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Manufacture of wood fiber reinforced polyvinyl acetate rigid foams

Odun lifi takviyeli polivinil asetat rijit köpüklerin üretimi

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Abstract

In this work, rigid foams designed and manufactured using the freeze drying technique were made from polyvinyl acetate (PVAc), bleached kraft pulp and unbleached kraft pulp. The rigid foams designed as an environmentally-friendly product with no pentane or hydrochlorofluorocarbon included in the manufacturing process. The PVAc based foams were reinforced with different kraft pulp contents. Their performance properties such as compressive and flexural strength, physical and morphological properties were investigated according to relevant standards. The foam densities ranged from 0,017 g/cm^3 with %17,65 coefficient of variation (CV) to 0,137 g/cm^3 with %2,33 CV. The compression resistance was found between 0,001 N/mm^2 with %50,00 CV and 0,03 N/mm^2 with %5,98 CV. The flexural resistance was found between 0,005 N/mm^2 with %20,00 CV and 0,11 N/mm^2 with %6,06 CV. Optimum properties were observed at B-4 (PVAc/ Bleached Kraft pulp 1/0.8). Bleached kraft pulp reinforcement gave better results on performance characteristics of foam materials compared to unbleached kraft pulp reinforcement. Overall test results showed that the PVAc based rigid foams have promising results.

Keywords: Rigid foam, freeze-drying technique, polyvinyl acetate, kraft pulp, mechanical properties

Öz

Bu çalışmada, dondur-kurut tekniği kullanılarak tasarlanmış ve üretilmiş rijit köpükler polivinil asetattan (PVAc), ağartılmış kraft hamurundan ve ağartılmamış kraft hamurundan yapılmıştır. Rijit köpük çevre dostu bir ürün olarak üretim aşamasında pentan veya hidrokloroflorokarbon içermemektedir. PVAc bazlı köpük farklı oranlarda kraft hamuru ile güçlendirilmiştir. Basınç, eğilme kuvvetleri, fiziksel ve morfolojik özellikleri gibi performans özellikleri ilgili standartlara göre incelenmiştir. Köpük yoğunlukları %17,65 varyasyon katsayısı (CV) ile 0,018 g/cm^3 ve %2,33 CV ile 0,137 g/cm^3 arasında değişmektedir. Basınç direnci %50,00 CV ile 0,001 N/mm^2 ve %5,98 CV ile 0.03 N/mm^2 arasında değişmektedir. Eğilme direnci ise %20,00 CV ile 0,005 N/mm^2 ve %6,06 CV ile 0,11 N/mm^2 arasında bulunmuştur. Optimum özellikler B-4'ten (PVAc/Ağartılmış Kraft hamuru 1/0.8) elde edilmiştir. Ağartılmış kraft hamur takviyesi, ağartılmamış kraft hamur takviyesine kıyasla köpük malzemenin performans özellikleri üzerinde daha iyi sonuçlar vermiştir. Tüm test sonuçlarına göre PVAc bazlı rijit köpüğün umut verici sonuçlar sergilediği gözlenmiştir.

Anahtar Kelimeler: Rijit köpük, dondur-kurut tekniği, polivinil asetat, kraft hamuru, mekanik özellikler

1. Introduction

The low prices and reliable properties including but not limited to lightweight, good sound absorption and high thermal insulation made the foam materials attractive as an everyday item. Therefore, a dramatic rise occurred in the foam material sector. Researchers focused on investigating the novel application possibilities, unique characteristics, procedures and manufacturing techniques (Lee and Ramesh, 2004). Currently, there are many foaming techniques such as extrusion, (Guan and Hanna, 2004) freeze-drying, (Glenn and Irving, 1995) microwave heating, (Sjöqvist and Gatenholm, 2005) wafer baking, (Soykeabkaew et al., 2004; Glenn et al., 2001) and compression/expansion (Glenn and Orts, 2001).

One of the most widely used rigid foam types is expanded polystyrene foams (EPS). Polystyrene is a solid material at room temperature that can be molded at the high temperatures and re-solidified for required fields. The possible expansion of polystyrene is about forty times of the original volume. EPS foam has been used in many applications like thermal insulation board in buildings, packaging, cushioning of valuable goods and food packaging. However, EPS foam has some drawbacks such as lack of resistance to organic solvents, being hazardous to environment, flammability, and releasing hazardous gases under burning conditions (Thoughtco, 2019).

Nowadays, the majority of commercial foam products are manufactured from polystyrene and polyurethane. However, these polymers are petroleum-based and require a long time to decompose in nature. Therefore, many studies have been focused on the production techniques of biodegradable foams (Ahmadzadeh et al., 2015). First, the foam production process from paper fibers started in the early 1960s and pulp fibers were produced by high shear power by dispersing them with water/foam-forming material (Radvan, 1964). Today, the foam material is produced from many different natural polymers by various methods. Most of the studies were conducted by making the polymers at high temperatures and then producing foam with the help of various gases. In Soykeabkoew's study, (Soykeabkoew et al., 2004) foams were manufactured from starch which was obtained from cassava and jute fiber. Starch and jute fiber were cross-linked with each other, and improvements in bending resistance were determined. Corn starch-based foams were produced by adding mono stearyl citrate which improved the water resistance (Shogren et al., 2002). Foams were produced with starch and latex. The latex enhanced bending resistance and water resistance (Shey et al., 2006). In another study, biodegradable foams were produced with cassava starch, sunflower proteins, and cellulose fibers through a baking process. When the amount of fiber increased from %10 to %20 in foam production, its mechanical properties increased. Optimum properties were obtained from 20% fiber and %10 sunflower mixture (Salgado et al., 2008).

The freeze-drying process includes three main parts: freezing, primary drying and secondary drying (Jennings, 1999). At first, the water molecules form ice crystals after the freezing process. Then the drying process starts under specialized pressure and temperature conditions. During the drying phase, the final pore morphology is created as ice crystals are sublimated. Therefore, the foam structure is immediately linked to the size and distribution of the frozen system (Svagan et al., 2008).

The PVAc glue has been mostly used in the adhesive sector due to its reliable cost. However, the use of PVAc has been decreasing. In this research, it was used as a matrix material. The unique freeze-drying technique was preferred for the manufacturing process. In this study, the bleached kraft pulp and unbleached kraft pulp were used to reinforce PVAc based rigid foams. The PVAc glues are easily available, inexpensive and have very few volatile organic components. Under certain conditions, PVAc was shown to biodegrade (Trejo, 1988; Crowly et al., 2005). Biodegradation is further guaranteed under the particular circumstances for biodegradation only after the expected lifetime of the product. The raw materials for PVAc are derived from fossil resources, primarily crude oil. These raw materials can be completely replaced by feedstock and switched to ethanol, which can be produced from renewable sources such as sugar cane, corn or preferably straw and other non-food parts of plants. (Amann and Minge, 2011). PVAc is a rather polar, hygroscopically restrained polymer. It is expected that the hydroxyl-rich surface of cellulose can develop strong interactions with ester links protruding abundantly from the PVAc. The resultant hydrogen bonds are expected to significantly strengthen the interface and have a positive influence on the mechanical characteristics of the materials (De Rodriguez et al., 2006).

According to The British Wood and Pulp Association, over %95 of the chemical pulp in the world is made through the kraft process, which uses caustic soda and sodium sulphate to dissolve the woodchips. There are two types of kraft pulps: unbleached kraft pulp and bleached kraft pulp. Unbleached kraft pulp is brown color due to the lignin

content found in its structure, it is generally used to manufacture corrugated boards and paper bags. The bleached kraft pulp is bleached with chemicals such as hypochlorite and peroxide. The lignin is removed through the production process so the color of the bleached pulp is whitish. Bleached kraft pulp is generally used in the tissue and pulp industry. (BWPA, 2019). As lignin and hemicellulose are removed from the fiber wall, pores are formed during the cooking process. The pores range between 5 and 15-20 nm in diameter. These pores provide the fibers with their flexibility and compressibility to create strong fiber-fiber bonds. This is going to lead to good mechanical properties. As the degree of polymerization (DP) of cellulose decreases in pulp, the individual strength of the fiber is weakened. Eventually, this phenome leads to poor mechanical properties. In addition, the viscosity value of kraft pulp is a linear relationship with zeta potential (Ek et al., 2009).

In similar studies, nanocomposite-based PVAc-reinforced cellulose nanofibril and cellulose nanocrystals have been extensively studied (De Rodriguez et al., 2006; Geng et al., 2016; Gong et al., 2011a; Gong et al., 2011b; Mathew et al, 2011). However, due to the poor distribution of cellulose nanomaterials, the development of new composite was not completed (Iwamoto et al., 2009; Iwatake et al., 2008; Roohani et al., 2008; Suryanegara et al., 2009). Hamou et al. (2018) studied biobased film production by adding PVAc glue to nanofibril cellulose. As a result of their study, the increase in were the mechanical properties of the films was observed. Kaboorani et al. (2012) added microcrystalline cellulose to the PVAc glue to investigate the effect of bonding strength. The increase in modulus of elasticity and bond strength was determined.

In this study, PVAc rigid foams reinforced with bleached and unbleached kraft pulp were manufactured. Their physical, morphological and performance properties were investigated.

2. Material and Method

In this work, the Mad Wolf brand polyvinyl acetate glue (PVAc: Super transparent framework adhesive made in Akpinar Building Materials Industry and Trade Inc./Istanbul) was purchased from a local supplier. The dry matter rate of PVAc glue was %42,8. The density of PVAc was 1,2 gr/cm^3. PVAc was reinforced with OYKA (Paper Packing Industry and Trade Inc.) brand unbleached kraft pulp and EUROPAP (Tezol Paper Industry and Trade Inc.) brand bleached kraft pulp. According to the information obtained from OYKA Paper Factory, Scotch pine is used in the cooking process in which the active alkali ratio is 125 g/lt, the sulphide rate is %27. The white solution coming from the recovery unit is used. %23 effective alkaline is used during cooking. The unbleached kraft pulp properties had a kappa number of 26,55 and a viscosity of 1155 mL/g. The ISO brightness of the unbleached pulp was determined as %27,11. The degree of polymerization is 1214. According to the information obtained from the EUROPAP tissue factory, the eastern spruce wood is used in the cooking process in which the active alkali ratio is 130 g/lt and the sulfidity rate is %30. The white solution coming from the recovery unit is used. %25 effective alkaline is used during cooking. After the cooking process, the pressurized peroxide bleaching process was applied in the bleaching unit. The temperature was maintained at 95^{0}C degrees during the process. %0.5 oxygen and %2.5 hydrogen peroxide were used in the pulp production. The bleached pulp produced had a kappa number of 3,5 and a viscosity of 650 mL/g. The ISO brightness of the bleached pulp was determined as %90. The degree of polymerization is 933. The kraft pulp was added in six (6) different ratios (PVAc / Kraft pulp (bleached and unbleached) - 1/0, 1/0.2, 1/0.4, 1/0.6, 1/0.8 and 1/1).

The 20 g. PVAc glue was used as a starting material for each suspension then dissolved with distilled water for 1 hour at 1000 RPM. Then, bleached and unbleached kraft pulps were added to the suspension depending on pre-determined ratios and shear-mixed for 15 minutes at 1500 RPM. The total suspension weight was 200 g. It was poured into molds and placed in the freezer at -25^{0}C for 24 hours. The frozen samples were freeze-dried in a lyophilizer at a condenser temperature of -50^{0}C under approx. 0,40 mBar pressures for 72 hours to manufacture the rigid foams. The manufacturing proses is demonstrated in Figure 1.

The microstructure of PVAc-Kraft pulp rigid foam specimens was investigated using JSM-7600F brand scanning electron microscopy (SEM) and pore sizes of foam were determined with Matlab software. Seven (7) test specimen were prepared for each mechanical and physical test. Prepared samples were kept in laboratory conditions for 24 hours for conditioning. The density calculations were made according to ASTM C303 (2010), the 3-point bending tests were performed according to ASTM C203 (2012) standard, and the compression tests were performed according to ASTM C165 (2007). The density, flexural and compressive strength data were compared using one-way means/ANOVA to check for significant differences (alpha= 0,01). Significant differences between the

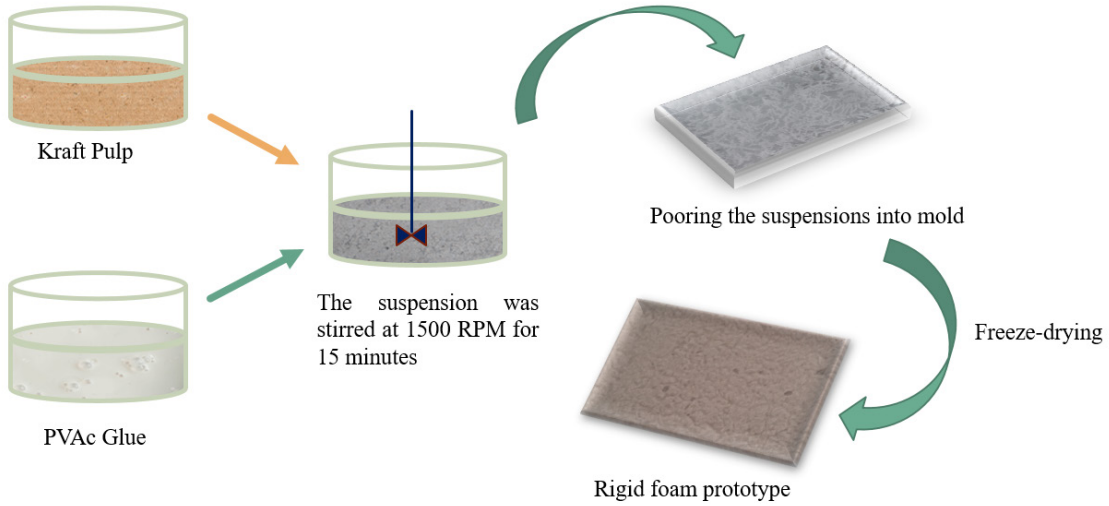

Figure 1. Manufacturing process of PVAc-Kraft pulp rigid foam.
Şekil 1. PVAc-Kraft hamuru rijit köpüğün üretim aşaması

groups were determined using Tukey-Kreamer Honest Significant Differences (HSD) test (alpha= 0,05).

3. Results and Discussion

In this study, PVAc was chosen for matrix formation because PVAc contains abundant ester bonds and forms strong bonds with the wood fiber added as a reinforcer. PVAc is degraded by *Penicillium* and *Aspergillosis fungi* (Trejo 1988). Also, PVAc-based composite material that completes its lifetime will be consumed by these fungi and its extinction time will be shortened, thus the environmental load will be reduced. On the other hand, PVAc does not cause carcinogenic effects, since it does not contain the formaldehyde content, which is dangerous for human health. So the harmless material both in terms of environment and human health will be obtained.

3.1. Density

The density of rigid foams was given in Table 1.

Table 1. The density values of manufactured rigid foams
Tablo 1. Üretilen rijit köpüklerin yoğunluk değerleri

Codes	Kraft Pulp Type	PVAc / Kraft Pulp Ratio	PVAc Glue (g)	Kraft Pulp (g)	Solvent (g)	Density (g/cm^3)
P-0	-	1/0	20	0	200	0,017 (17,65) A
U-1	Unbleached	1/0,2	20	4	200	0,047 (0,09) B
U-2	Unbleached	1/0,4	20	8	200	0,081 (0,32) C
U-3	Unbleached	1/0,6	20	12	200	0,083 (3,09) C
U-4	Unbleached	1/0,8	20	16	200	0,109 (3,05) D
U-5	Unbleached	1/1	20	20	200	0,128 (1,61) E
B-1	Bleached	1/0,2	20	4	200	0,064 (5,28) F
B-2	Bleached	1/0,4	20	8	200	0,080 (9,70) C
B-3	Bleached	1/0,6	20	12	200	0,089 (5,93) C
B-4	Bleached	1/0,8	20	16	200	0,107 (8,37) D
B-5	Bleached	1/1	20	20	200	0,137 (2,33) E

Note: Parentheses indicate the coefficient of variation (CV, %). A, B, C, D, E. F letters indicate the significant differences between the groups.

According to the test results, the density of PVAc - bleached and unbleached kraft pulp rigid foams was found changing between 0,017 g/cm^3 with a %17,65 coefficient of variation (CV) to 0,137 g/cm^3 with %2,33 CV. It was clearly shown that once the kraft pulp content increase, the density of rigid foams increases as expected (Table 1).

3.2. Morphology

The microstructure of the foams is provided at different magnifications (x25, x50 and x500) in the SEM images given in Figure 2, Figure 3 and Figure 4. The irregular pore formation with different diameters was observed.

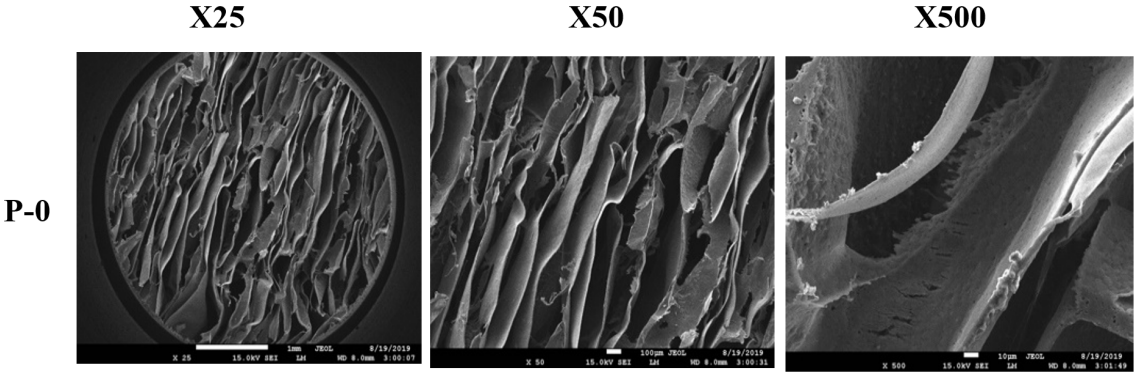

Figure 2. The SEM images of Pure PVAc rigid foam (P-0) at x25, x50 and x500 magnifications
Şekil 2. x25, x50 ve x500 büyütmede saf PVAc rijit köpüğün (P-0) SEM görüntüleri

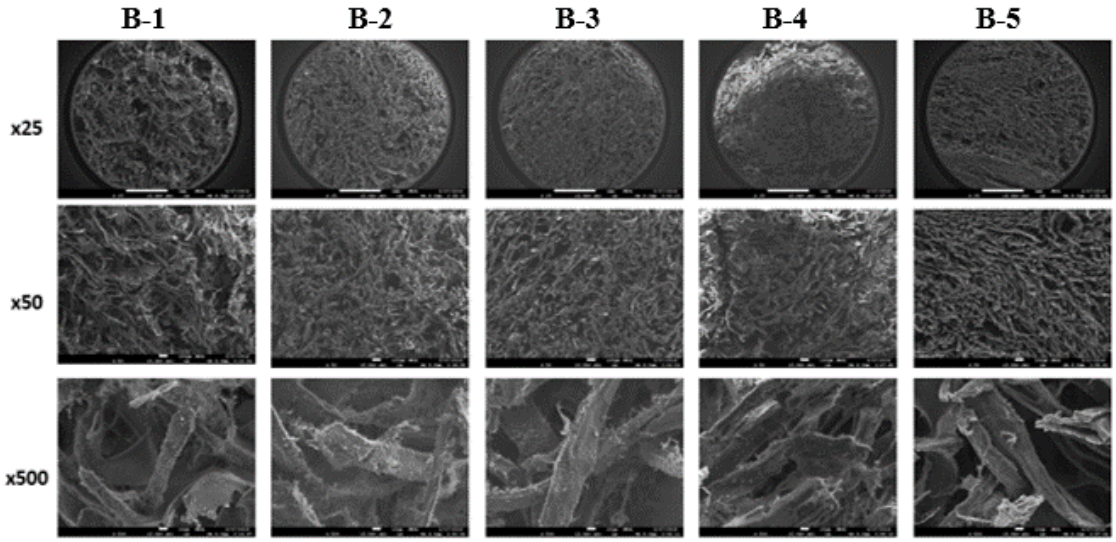

Figure 3. The SEM images of PVAc rigid foam reinforced with different rate of bleached kraft pulp at x25, x50 and x500 magnifications
Şekil 3. x25, x50 ve x500 büyütmede farklı oranlarda ağartılmış kraft hamuru takviyeli PVAc rijit köpüğün SEM görüntüleri

Pure PVAc rigid foam (P-0) was seen to have a more regular pore shape than PVAc with pulp fiber rigid foam. The amount of PVAc was kept constant and the effect of the fiber reinforcement on the foam material was investigated. The increase in fiber amount decreased the inter-pore gap. At the same time, as the amount of fiber increased, the density increased and a tighter structure was formed. Especially at x500 magnification, the adsorption of PVAc on the fiber was visible. Although the morphological structure of starch and nano-fibril cellulose foam material had regular pore shapes (Svagan et al., 2008; Dash et al., 2012), morphological structure of our production was seen to be irregular pore shapes and tight structure. The reason for this is that the individual particle size of the kraft pulp

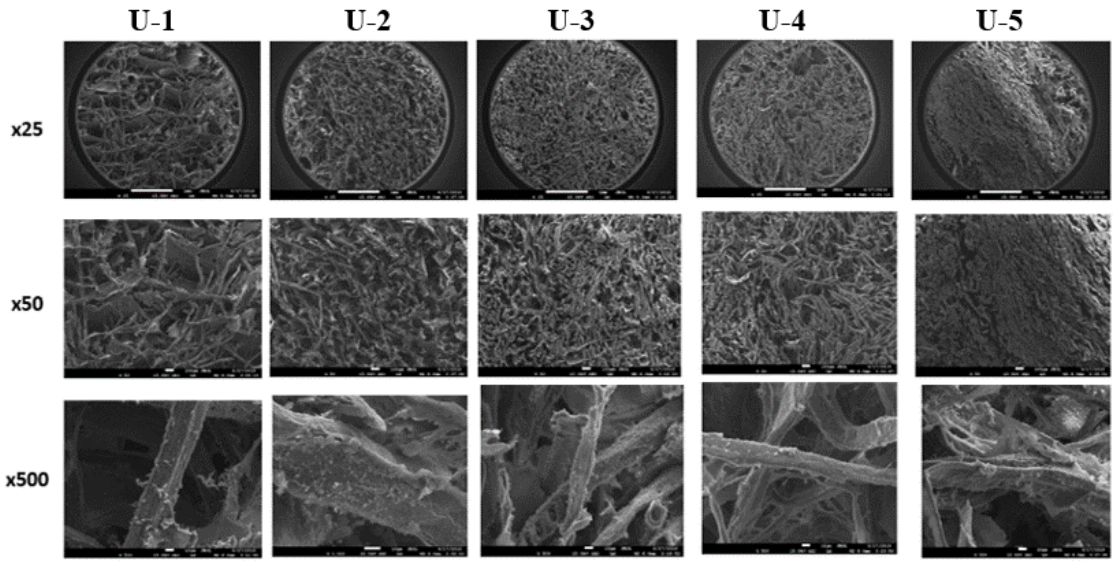

Figure 4. Representative SEM micrographs of PVAc rigid foam reinforced with different rate of unbleached kraft pulp at x25, x50 and x500 magnifications
Şekil 4. x25, x50 ve x500 büyütmede farklı oranlarda ağartılmamış kraft hamuru takviyeli PVAc rijit köpüğün SEM görüntüleri

was in the macro dimension and this macro dimension ruined structure and interpore gaps. Similar irregular and tight images were obtained on materials produced using kraft pulp (Heydarifard et al., 2017; Ottenhall et al., 2018).

As given in Table 2, the mechanical properties increased with increasing the density except for PVAc/ bleached kraft pulp ratio at 1/1. It was observed that the fibers were not disintegrated well and aggregation occurs when preparing the mixture at this ratio. The pore size determination was studied using MatLab Software. It was found that there was a great variety of pore diameters. The pore diameters ranged from several micrometers to millimeters. The main reason for this variety is thought to be related to the production process, where the sublimation process couldn't be controlled homogenously.

3.3. Mechanical properties of the rigid foams

The mechanical properties of produced rigid foams were given in Table 2.

Table 2. The mechanical properties of produced rigid foams
Tablo 2. Üretilen rijit köpüklerin mekanik özellikleri

Codes	Density (g/cm^3)	Flexural Resistance (N/mm^2)	Compression Resistance (N/mm^2)
P-0	0,017 (17,65) A	0,005 (20,00) A	0,001 (50,00) A
U-1	0,047 (0,09) B	0,032 (20,73) B	0,007 (63,13) A
U-2	0,081 (0,32) C	0,076 (11,02) C	0,018 (24,15) BC
U-3	0,083 (3,09) C	0,082 (11,80) C	0,016 (22,16) B
U-4	0,109 (3,05) D	0,086 (13,55) C	0,018 (2,22) BC
U-5	0,128 (1,61) E	0,11 (6,06) D	0,021 (7,70) BCD
B-1	0,064 (5,28) F	0,039 (24,85) B	0,019 (4,81) BC
B-2	0,080 (9,70) C	0,078 (17,42) C	0,025 (10,83) CDE
B-3	0,089 (5,93) C	0,090 (3,07) CD	0,027 (8,32) DE
B-4	0,107 (8,37) D	0,095 (21,53) CD	0,030 (5,98) E
B-5	0,137 (2,33) E	0,073 (9,03) C	0,016 (48,24) B

Note: Parentheses indicate the coefficient of variation (CV, %). A, B, C, D, E. F letters indicate the significant differences between the groups.

The flexural resistance was found between 0,005 N/mm^2 with %20,00 CV and 0,11 N/mm^2 with %6,06 CV. The highest flexural resistance was obtained from U-5. However, U-5, B-3, and B-4 statistically belong to the same group in terms of flexural resistance. The lowest flexural strength was obtained from P-0 according to Tukey-Kreamer Honestly Significant Differences (HSD) test. High coefficient of variation values was obtained from P-0 and U-1, it is thought that due to low solid content, the starting suspension did not mix homogenously and the material showed different behavior in different locations under external loads.

Flexural resistance increases with increasing fiber concentration in rigid foams which are reinforced with unbleached pulp. However, in the bleached pulp, especially B-5 the flexural resistance decreased. The reason for this decrease was thought to be due to the fact that the pulp came in pulp sheets and that the fibers were not disintegrated well and aggregation occurred when preparing the mixture. On the other hand, unbleached kraft pulp was not aggregated at a high concentration such as a 1/1 mixture ratio. Because unbleached kraft pulp was never dried pulp, it was easily disintegrated at suspension. The compression resistance found between 0,001 N/mm^2 with %50,00 CV and 0,03 N/mm^2 with %5,98 CV. The highest compression resistance was obtained from B-4. However, B-2 and B-3 statistically belong to the same group with B-4 in terms of the compression resistance value. The lowest compression strength was obtained from P-0 and U-1 as expected. The compression resistance increased with increasing fiber concentration in rigid foams.

Once the effectiveness and the impact on the final products' mechanical performance were compared, the bleached kraft pulp reinforcement was found to be higher. This is thought to be related to the lignin-free structure of bleached kraft pulp. The higher amount of fibers found in bleached kraft pulp produced higher performance properties compared to the unbleached kraft pulp reinforcements.

The density of commercial foam materials used for insulation ranges from 0.015 to 0.16 g/cm^3, and the compressive and flexural resistance changes between 0.089 to 0.55 N/mm^2 and 0.1 to 0.86 N/mm^2 respectively (Lee and Ramesh, 2004). Our results appear to be in the acceptable range compared to commercially available polyurethane, polystyrene and phenolic foams.

It was observed that by adding fiber, the rigid foam material improves its performance under bending and compression loads. It is thought that the PVAc has a strong interaction between abundant ester bonds with fiber surface (Geng et al., 2016). Besides, hydrogen bonds have a positive effect on the mechanical properties of the foam material (Iwatake et al. 2008). At the same time, the increase in the density and the increase in the amount of material to respond to the unit load against the unit area improved the mechanical properties.

Kang et al. (2014) produced foam material by using recycled fiber starch and polypropylene type glue. The compressive resistance was found 0.067 N/mm^2 and the flexural resistance was found 0.026 N/mm^2 at a density of 0.02 g/cm^3, which was higher than our results. In another study, the compressive strength of foam material produced using %2 nano cellulose was found to be 0.013 N/mm^2 (Yang et al., 2017). Yildirim (2018) found the compressive resistance to 0.169 N/mm^2. In another study, compressive resistances of the foams produced from the microfibril cellulose, dopamine and silane were found between 0.76 - 1.35 N/mm^2 (Li et al., 2017).

4. Conclusions

In this work, the innovative rigid foams were designed and manufactured without using foaming agents. The kraft pulp at different ratios (PVAc/ Kraft pulp (bleached and unbleached) - 1/0, 1/0.2, 1/0.4, 1/0.6, 1/0.8 and 1/1) was mixed with PVAc. Due to PVAc's low mechanical properties, kraft pulp was used as a reinforcing material and produced an increase in the performance properties of the final product. Increasing the kraft pulp content increased the density as expected. The addition of the fiber and increasing the amount of fiber produced higher mechanical properties except for PVAc/ bleached kraft pulp ratio at 1/1. It was found that the fibers were not disintegrated well and aggregation occurs when preparing the mixture at this ratio. The optimum properties were observed B-4, which showed promising mechanical properties. The bleached kraft pulp reinforcement produced higher enhancements on performance properties compared to unbleached kraft pulp reinforcement.

Future studies will focus on economic assessment and environmental load reductions. Fire retardant properties and thermal properties of the foam are being studied.

Acknowledegments

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