2800–3000 cm^{-1} range contribute to strengthening the adhesive structure through additional crosslinking, but excessive amounts can lead to increased brittleness. C=C groups present in biochar and lignin, indicated by bands at 1620–1570 cm^{-1} may also react with isocyanates, introducing additional crosslinks. This improves the structure's integrity but can reduce flexibility and increase stiffness. Water, identified in bamboo fibers, hemp fibers, textile fibers, and sawdust (bands in the 3200–3600 cm^{-1} range), can negatively affect polyurethane adhesives by participating in hydrolysis reactions and altering the material's chemical structure. This can slow polymerization and reduce adhesive strength and durability. Therefore, controlling the moisture content of raw materials is essential to achieving the desired mechanical properties.

In summary, the presence of –OH, C=O, and C–H functional groups, along with moisture content, significantly influences the properties of polyurethane adhesives. Hydroxyl groups improve bonding with isocyanates but may lead to over-crosslinking. Carbonyl groups increase hardness but reduce flexibility. In particular, water present in textile fibers may promote material degradation and reduce adhesive quality. The absence of distinct C–C peaks in the FTIR spectra of organic fibers, biochar, and lignin is likely due to overlapping bands in the 1000–1400 cm^{-1} region, which commonly includes combined signals from C–C, C–O, and C–H vibrations, making it difficult to assign specific bands. Bamboo, hemp, and textile fibers exhibited a greater number of absorption bands than other materials, reflecting their complex lignocellulosic composition, which includes cellulose, hemicellulose, lignin, and, in the case of textile fibers, possible protein residues. Characteristic features include strong C–H stretching near 2900 cm^{-1}, CH_2/CH_3 bending near 1450 cm^{-1}, and C–O or C–O–C stretching in the 1300–1000 cm^{-1} region. Out-of-plane C–H bending associated with aromatic structures consistently appears around 550–560 cm^{-1}. No significant absorption bands were observed in the 1900–2500 cm^{-1} spectral region, which is typically inactive for both organic and inorganic waste materials. Any weak signals observed in this region are likely due to baseline fluctuations or atmospheric CO_2.

Elution of pollutants from waste

A detailed leachability analysis was conducted to evaluate the potential environmental impact of different types of waste. This analysis aimed to determine the levels of heavy metals and other potentially harmful elements present in the waste materials. Leachability analysis is a key component of environmental risk assessment as it allows the identification and quantification of potentially toxic substances that may be released from waste materials into the environment.

The results of leachability tests for pollutants in the waste materials, presented in Table 4, show that most of the tested samples meet the limits specified in the Regulation of the Minister of Economy (Journal of Laws of the Republic of Poland, 2015, item 1277). However, some materials did not meet the legal requirements. Textile fibers exceeded the permissible limit for antimony (Sb). According to the regulation, the maximum allowable leaching value for Sb is 0.7 mg/kg dry matter. The measured value in textile fibers was 0.72 mg/kg dry matter. This slight exceedance may result from the use of antimony-based flame retardants, which are commonly applied in synthetic fabrics. The elevated level may limit further use of this material unless proper treatment is applied. Risk management strategies should also be considered. Glass from photovoltaic panels met the legal requirements; however, the measured Sb value of 0.69 mg/kg of dry matter was close to the limit. Although compliant, this result indicates a narrow safety margin. Further monitoring is recommended to ensure continued safety.

Excess zinc (Zn) was found in lignin and hemp fibers. In both cases, the values exceeded the 2 mg/kg dry matter threshold. This may be due to contamination from chemical processing or zinc-based additives in lignin. Hemp fibers may also accumulate heavy metals from fertilized or polluted soil. Like Sb, the use of these materials in polyurethane adhesives may require either purification or careful raw material selection .Other trace elements (Pb, Cr, As, Cd, Se, Hg, Mo, Cu, Ba, Ni) may originate from various sources, including environmental contamination (soil or water), industrial additives (e.g., dyes, preservatives), or secondary pollution during storage or transport. Although most values were within limits, their presence highlights the need for quality control and monitoring of waste-based materials intended for industrial use.

In summary, trace metals found near or above legal limits confirm the importance of raw material selection, pre-treatment, and continuous monitoring when using waste materials as fillers in polyurethane adhesive formulations.

Additionally, other parameters were analyzed (Table 5), such as DOC, TDS, sulfates, fluorides, and chlorides. For DOC, the permissible limit is 800 mg/kg dry matter. Values exceeding this limit were observed in the following materials: sawdust (7,500 mg/kg dry matter), bamboo fibers (6,400 mg/kg dry matter), biochar (1,900 mg/kg dry matter), lignin (461 482 mg/kg dry matter), and hemp fibers (8,500 mg/kg dry matter). For TDS, the permissible limit is 60,000 mg/kg dry matter. Materials exceeding this limit include lignin (17,200 mg/kg dry matter) and fly ash (74,100 mg/kg dry matter). For sulfates, the permissible limit is 2,000 mg/kg dry matter. The following materials exceeded this limit: fly ash from biomass combustion in a fluidized bed (4,230 mg/kg dry matter), fly ash (26,800 mg/kg dry matter), fly ash BC (12,300 mg/kg dry matter), and lignin (17,200 mg/kg dry matter). Fluoride concantrations in all tested materials were within the permissible limits. However, for chlorides, the permissible limit is 15,000 mg/kg dry matter, and lignin exceeded this limit, with a value of 23,000 mg/kg dry matter.

In conclusion, most of the waste materials tested show low pollutant leaching and meet the criteria set out in the

Table 4. The results of the leachability of metals from waste materials [mg/kg s.m.].

Parameter	**Zn**	**Cu**	**As**	**Ba**	**Pb**	**Ni**	**Mo**	**Cr**	**Cd**	**Sb**	**Se**	**Hg**
Lignin	**10.00**	0.65	<0.025	0.64	<0.1	<0.04	<0.04	0.57	<0.005	<0.025	<0.025	<0.01
biochar	0.76	<0.04	<0.025	2.40	<0.1	<0.04	<0.04	<0.03	<0.005	<0.025	<0.025	<0.01
bamboo fibers	1.10	<0.04	<0.025	4.10	<0.1	<0.04	<0.04	<0.03	<0.005	<0.025	<0.025	<0.01
hemp fibers	**20.00**	1.40	0.090	16.00	<0.1	<0.04	<0.04	<0.03	0.110	<0.025	0.069	<0.01
textile fibers	**13.00**	1.70	<0.025	12.00	<0.1	0.49	1.20	0.83	0.120	**0.720**	<0.025	<0.01
eggshell	0.65	<0.04	<0.025	1.20	<0.1	<0.04	<0.04	<0.03	<0.005	<0.025	<0.025	<0.01
sawdust	5.00	< 0.04	< 0.025	2.50	<0.1	<0.04	<0.04	<0.03	<0.005	< 0.025	<0.025	<0.01
glass	0.67	<0.04	<0.025	1.20	<0.1	<0.04	<0.04	<0.03	<0.005	0.690	<0.025	<0.01
fly ash BC	<0.05	<0.04	<0.025	3.30	<0.1	<0.04	0.92	2.10	<0.005	<0.025	0.270	<0.01
bottom ash	<0.05	<0.04	<0.025	1.20	<0.1	<0.04	<0.04	<0.03	<0.005	<0.025	<0.025	<0.01
fly ash	4.00	<0.04	<0.025	3.40	<0.1	<0.04	1.30	1.60	<0.005	<0.025	<0.025	<0.01
slag	<0.05	<0.04	<0.025	1.60	<0.1	<0.04	<0.04	<0.03	<0.005	0.035	<0.025	<0.01
fly ash BCFBB	< 0.05	<0.04	<0.025	7.60	<0.1	<0.04	2.60	1.20	<0.005	0.069	0.120	<0.01
sand	0.99	<0.04	<0.025	4.20	<0.1	<0.04	<0.04	<0.03	<0.005	<0.025	<0.025	<0.01
*acceptable limit	≤5.00	≤50.00	≤2.000	≤100	≤10	≤10	≤10	≤10	≤1	≤0.7	≤0.5	≤0.2
* The acceptable threshold values were adopted based on the Regulation of the Minister of Economy of 16 July 2015 on the acceptance of waste at landfills for inert waste (Journal of Laws 2015, item 1277)												

Minister for the Economy's regulation. However, some materials, such as sawdust, sand from fluidized beds, bamboo fibers, biochar, lignin, and hemp fibers, require further studies and possible pretreatment to minimize their potential environmental impact.

Evaluation of Selected Waste Fillers in the Formulation of 2C Polyurethane Adhesives

Based on the results of physicochemical characterization, FTIR analysis, and pollutant leachability, three waste materials – fly ash, fly ash from fluidized bed combustion (BCFBB), and photovoltaic panel glass – were identified as the most promising candidates for application in two-component polyurethane adhesives.

According to the selection criteria, the following three materials were selected for preliminary synthesis trials to assess their compatibility with the polyurethane system. These materials fulfilled the key requirements, including low moisture content, environmentally safe leaching behavior, and favorable particle size distribution.

Particle size was a critical factor in the selection process. Materials with fine particles ensure better homogenization within the adhesive matrix and eliminate the need for additional grinding. Ideally, fillers should exhibit a particle size distribution comparable to commonly used industrial materials such as kaolin or ground calcium carbonate (GCC). Kaolin typically has a median particle size (D50) of 1–10 µm, while GCC falls within D50 range of 5–20 µm, with most particles smaller than 63 µm (passing a 250 mesh sieve). Such fillers disperse well in polyurethane systems and contribute to a homogeneous structure.

In contrast, coarser waste materials tend to agglomerate and require additional processing, such as grinding in colloid mills, which increases both complexity and cost. Therefore, priority was given to materials in which no more than 70% of the mass was retained on a 0.25 mm sieve. To assess the suitability of the selected waste materials as fillers for polyurethane adhesives, three syntheses were performed, with each waste component making up 50% of the mixture by weight. The formulation containing photovoltaic panel glass proved unsuitable. As shown in Figure 4, the filler did not integrate with the adhesive matrix and settled at the bottom. Moreover, despite prior grinding, the glass particles retained sharp edges, which caused discomfort and difficulty during manual dosing in the laboratory. This characteristic could also pose challenges in industrial settings, potentially affecting operator safety and handling comfort. The fluidized bed fly ash also exhibited poor compatibility. Despite increased mixing speed, the material formed agglomerates, preventing homogeneous dispersion (Figure 5).

In contrast, conventional fly ash (from pulverized coal combustion) showed good dispersibility, stable integration, and homogeneous structure formation during synthesis. Therefore, it was selected for further adhesion testing.

To evaluate the effect of filler type on adhesive properties, two separate syntheses of polyurethane adhesive were conducted – one using fly ash as the sole filler and the other using chalk. Additionally, formulations containing various proportions of both fillers were prepared.

The research results indicate that chalk used as the sole filler provides the highest stability and the best overall properties, especially at low temperatures (20°C - 4.76). However, as the temperature increases, these values decrease significantly, reaching 0.80 at 100°C. In comparison, polyurethane adhesives containing 100% fly ash exhibit much lower values, decreasing from 2.03 at 20°C to just 0.10 at 100°C, indicating weaker

Table 5. The results of pollutant elution rate (mg/kg) from waste materials [mg/kg s.m.].

Parameter	DOC	TDS	Sulfates	Fluorides	Chlorides
Units	mg/kg s.m.				
lignin	**461 482**	**877 900**	17 200	35	**23 000**
biochar	**1 900**	32 500	<100	<1.0	1 750
bamboo fibers	**6 400**	11 000	<100	<1.0	80
hemp fibers	**8 500**	12 700	1 110	<1.0	397
textile fibers	<20	7 820	1 360	<1.0	423
eggshell	<20	6 060	<100	<1.0	254
sawdust	**7 500**	9 860	< 100	< 1.0	54
glass	66	880	<100	19	<50
fly ash BC	35	33 400	**12 300**	1.7	6 390
bottom ash	< 20	< 5 700	**3 630**	< 1.0	116
fly ash	150	54 100	**26 800**	52	4 190
slag	<20	3 920	1 800	3	319
fly ash BCFBB	690	8 690	**4 230**	20	80
sand	46	14 400	750	17	99
*Acceptable limit	800	60 000	2 000	150	1500

* The acceptable threshold values were adopted based on the Regulation of the Minister of Economy of 16 July 2015 on the acceptance of waste at landfills for inert waste (Journal of Laws 2015, item 1277)

Fig 4. Lack of compatibility of glass with the polyurethane system

Fig 5. Appearance of component A with the addition of fly ash from fluidized bed combustion

stability and reduced effectiveness as a filler. Analysis of fly ash and chalk mixtures shows that increasing the chalk content improves material performance. A mixture containing 75% fly ash and 25% chalk yields values of 2.34 at 20°C and 0.30 at 100°C, while the 50% fly ash and 50% chalk variant reaches 2.99 at 20°C and 0.30 at 100°C. With further increases in chalk content, performance continues to improve – the 25% fly ash and 75% chalk mixture achieves 4.24 at 20°C and 0.50 at 100°C. The formulation containing 10% fly ash and 90% chalk produces values nearly identical to those of pure chalk (4.50 at 20°C and 0.78 at 100°C). These results indicate that fly ash alone is not an effective substitute for chalk. However, a small addition of fly ash (up to 10%) does not negatively affect the adhesive properties. At higher fly ash contents, noticeable reductions in performance are observed, particularly at elevated temperatures.

Discussion

The study evaluated the potential of 15 waste materials as fillers in two-component polyurethane (2C PU) adhesives, with a focus on their compatibility, mechanical properties and economic feasibility. The findings can be summarized in the following key points:

1. Fly ash as a promising filler.

The present study demonstrated that fly ash exhibited the best compatibility with the polyurethane system among the analyzed waste materials, forming a uniform and stable mixture without visible sedimentation or phase separation. Similar effects have been reported by other authors. Kuźnia et al. (2022, 2024) observed that polyol mixed with 5–10 wt% fly ash produced a homogeneous blend prior to isocyanate addition, indicating good miscibility and the absence of dispersion issues during mechanical mixing. These findings confirm favorable interactions between fly ash particles and the polyurethane matrix, resulting in stable mixtures.

Magiera et al. (2025) further reported that fly ash, as a by-product of coal combustion, enhances the mechanical strength and thermal stability of polyurethane systems, indirectly confirming good interfacial compatibility. Li et al. (2025) showed that chemical modification of fly ash with epoxy-functional silane agents significantly improved interfacial bonding and reduced agglomeration in polyurethane composites. The modified filler achieved approximately 45% higher mechanical strength and 64.5% greater interfacial adhesion compared to the unmodified one, confirming the strong affinity between fly ash and the polyurethane matrix.

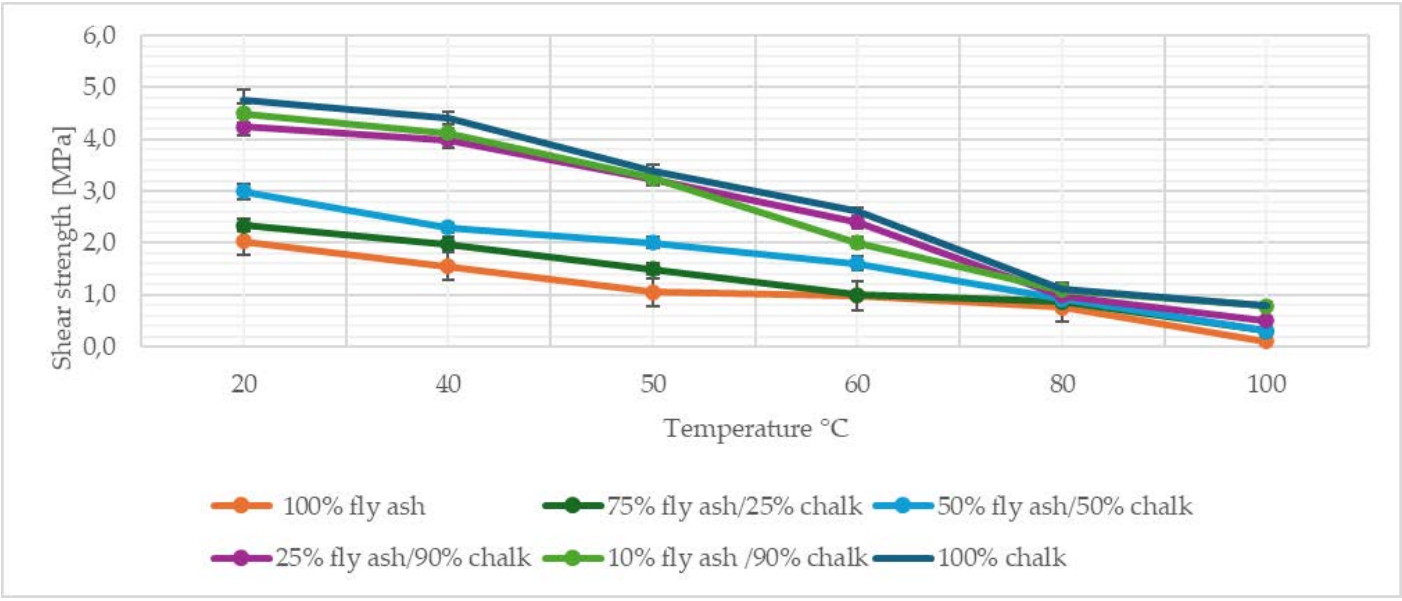

Fig 6. Change in shear strength as a function of temperature for various polyurethane adhesive formulations containing fly ash and chalk. Results represent mean values based on 10 measure-ments; error bars indicate standard deviation.

Collectively, these studies indicate that fly ash can be effectively incorporated into polyurethane systems, forming homogeneous and process-stable mixtures, consistent with the observations of the present study.

Having established the good compatibility of fly ash with the polyurethane system, its influence on the adhesive's mechanical performance was further evaluated. Shear strength tests revealed that fly ash can partially replace conventional fillers such as chalk. The highest strength values were obtained for formulations containing 10% fly ash and 90% chalk (4.50 MPa) and 25% fly ash and 75% chalk (4.24 MPa), demonstrating that the fly ash-to-chalk ratio strongly affects adhesive performance. In contrast, the formulation containing 100% fly ash exhibited much lower strength—2.03 MPa at 20 °C and only 0.10 MPa at 100 °C—indicating limited thermal stability and weak reinforcement at elevated temperatures.

The decrease in strength with increasing temperature is consistent with literature data on polyurethanes (Wirpsza, 1991), which indicate that these materials maintain stable physical properties between 20–60 °C, while at 100 °C their strength decreases by about 50%. This reduction results from the lower viscosity of the soft segments and a decrease in the stress required for plastic deformation within the domain structure. Therefore, the decline in shear strength observed for the 100% fly ash formulation corresponds to the expected thermal behavior of polyurethane adhesives and is further intensified by poor dispersion and limited interfacial adhesion at high filler loadings.

Relatively few studies have addressed the use of fly ash in polyurethane adhesives, particularly in two-component (2K PU) systems. One of the few examples is the work of Pęczek et al. (2025), who confirmed that a moderate addition of fly ash improves viscosity, uniformity, and mechanical strength of 2K polyurethane adhesives, whereas excessive filler loading leads to particle agglomeration and reduced cohesion.

More extensive research has been conducted on other polyurethane materials such as foams, composites, and elastomers. Zhai et al. (2024) found that uniformly dispersed fly ash enhances cohesion and structural integrity in polyurethane composites, while exceeding the optimal filler content results in microdefects and reduced strength. Kuźnia et al. (2024) reported that silanized fly ash improves interfacial adhesion and compressive strength in polyurethane foams. Similarly, Özer and Bektaşoğlu (2025) observed that evenly distributed fly ash cenospheres increased both compressive strength and thermal insulation properties of foams without compromising structural stability.

Dağ (2025) identified fly ash as the most compatible industrial waste filler for polyurethane matrices, forming strong interfacial bonds and maintaining good dispersion. Wang et al. (2025) likewise reported that fly ash filled microvoids between polymer chains in PU/water-glass composites, producing a compact structure that enhanced load transfer and reduced microcrack formation.

Taken together, both literature data and the results of the present study confirm that a moderate amount of fly ash improves cohesion and mechanical strength in polyurethane systems, whereas excessive content leads to agglomeration and deterioration of performance. These findings clearly demonstrate that fly ash can successfully replace part of conventional mineral fillers in two-component polyurethane adhesives, provided that its content is properly optimized and uniform dispersion within the matrix is ensured.

2. Challenges with other waste materials.

Materials such as glass from photovoltaic panels and ash from fluidized bed combustion were found to be unsuitable for polyurethane systems due to poor integration, manifested by particle settling and agglomeration. Similar issues with glass-derived waste have been reported in other material systems. Máčalová et al. (2021) observed that incorporating glass from end-of-life photovoltaic panels into cement composites required careful control of particle size and surface treatment to ensure uniform dispersion and adhesion within the matrix. Likewise, Smoleń et al. (2024) reported that waste glass from solar thermal collectors exhibited limited compatibility in polymer concretes, leading to reduced cohesion and local inhomogeneities when used without modification. Nevertheless, photovoltaic glass waste remains a valuable secondary raw material. When finely ground or surface-treated, it can partially replace silica sand or cement in cementitious or polymer-based composites, improving mechanical strength, chemical stability, and sustainability (Máčalová et al., 2021; Smoleń et al., 2024).

For fly ash derived from fluidized bed combustion, Kuźnia et al. (2022) compared different ash types and found that unmodified particles showed poor dispersion and interfacial adhesion in polyurethane matrices, whereas silane modification significantly improved filler distribution and mechanical performance. These findings confirm that both photovoltaic glass waste and fluidized bed combustion ash require surface treatment or coupling agents to achieve satisfactory compatibility with polymer systems.

Due to their high moisture content, materials such as lignin, sawdust, and textile fibers were excluded from further testing, as excessive water interferes with polyurethane crosslinking. According to Peres et al. (2023), elevated moisture levels in polyol components or fillers reduce the reactivity of hydroxyl groups, lowering isocyanate conversion efficiency and consequently deteriorating adhesive performance. Similarly, Lisý et al. (2022) and Jiju et al. (2025) observed that the intrinsic polarity and moisture absorption capacity of lignin hinder its compatibility with hydrophobic polyurethane matrices unless chemical modification is applied. Nevertheless, lignin remains a promising renewable raw material in other chemical sectors; its aromatic structure enables its use in bio-based resins, adhesives, and biodegradable plastics (Jiju et al., 2025).

Sawdust was also excluded from polyurethane formulations due to its high moisture content and poor compatibility with hydrophobic matrices. However, it can be effectively reused in the construction industry as a partial replacement for sand or cement in lightweight composites, thereby improving sustainability and reducing environmental impact. Olaiya et al. (2023) demonstrated that incorporating sawdust into cementitious materials enhances thermal insulation and reduces density while maintaining adequate mechanical strength, making it a promising component for sustainable building materials.

Eggshells and fluidized bed sand were disqualified due to inadequate granulation, which hindered uniform dispersion and interfacial bonding in the polyurethane matrix. Nevertheless, eggshells, rich in calcium carbonate, can be valorized in other sectors as natural additives or fillers. Babalola et al. (2024) reported that waste eggshell powder serves as an efficient and eco-friendly source of calcium for agricultural fertilizers and soil conditioners. Similarly, Chen et al. (2022) showed that calcined eggshells can act as precursors for calcium-based sorbents used in environmental purification and gas adsorption.

Sulfur was excluded from polyurethane adhesive formulations due to poor compatibility with organic matrices and the potential emission of toxic gases (SO_2 and SO_3) during thermal degradation. Its presence in polymer systems may cause undesirable reactions at processing temperatures, leading to discoloration, odor release, and reduced thermal stability. Fediuk et al. (2020) noted that elemental sulfur oxidizes at elevated temperatures, forming sulfur oxides that pose both technological and environmental hazards, thus limiting its direct use in polymer composites.

Nevertheless, sulfur remains a valuable industrial material that can be effectively used in circular-economy applications. Studies have demonstrated that it can serve as a binder in sulfur-based concretes and polymer composites, providing excellent chemical resistance, water impermeability, and durability under aggressive environmental conditions (Gutarowska et al., 2019; Fediuk et al., 2020). Such materials are recommended for sewage systems, acid-resistant structures, and infrastructures exposed to chemical corrosion. Furthermore, recent advances in polymer chemistry have enabled the development of "inverse vulcanized sulfur polymers," which utilize elemental sulfur as a primary component in cross-linked polymer networks. These materials exhibit high thermal and chemical stability and are currently being explored for energy storage, mercury capture, and self-healing applications (Diniz et al., 2025). Such innovations demonstrate that while sulfur is unsuitable as a direct filler in polyurethane adhesives, it retains significant potential as a sustainable raw material for next-generation functional composites.

3. Economic and environmental considerations.

The incorporation of waste-derived fillers, particularly fly ash, proved to be a cost-effective approach to material formulation, as it reduces dependence on virgin resources and supports circular economy principles. The results of this study indicate that such by-products can contribute to lowering production costs and improving the environmental profile of polyurethane-based materials. This interpretation is supported by previous studies, in which similar benefits were observed for various composite systems (Alhazmi et al., 2021; Mort, 2022; Garbacz, 2013).

However, the need for preprocessing — including drying, grinding, and impurity removal — may incur additional energy and labor costs, partially offsetting the initial material cost savings. Comparable conclusions were drawn by Vinod Kumar et al. (2018) and Formela et al. (2022), who emphasized that the cost-effectiveness of waste-derived fillers strongly depends on the scale and efficiency of the processing methods employed. Therefore, a comprehensive industrial-scale cost analysis remains essential to fully assess the economic feasibility of such materials, as financial constraints remain a major barrier to large-scale recycling applications (Shehab et al., 2023; Formela et al., 2022).

4. Correlation between preliminary analyses and material selection.

FTIR and leachability analyses played a crucial role in the preliminary screening process, despite not all materials being tested for adhesive performance. These methods enabled the identification of fillers exhibiting excessive chemical reactivity or potential environmental hazards, allowing the early elimination of unsuitable materials prior to mechanical testing. While this study focuses on polyurethane-based systems, such a multistage evaluation—combining chemical characterization and environmental screening—is consistent with approaches widely adopted in other material technologies. Numerous studies emphasize that, irrespective of the material base, assessing chemical compatibility and environmental safety is an essential step in the design and qualification of composites containing waste-derived fillers. Bordoloi et al. (2024) highlighted the need to confirm the chemical stability and reduced metal leachability of modified fly ash before its use in polyolefin-based composites. Goh et al. (2017) reported that similar evaluations are indispensable for epoxy composites containing municipal solid waste incineration ashes, whereas Alghamdi (2021) stressed the importance of verifying filler–matrix compatibility in fly ash-filled HDPE systems.

Collectively, these findings demonstrate that, irrespective of the matrix type, whether polyurethane, epoxy, polyolefin, or cement-based, the preliminary evaluation of chemical reactivity and environmental safety constitutes a universal criterion for filler selection, ensuring both composite stability and compliance with sustainable development principles.

5. Future research directions.

- Adhesion and Cohesion Optimization: Conducting detailed adhesion tests on various substrates (e.g., metals, plastics, and wood) under different environmental conditions (temperature and humidity) to evaluate the performance of fly ash-based adhesives in real-world applications.
- Long-Term Durability: Assessing the aging behavior of the adhesive under simulated long-term operational conditions to ensure its reliability over time.
- Sustainability and Circular Economy Impact: Assessing the potential of waste-derived fillers to support circular economy principles by reducing the consumption of virgin raw materials, minimizing waste generation, and lowering CO_2 emissions. This approach not only enhances resource efficiency but also offers economic benefits by reducing dependency on conventional, often more expensive, fillers.

By addressing these research directions, the potential of fly ash and other waste materials as sustainable fillers in PU adhesives can be fully realized, contributing to the development of eco-friendly and high-performance adhesive systems.

Summary

The findings of this study support the concept of replacing conventional mineral fillers in polyurethane adhesives with selected industrial and organic waste materials. Among the tested candidates, fly ash derived from pulverized coal combustion proved to be the most compatible with the two-

component polyurethane (2C PU) system. Formulations with partial chalk replacement (up to 30%) achieved satisfactory mechanical performance under standard conditions.

While the results are promising, the study was limited to a single application type (wood bonding), a fixed filler content (50 wt%), and short-term performance testing. Therefore, further research is needed to verify the feasibility of these materials in real production environments and across broader application domains. Future studies should focus on scaling up adhesive production for pilot or industrial trials, assessing cost-efficiency and processability at larger scale, and evaluating long-term durability under combined moisture, thermal, and mechanical loading. By identifying sustainable filler alternatives, this work contributes to reducing dependency on virgin raw materials and supports the development of adhesives aligned with circular economy principles.

Appendix 1

Note: A 100% Passing value at the coarsest sieve indicates that the entire sample passed through that mesh, with no particles retained above it. This does not imply the presence of zero-sized particles, but rather that all particles are smaller than the mesh size of the largest sieve used in the analysis.

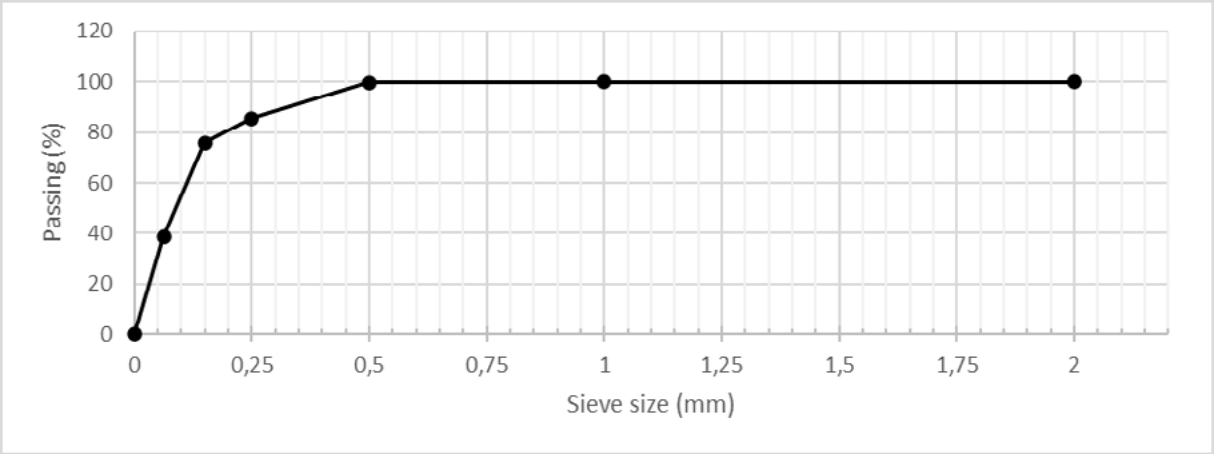

Chart 1. Particle size distribution curves for Lignin.

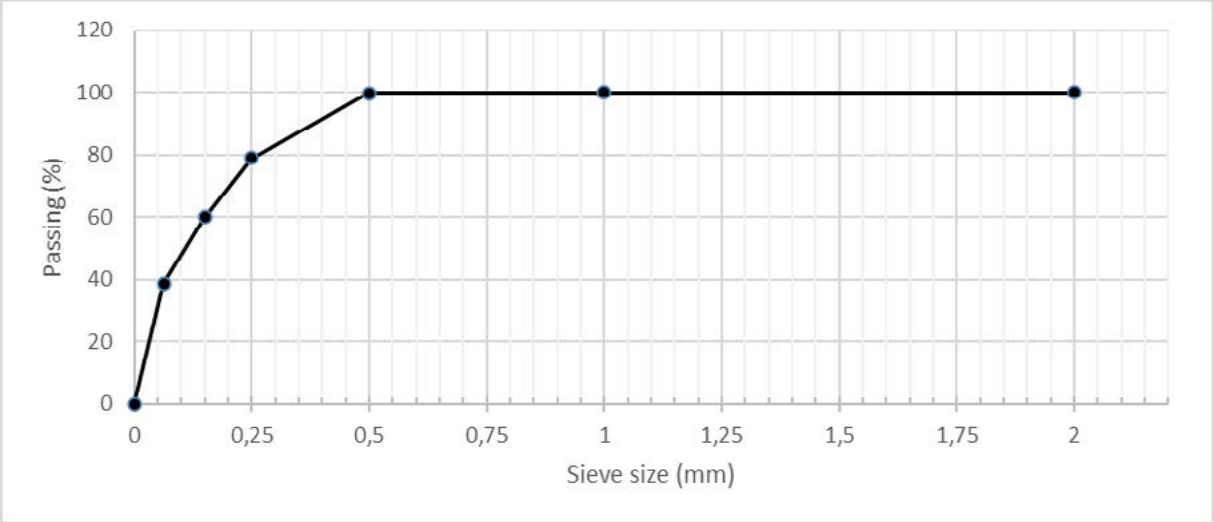

Chart 2. Particle size distribution curves for Biochar.

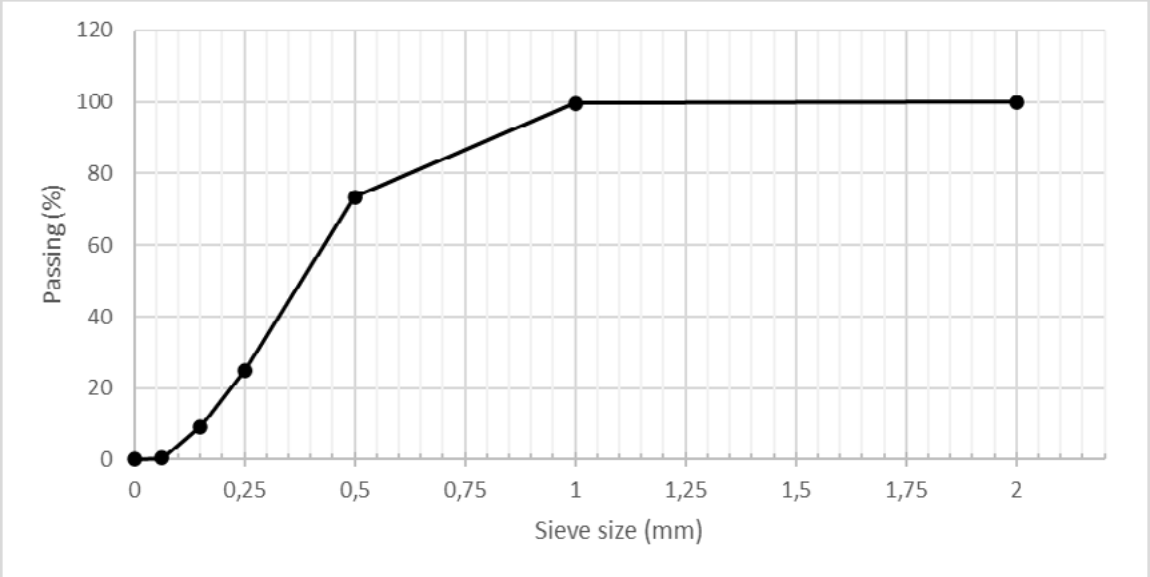

Chart 3. Particle size distribution curves for Bamboo fibers.

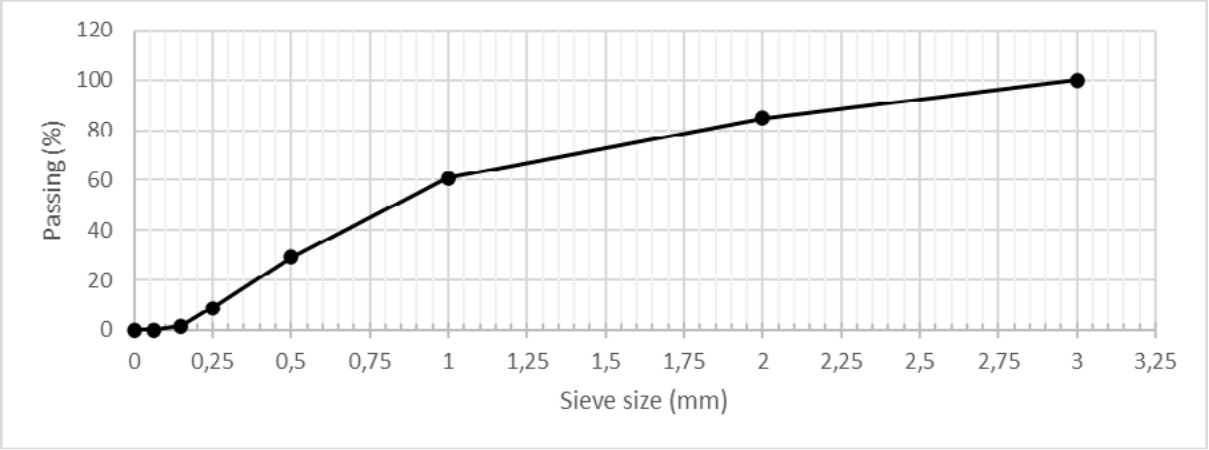

Chart 4. Particle size distribution curves for Hemp fibers.

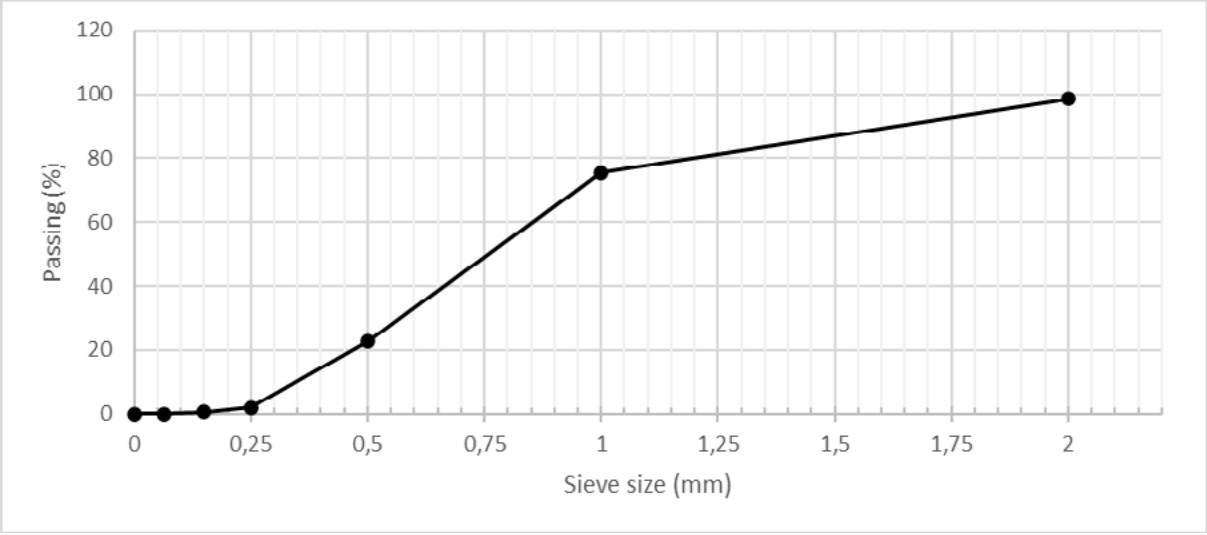

Chart 5. Particle size distribution curves for Sawdust.

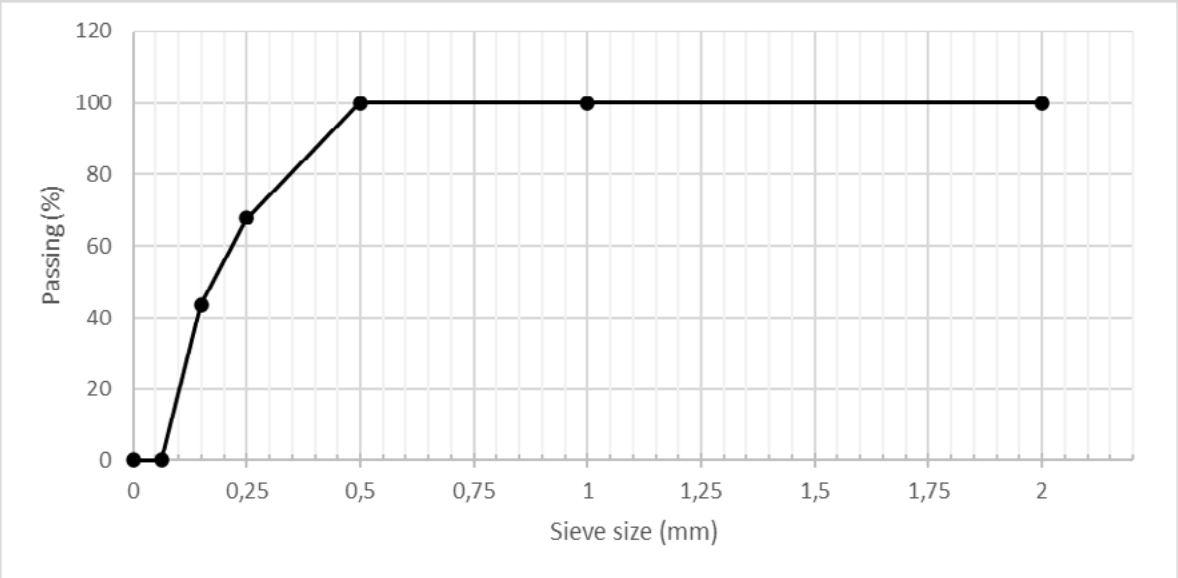

Chart 6. Particle s size distribution curves for Sulfur.

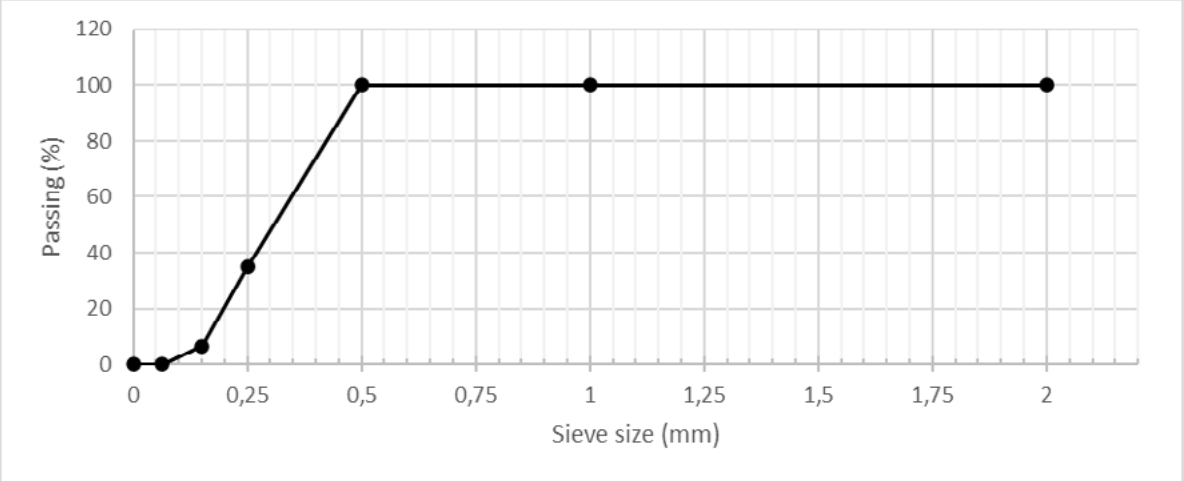

Chart 7. Particle size distribution curves for Glass.

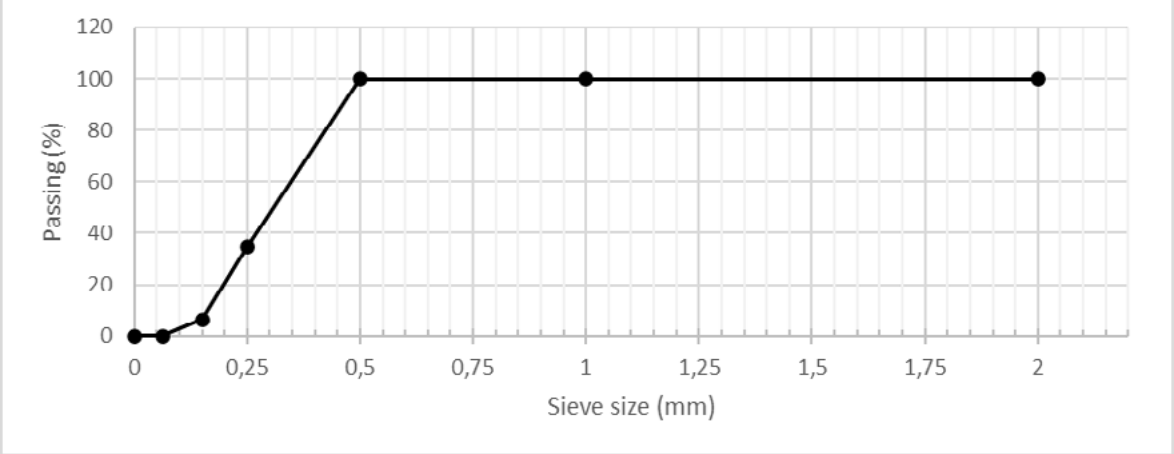

Chart 8. Particle size distribution curves for Fly ash BC.

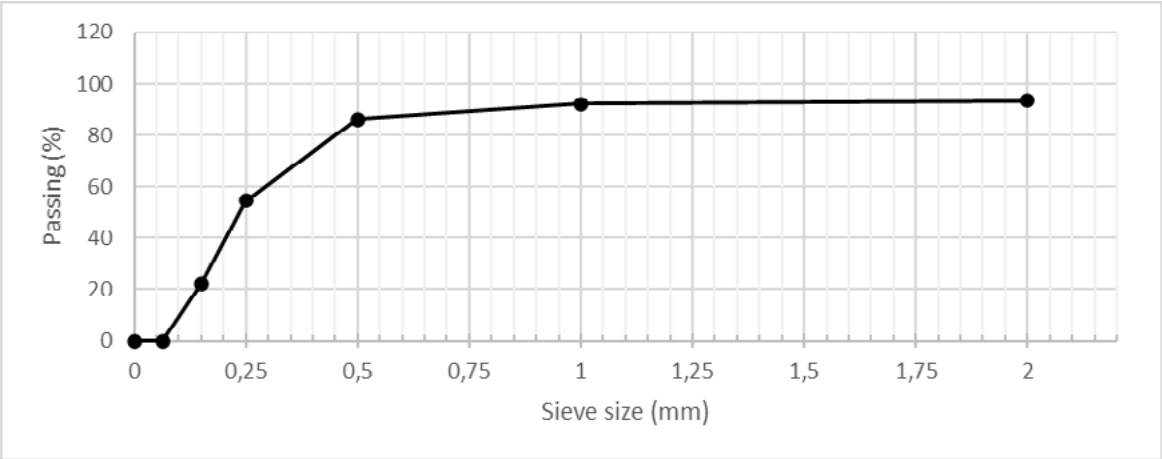

Chart 9. Particle size distribution curves for Bottom ash.

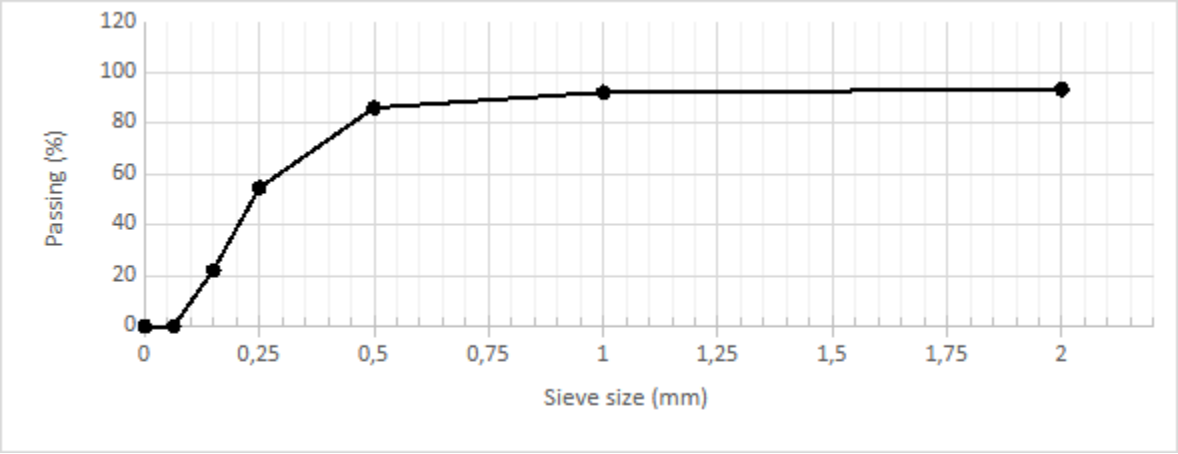

Chart 10. Particle size distribution curves for Fly ash.

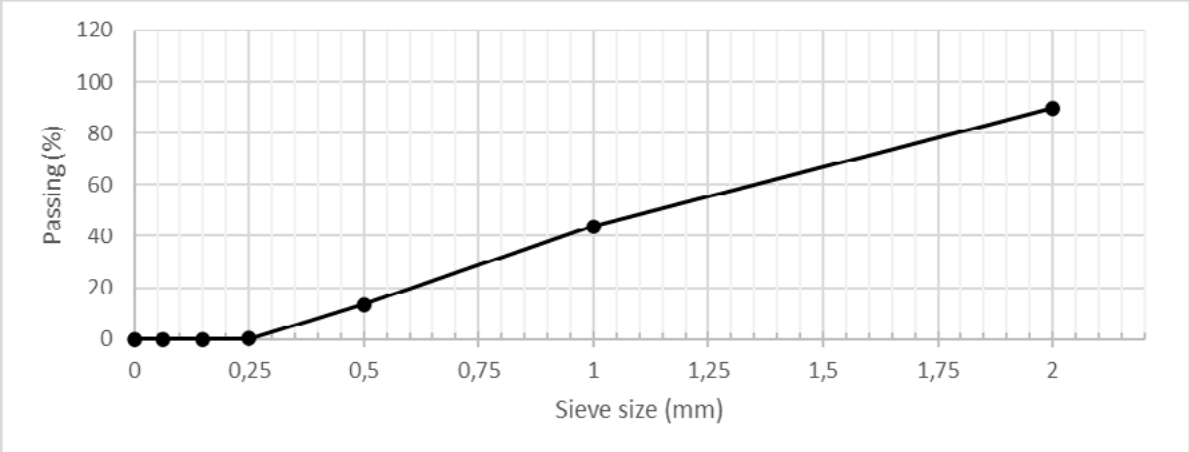

Chart 11. Particle size distribution curves for Slag.

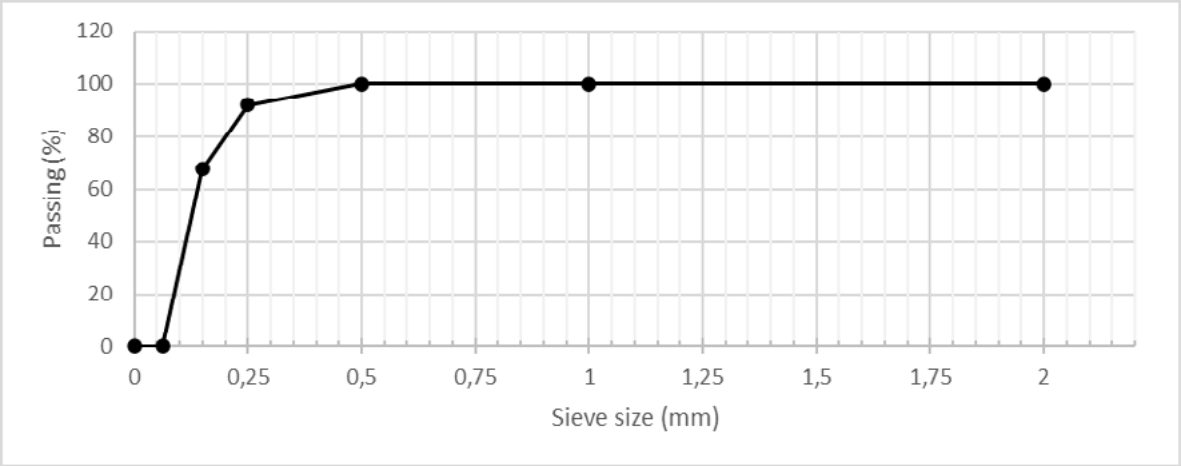

Chart 12. Particle size distribution curves for Fly ash BCFBB.

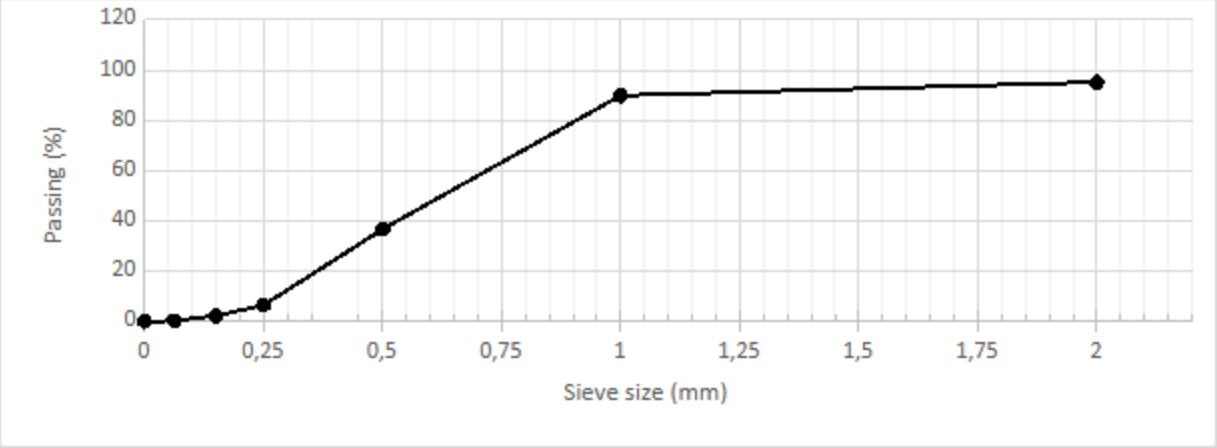

Chart 13. Particle size distribution curves for and from fluidized beds.

Appendix 2

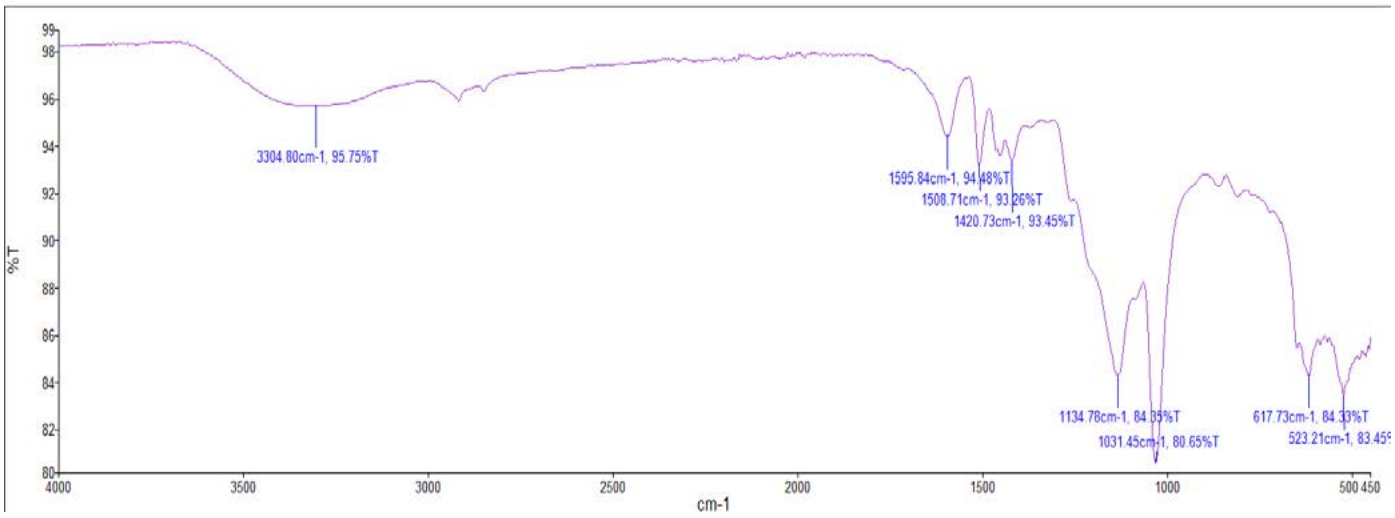

Fig. 1. ATR-FTIR Spectrum of lignin.

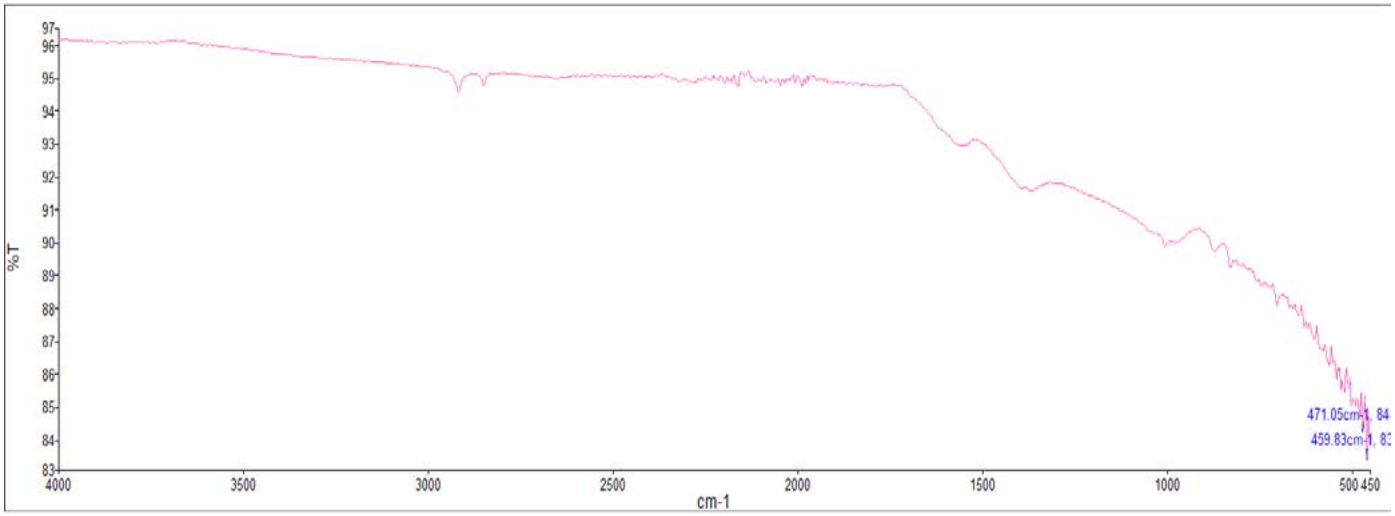

Fig. 2. ATR-FTIR Spectrum of biochar.

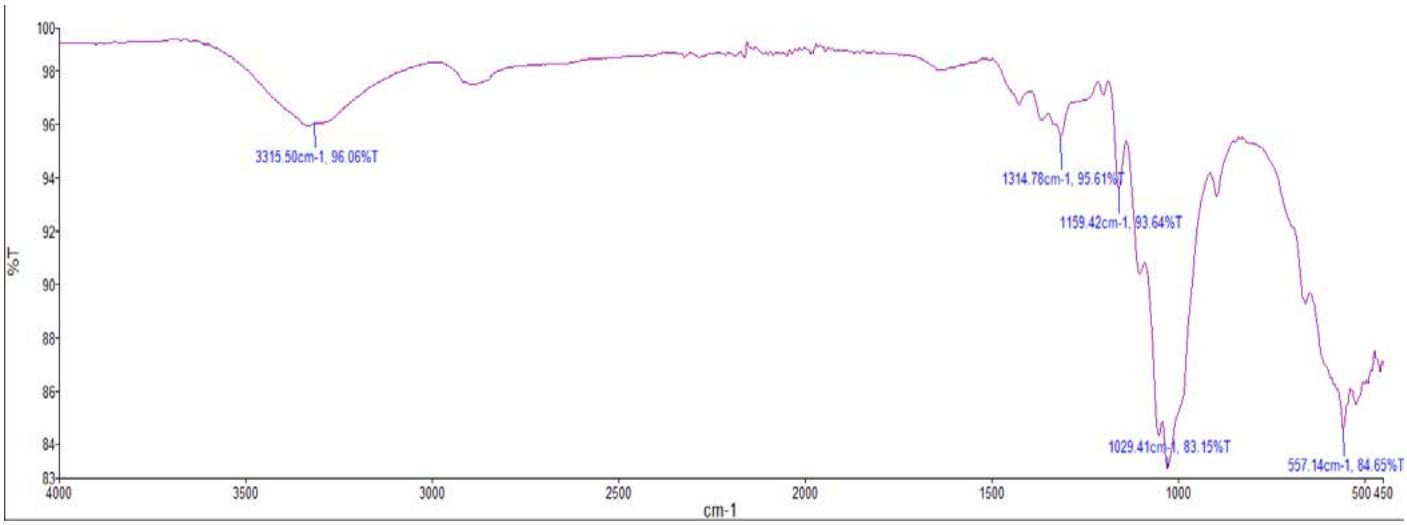

Fig. 3. ATR-FTIR Spectrum of bamboo fibers.

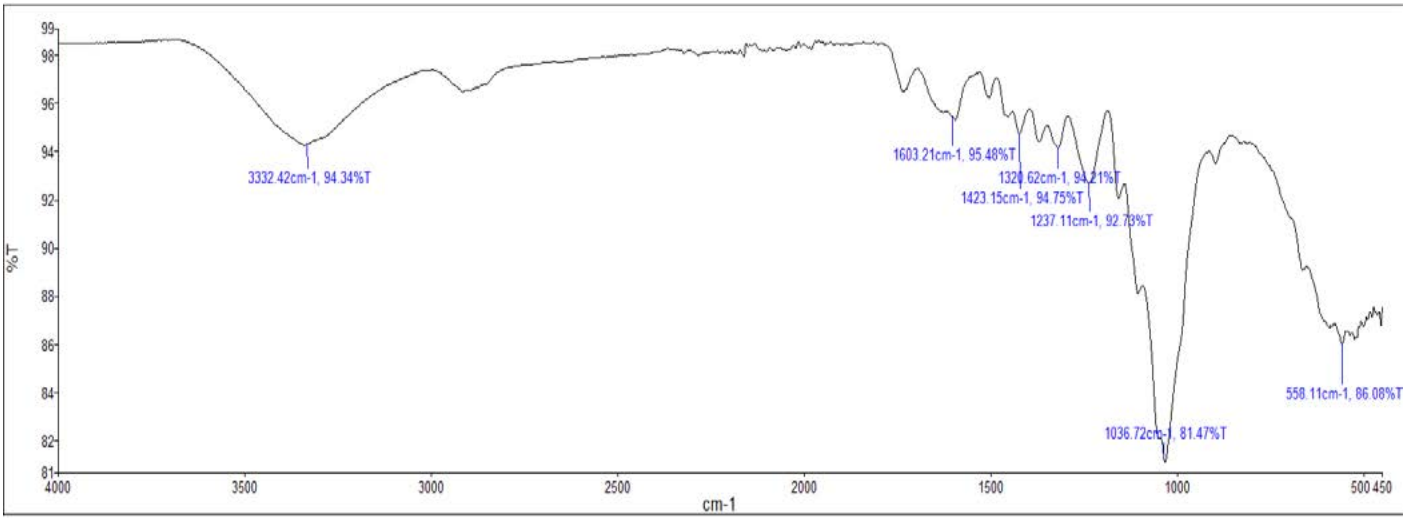

Fig. 4. ATR-FTIR Spectrum of hemp fibers.

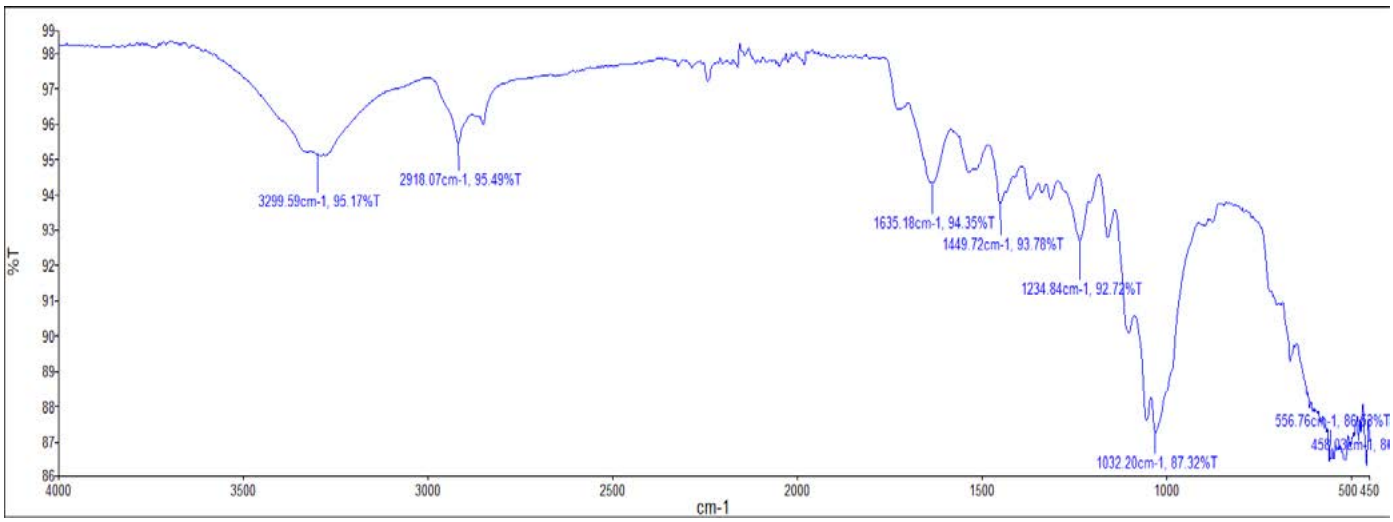

Fig. 5. ATR-FTIR Spectrum of textile fibers.

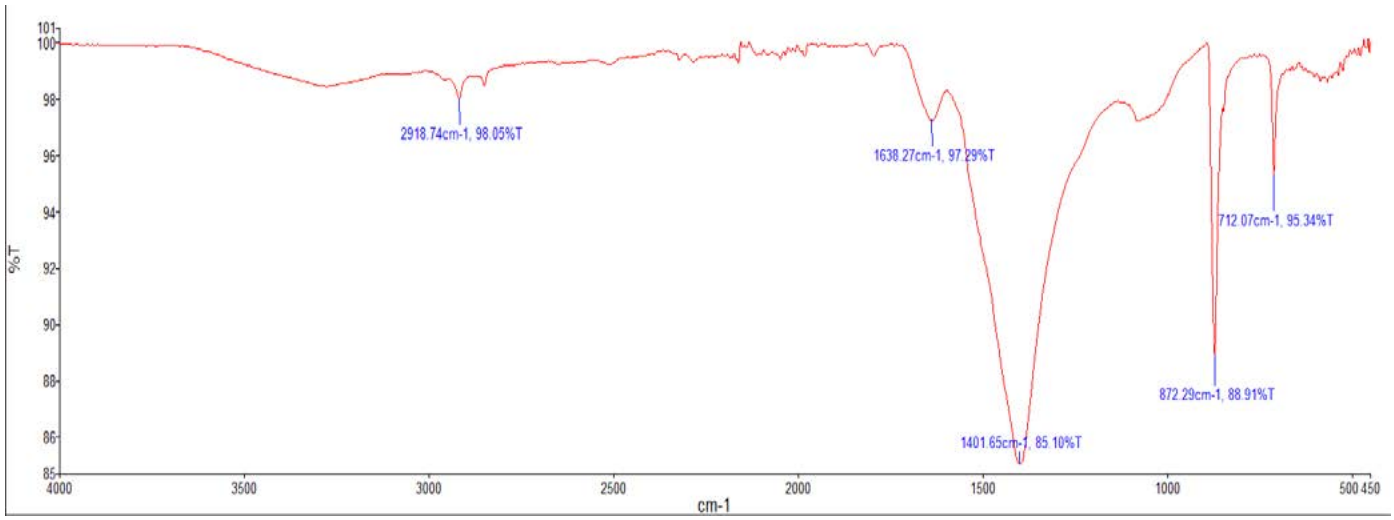

Fig. 6. ATR-FTIR Spectrum of eggshells.

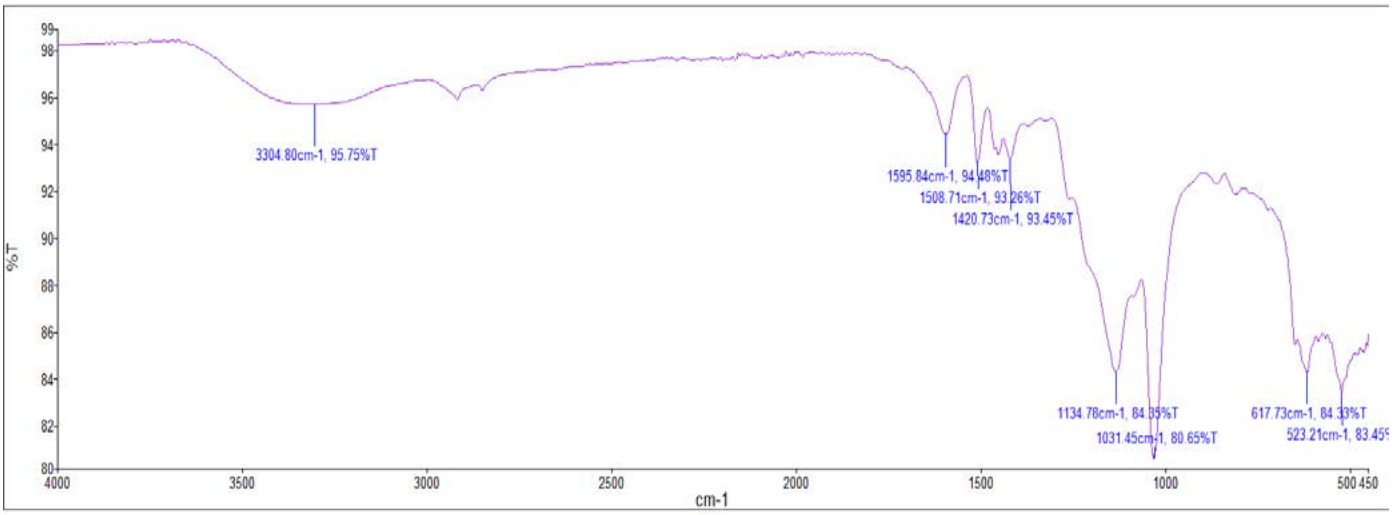

Fig. 7. ATR-FTIR Spectrum of sawdust.

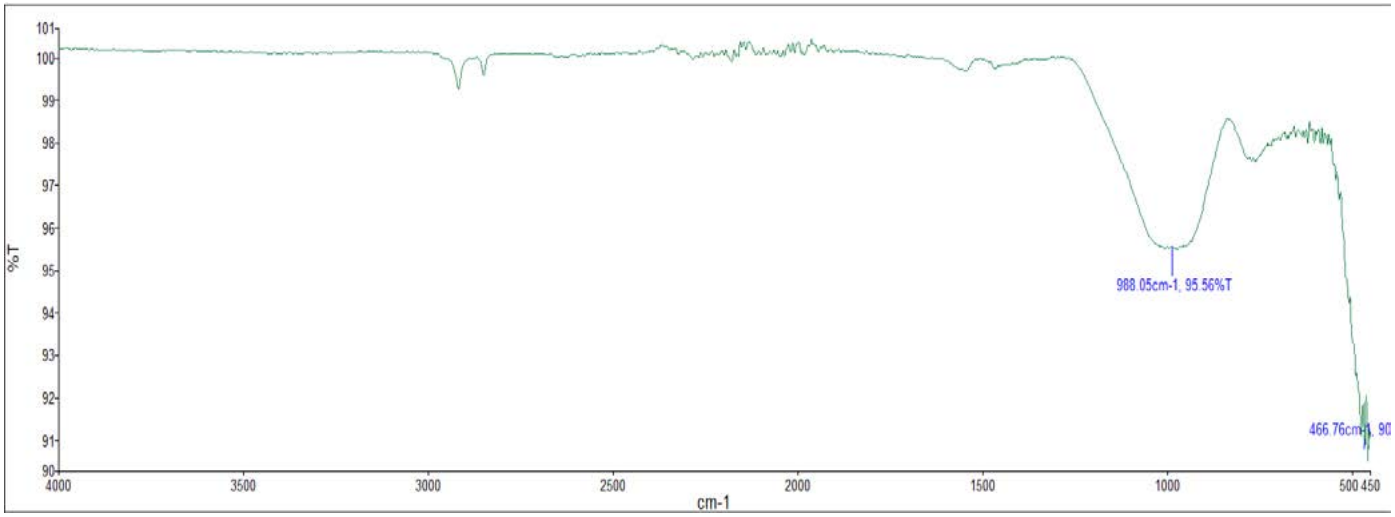

Fig. 8. ATR-FTIR Spectrum of glass from photovoltaic panels.

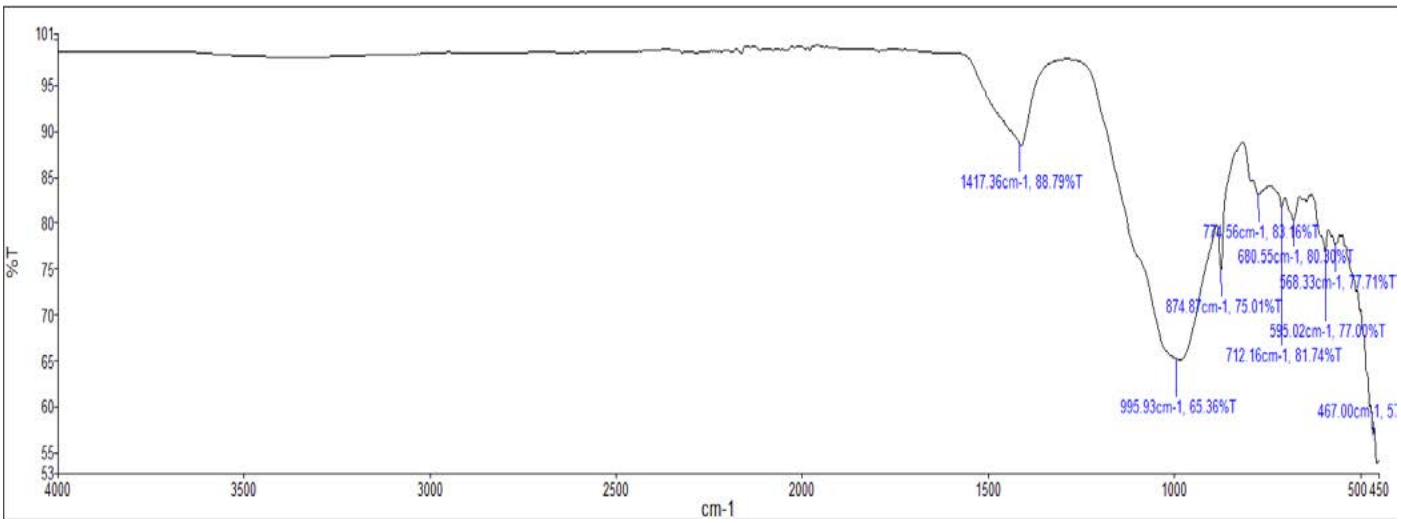

Fig. 9. ATR-FTIR Spectrum of fly ash from biomass combustion.

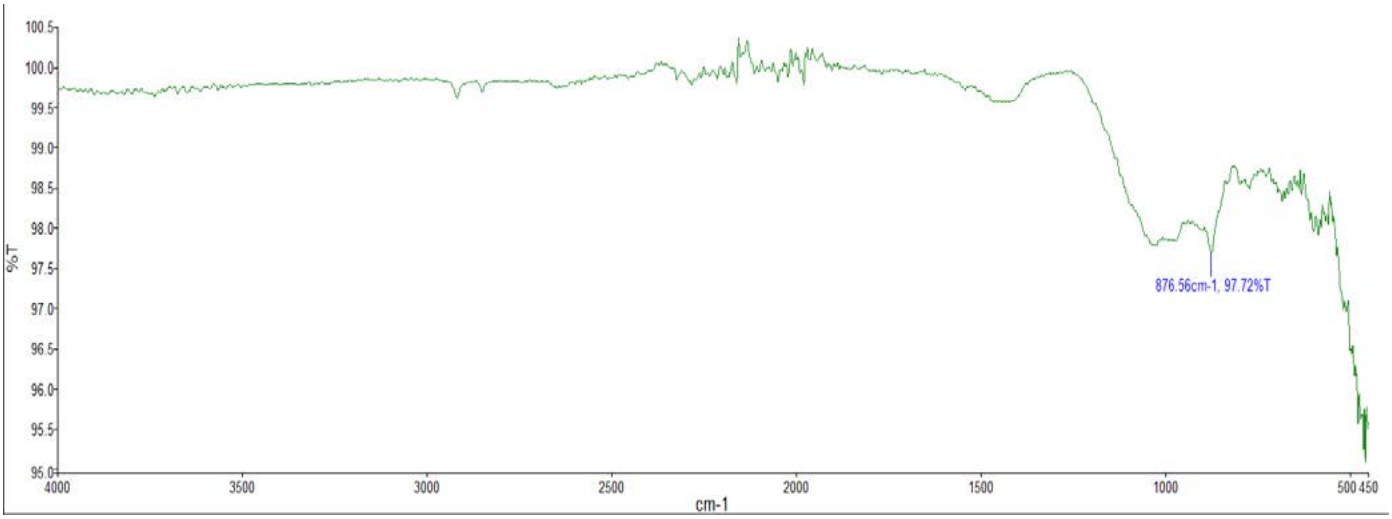

Fig. 10. ATR-FTIR Spectrum of bottom ash.

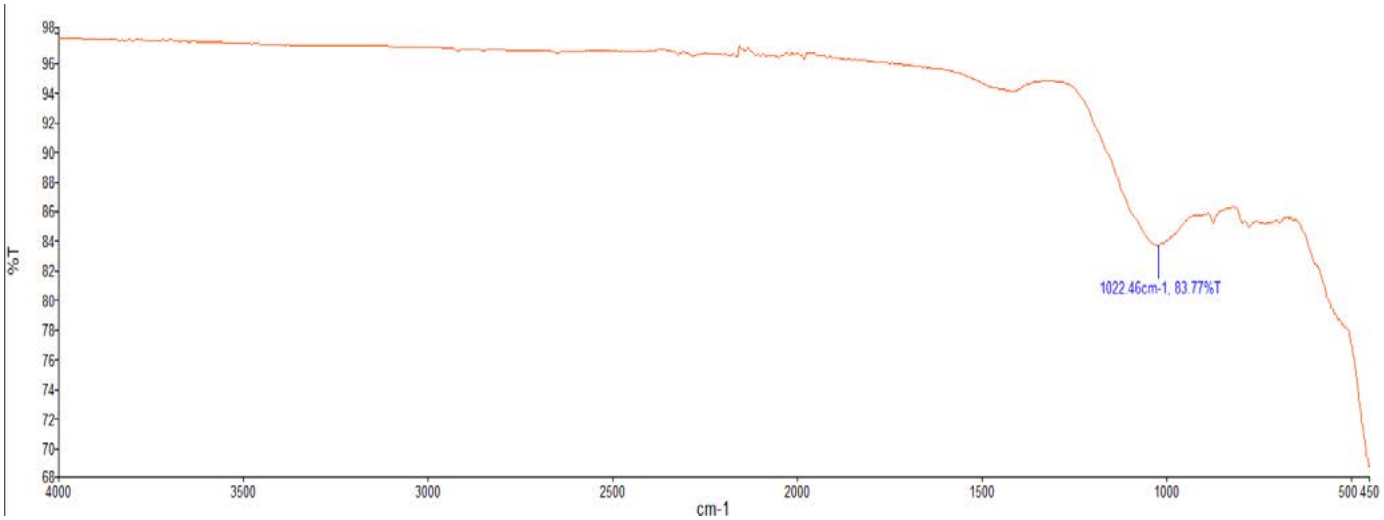

Fig. 11. ATR-FTIR Spectrum of fly ash.

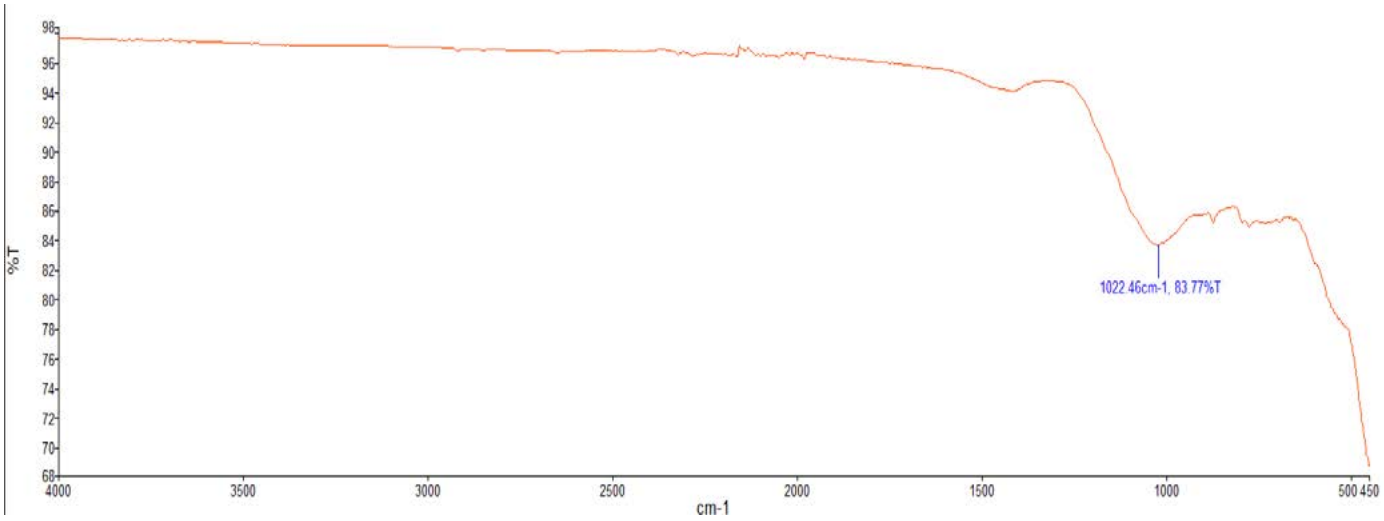

Fig. 12. ATR-FTIR Spectrum of slag.

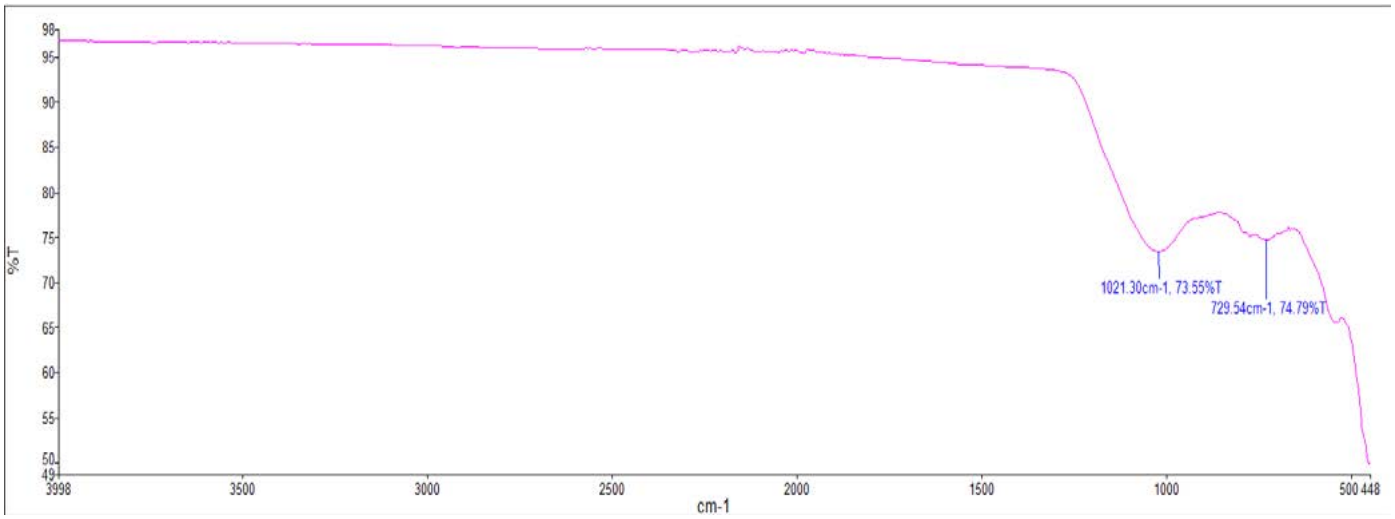

Fig. 13. ATR-FTIR Spectrum of Fly ash from biomass combustion in a fluidized bed boiler.

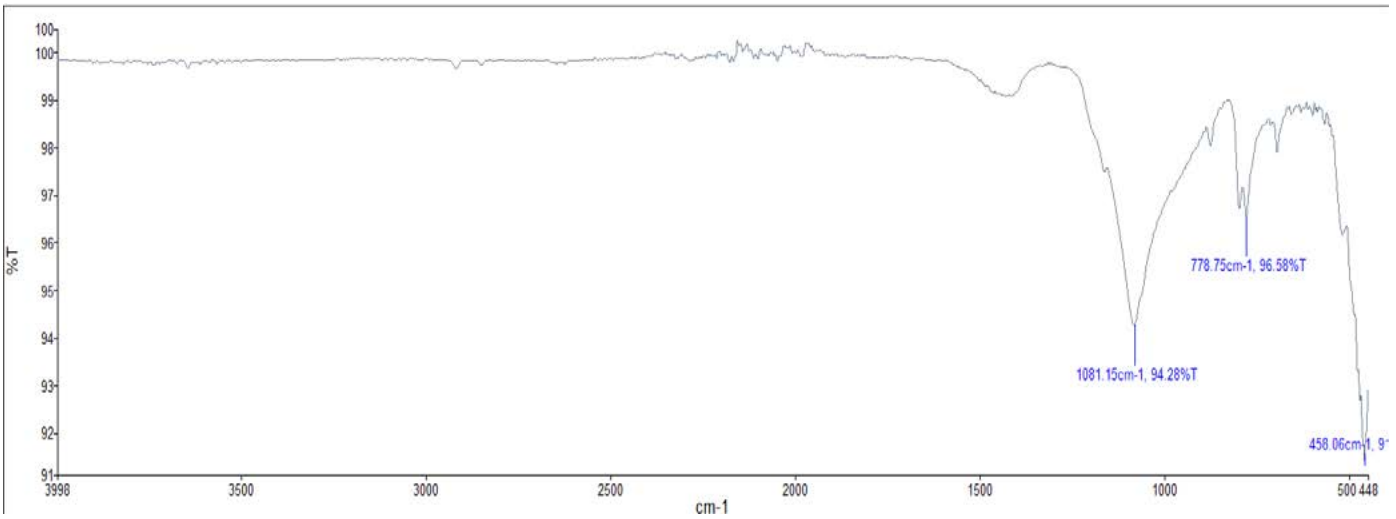

Fig. 14. ATR-FTIR Spectrum of sand from fluidized beds.

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Ocena odpadów przemysłowych i organicznych pod kątem ich przydatności w klejach poliuretanowych.

Streszczenie. W niniejszej pracy przeprowadzono ocenę potencjału zastosowania 15 rodzajów odpadów przemysłowych i organicznych jako wypełniaczy w dwuskładnikowych klejach poliuretanowych (2K PU). Do badania wybrano m.in. popiół lotny, popiół denny, popiół z kotłów fluidalnych, żużel, szkło z paneli fotowoltaicznych, siarkę, ligninę, biowęgiel, włókna tekstylne, włókna konopne, trociny, skorupki jaj, włókna bambusowe oraz piasek fluidalny. Materiały zostały poddane charakterystyce fizykochemicznej, obejmującej oznaczenie zawartości wilgoci, części organicznych, części palnych i popiołu, ciepła spalania, analizę sitową, określenie zawartości węgla, wodoru, azotu i siarki, a także analizę widm w podczerwieni metodą ATR-FTIR oraz ocenę podatności na wymywanie metali ciężkich. Przygotowano formulacje klejów o różnych proporcjach popiołu lotnego do kredy i poddano je badaniom wytrzymałości na ścinanie. Najwyższą wartość (4,50 MPa w temperaturze 20 °C) uzyskano dla układu zawierającego 10% popiołu lotnego i 90% kredy, co wskazuje na korzystny efekt synergiczny. Dla porównania, formulacja zawierająca wyłącznie popiół lotny (100%) wykazała znaczny spadek wytrzymałości w podwyższonej temperaturze (0,10 MPa przy 100 °C), co potwierdza jej ograniczoną stabilność termiczną. Uzyskane wyniki wskazują, że popiół lotny może stanowić zrównoważony i ekonomicznie opłacalny częściowy wypełniacz w klejach poliuretanowych, wpisujący się w założenia gospodarki o obiegu zamkniętym. Należy jednak uwzględnić jego ograniczoną stabilność termiczną przy projektowaniu formulacji przeznaczonych do stosowania w podwyższonych temperaturach.