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Effects of different nano fillers on the physical and mechanical properties of medium density fiberboards (MDF)

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Abstract

In this study, the physical and mechanical properties of medium density fiber boards (MDF) produced with urea formaldehyde adhesive reinforced with nano-boron nitride (BN) and nano-titanium dioxide (TiO_2) were investigated. 0.5% and 1.5% nano-filled urea formaldehyde adhesives were used in the production of test samples. The density, thickness swelling, water uptake, internal bond strength, bending strength, modulus of elasticity in bending and SEM analysis of the boards prepered were determined. According to the laboratory tests conducted, there was no significant change in the density of the boards after nano filling. Physical properties such as water intake and thickness swelling were determined to decrease with the addition of nano particles. The mechanical properties of the boards increased with the addition of both nano-BN and TiO_2. According to the a results of thermogravimetric analysis, it has been determined that the thermal stability of urea formaldehyde adhesive with TiO_2 is increased and maximum mass loss temperatures are higher than the addition of nano-BN. The morphological structure of the urea formaldehyde adhesive was visualized with an electron microscope, and the nanoparticle dispersions were displayed and the particles were also analyzed by EDAX analysis. As a result, it has been determined that MDFs prepared with nanoparticle adhesives have improved physical, mechanical and thermal properties. Particularly, all samples with nano particles have been achieved the standards required in their mechanical properties. As a result, it can be sad that the adding nano-particles have positive effect on the physical and mechanical properties of the panels.

Anahtar Kelimeler: Fiberboards, Nanoparticles, Wood-based composites, Material characterization, Nano Boron Nitride, Nano Titanium dioxide.

Orta Yoğunluklu Liflevhaların (OYL) Fiziksel ve Mekanik Özellikleri Üzerine Farklı Nano Dolguların Etkileri

Öz

Bu çalışmada, nano bor nitrür (BN) ve nano titanium dioksit (TiO_2) ile güçlendirilmiş üre formaldehit tutkalı ile üretilen orta yoğunluklu liflevhaların (MDF) fiziksel ve mekanik özellikleri araştırılmıştır. %0,5 ve %1,5 nano dolgulu üre formaldehit tutkalı test örneklerinin hazırlanmasında kullanılmıştır. Hazırlanan levhaların yoğunluğu, kalınlığına şişmesi, su alması, iç yapışma direnci, eğilme direnci ve eğilmede elastikiyet modülü ve ayrıca tutkalın SEM analizleri gerçekleştirilmiştir. Yapılan laboratuvar testlerine göre, levhaların yoğunluklarında nano dolgu sonrası önemli bir değişim meydana gelmemiştir. Su alma ve kalınlığına şişme gibi fiziksel özellikler nano dolgularla azaldığı belirlenmiştir. Levhaların mekanik özellikleri ise hem nano-BN hemde TiO_2 ilavesi ile artmıştır. Termogravimetrik analizine göre, genellikle TiO_2 içeren üre formaldehit tutkalının yanma kararlılığının arttığı saptanmıştır ve maksimum kütle kaybı sıcaklıklarının ise nano-BN ilavesiyle TiO_2 ilavesine göre daha yüksek olduğu belirlenmiştir. Üre formaldehit tutkalının morfolojik yapısı electron mükroskobuyla görüntülenmiş ve nano partiküllerin dağılımları görüntülenmiştir ve ayrıca EDAX analizi ile partiküllerin analizi gerçekleştirilmiştir. Sonuç olarak nano partikül ilaveli tutkallarla hazırlanan MDF'ların fiziksel, mekansel ve termal özelliklerinde iyileşmeler olduğu belirlenmiştir. Özellikle nano partikül ilavesiyle hazırlanan örnekler, mekanik özellikler için istenen standartlar yakalanmıştır. Sonuçta, nano partikül ilave etmenin panellerin mekanik ve fiziksel özelliklerini üzerine olumlu etkilerinin olduğu söylenebilir.

Anahtar Kelimeler: Liflevha, nanopartkül, odun esaslı kompozitler, malzeme karakterizasyonu, nano bor, nano titanyum dioksit.

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1. Introduction

In the production of fiberboards, raw material used are lower value round woods, forest thinning materials, sawdust, plywood residues, fuel woods and other lignocellulosic materials (Suchsland and Woodson 1986, Eroğlu et al. 2000). In recent years, special board production has become widespread by making improvements and modifications in chemical materials used in board production, and manufacturers has started to produce glossy surface boards by using various nano fillers such as nanoclays, nano-TiO_2, etc. with higher physical and mechanical properties, moisture and fire resistance (Kumar et al., 2013, İstek et al. 2017). Properties of urea formaldehyde adhesive affect reaction time, pH value, temperature, catalyst concentration and molar ratio of urea formaldehyde (Pizzi et al., 1994) and adding nano fillers were provided by improve on the properties of the adhesives (Alabduljabbar et al. 2020, Taghiyari et al., 2020).

Urea-formaldehyde (UF) resin has been widely used as a wood adhesive. It is economical, cures rapidly, has excellent bonding strength, and gives a clear appearance in finished products (Park and Causin, 2013). Intensive efforts are being made to improve physical and mechanical properties of urea formaldehyde by using various technologies (Pizzi et al., 1994; Park and Jeong, 2011). Reinforcement with nano-fillers such as SiO_2, Al_2O_3, clays, etc. in different thermosetting polymers has been conducted to the improvement of the physical and mechanical properties of the wood-based panels such as fiberboards, particleboards, etc (Kumar et al., 2013; Ashori and Nourbakhsh 2009; Taghiyari et al., 2016; Kumar et al., 2014; Chen et al., 2015; Zahedsheijani et al., 2011). In a study, it was found that addition of nano-SiO_2 to urea formaldehyde resin can improve the resin bond strength and the mechanical properties of wood (Jiang et al, 2013). In another study, Chen et al. (2018) studied the properties of fibreboard based on nanolignocelluloses/$CaCO_3$/PMMA composite, and they found he composites materials have good mechanical, dimensional stability, and thermal properties which enhanced as the filler loading increased. Candan and Akbulut (2015) studied on the physical and mechanical properties of nanoreinforced particleboard composites. The results obtained showed that nanomaterial reinforcement technique significantly affected the physical and mechanical performance properties of the particleboard composites.

Nanotechnology can provide a major opportunity to the wood panel industry to develop new innovative materials. The nano-fillers when homogenously dispersed in matrix can improve the properties of thermosetting resins, and the significantly improved physical and mechanical properties can be achieved due to nano-scaled fillers (Wegner et al. 2005, Wegner and Jones 2006). In the wood-based panel area of research, nano-fillers can be used to improvement of physical and mechanical properties of the panels according to the previous studies.

The aim of this paper is to examine the influence of nano boron nitride (BN) and nano titanium dioxide (TiO_2) on the physical and mechanical properties of MDF panels. It was determined how board quality changed depending on nano BN and nano TiO_2 particles used

2. Materials ve Methods

2.1. Materials

Wood fibers and urea formaldehyde (UF) resin used in this study were supplied from Kastamonu Integrated Wood Industry Incorporated Company. Fibers were produced in the plant's fiber unit using 70% Beech (Fagus oriantalis L.) and 30% Black Pine (Pinus nigra Arn.) wood chip mixes. 17.5% UF glue was used in board production according to the full dry fiber weight. UF glue has 57 ± 1% solids and 8.2 pH and was produced in the same institution. As a hardener, 1% of the ammonium chloride (NH_4Cl) solution was used. As a water-repellent agent liquid paraffin was added by an amount of 0.5% based on the weight of the fully dried fibers. In addition to these, nano-boron nitride (BN) and nano-titanium dioxide (TiO_2) were used in granular form at 0.5% and 1.5% ratios, respectively, based on the total dry fiber weight. These nanoparticles were obtained from a commercial enterprise.

2.2. Preparation of the urea formaldehyde

When the nano additive glue was prepared, homogenous mixture was provided by adding nanoparticles at 0.5% or 1.5% of the amount of glue solids. Liquid paraffin and hardener were then added and the solution was mixed and homogenized. The resulting glue mixture was sprayed onto the fibers in an externally spraying device.

2.3. Manufacturing of the Medium Density Fiberboards (MDF)

The experimental boards were produced by using a forming mold of dimensions 400x400 mm, by hand laying to obtain a board of about 715 kg/m^3 density and 16 mm thickness. 190°C press temperature, 40 kg/cm^2 press pressure and 5 minutes pressing time were used in the production of boards. Panel samples with nanoparticles such as TiO_2 and BN at 0.5% and 1.5% loading rate and control boards without the nanoparticles were produced three panels for each formulation under the same production conditions and the average results for totally fiveteen panel samples were compared.

2.4. Methods

The test samples and their dimensions were prepared in accordance with TS EN 325 (1999) and TS EN 326-1 (1999). After cutting, each test sample was conditioned at 20±2 °C and 65±5% relative humidity for 2 weeks before testing. Water absorption (WA), thickness swelling (TS), internal bonding strength (IB; tensile strength perpendicular to the surface of the board), modulus of rupture (MOR) and modulus of elasticity in bending (MOE) were determined using TS EN 317, TS EN 317, TS EN 319, TS EN 310, TS EN 310, respectively. The obtained resuts were evaluated according to Turkish Standard (TS) 64-1 EN 622-1, TS 64-2 EN 622-2 and 3. Panel density profile measurements were carried out in GreCon Raw Density Measuring System. For thermal analysis of the binder TGA/DTG-DTA methods were used. Also, for image analysis, scanning electron microscopy (SEM) was used to determine the distribution of nanoparticles in binding material.

3. Results and Discussion

3.1. Density profile of the MDFs

The density profiles of the 5 different fiberboard groups produced in this study are very close to each other and are shown in Fig. 1.

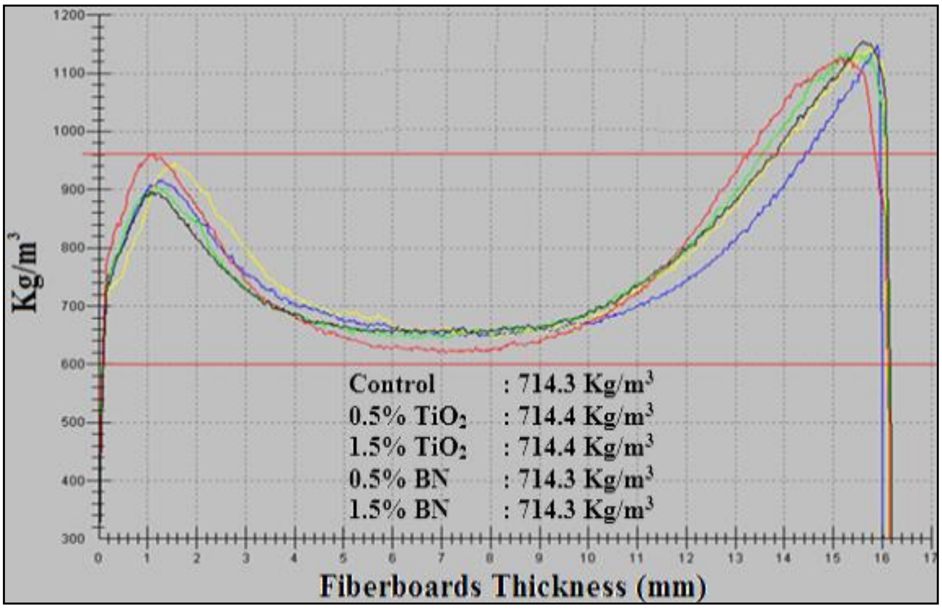

Figure 1. Density profiles of MDFs.

When the density profile was examined it was observed that there were significant differences between the upper surface and the lower surface densities of the boards. This case was explained by the waiting in cold environment and the mats get hot by carrying them with hot metal sheet during the production process in the laboratory. It has been determined that the densities of the test boards are within the target ranges. It was determined that there was no statistical difference among the densities of the experimental boards obtained according to the one-way ANOVA statistical studies. In study done by Nemli and Demirel (2007), it was found that there is a significant relationship between density profile and technological properties and the similar results were also found by Cengiz and Kalaycioglu (2005).

3.2. Physical and mechanical properties of the MDFs

The average values, standard deviations, Duncan test results for physical and mechanical properties of test samples were shown in Table 1.

Table 1. Physical and mechanical properties of test samples.

Properties		**Control**	**0.5 % BN**	**1.5 % BN**	**0.5 % TiO_2**	**1.5 % TiO_2**
Water Absorption (%) for	x	67.97 *A*	**45.68** *A*	**49.35** *B*	64.59 *A*	**48.89** *B*
24h (WA)	±s	2.41	4.02	2.37	4.40	5.10
Thickness Swelling (%)	x	32.54 *B*	**17.81** *A*	**19.16** *AB*	30.11 *C*	23.72 *B*
for 24h (TS)	±s	3.14	1.05	2.64	3.48	2.83
Internal bonding strength	x	0.71 *A*	**0.87** *AB*	**0.90** *B*	**0.88** *AB*	**0.94** *B*
(MPa)	±s	0.12	0.12	0.17	0.15	0.13
Modulus of rupture	x	28.43 *A*	**34.01** *B*	**31.46** *AB*	**32.82** *B*	**31.34** *AB*
(MPa)	±s	2.72	1.74	2.36	2.45	1.70
Modulus of elasticity in	x	3483 *A*	**4555** *B*	**4078** *AB*	**4294** *B*	**3931** *AB*
bending (MPa)	±s	435	393	599	533	475

x: average value, ±s: standard deviation, Means followed with the same letters such as A, B, C in the same column are being significantly significance among the groups ($p \leq 0.05$) according to the ANOVA statistical analysis.

As shown in Tab. 1, TS and WA values of the test samples were improved by the addition of the nano particles and the best as well as the lowest WA and TS values were obtained from the 0.5% BN added experimental boards. The relationship between nano particle addition and TS and WA (24-h) values were found to be statistically significant among groups. TS and WA increased with increasing of BN contents but they decreased with increasing in nano-TiO_2 particle loadings. As a result, both TS and WA of the samples with nano particle was lower than the control samples It is believed that the improvement in water uptake rates is due to the fact that nano particulates fill the urea formaldehyde molecule voids and bring a tighter structure. On the contrary, the increase in the thickness of the nano-TiO_2 added boards indicates that it does not form any bonds with urea formaldehyde.

According to TS EN 64-1 EN 622-2 standars for medium density fiberboards, thickness swelling value of the samples with BN were found to provide the requested values (min. 20% for thickness >10 mm) and the thickness ratio for the samples with BN were 17.8 for 0.5% BN and 19.2% for 2% BN. As seen as Tab.1, the mechanical properties of the test boards were significantly improved by addition of the nano particles (1.5 % BN and TiO_2) to the urea formaldehyde glue. It was determined that the tensile strength perpendicular to the surface of the test specimens was improved with nanoparticle addition. This improvement was found to be statistically significant on experimental boards with 1.5% BN and TiO_2 additions. It was determined that the highest IB strength was found as 0.94 MPa in 1.5% TiO_2 samples. As shown in Tab. 1, the addition of nanoparticles improved MOR and MOE but began to fall again when 1.5% is used. MOR and MOE values of both groups were analyzed statistically and the differences were found to be significant at 95% confidence level in compared to control plates and 0.5% nanoparticle-added boards.

As evaluating according to the TS EN 64-1 EN 622-2, the mechanical properties of the all samples were better than the requested strength and moludus (12 MPa, 0.1 MPa and 1800 MPa) in the standard and the strength, internal bonding and modulus of the samples changed from 28.4 MPa, 0.7 MPa and 3483 MPa to 34 MPa, 0.9 MPa and 4555 MPa, respectively. In a study, SiO_2 nano-fillers were added to ultra-low density fiberboards, and according the results obtained, it was determined that the addition of the nano-fillers increased the modulus of elasticity of bending, bending strength, and internal bonding strength of the fiberboards with nano-SiO_2. This is because the SiO_2 is inorganic filler, which could be making the material become more brittle (Chen et al., 2015). In another study, Kumar et al. (2014) investigated the effect of multi walled carbon nanotubes (MWCNT) on the physical and mechanical properties of MDF panels, and it was found that mechanical properties generally increased with the concentration of nano-fillers, and the addition of MWCNT in urea formaldehyde resin did not have much effect on the water absorption properties of MDF panels. Some scientific studies showed that the reinforcing with nano particle improved the phormaldehyde emission and thermal properties of the wood panels (Xia et al., 2009; Taghiyari et al. 2013).

3.3. Thermal analysis of the urea formaldehyde with the nano-fillers

After the reinforcing with nano particles, all properties of the adhesive was affected by the reinforcing and therefore, thermal stability of the adhesive was investigated with TGA after adding the nano particles to the matrix. Thermogravimetric analysis of the urea formaldehyde with nano boron nitride (BN) and nano titanium dioxide (TD) were shown in Fig. 3 and Fig.4. TG curves show the thermogravimetric analysis of the urea formaldehyde with the nano-fillers as a function of temperature. The main degradation points are at 100 – 180°C, 180 – 240°C (the temperatures obtained from TG curves), and 240 – 360°C (the temperature obtained from DTG curves). As seen as Fig. 3, TGA curves of the urea formaldehyde with nano-fillers showed to be similar to each one. The weight loss was found to range from 89.6% for 0.5%BN to 83% for 1.5%TD. The lowest weight loss of the urea formaldehyde was determined in the nano-TiO2 samples. The highest value of DTG was obtained at 295.5°C for 1.5%BN according to Fig.4. As a result, the urea formaldehyde with BN exhibited thermal degradation at higher temperatures. Besides, the lowest weight loss was found in urea formaldehyde with TD. Kumar et al. (2014) conducted a study about the reinforcement of urea formaldehyde with multi walled carbon nano tubes (MWCNT). The results showed that the addition of MWCNT does not affect the thermal stability of the urea formaldehyde; but, weight loss decreased in the presence of MWCNT.

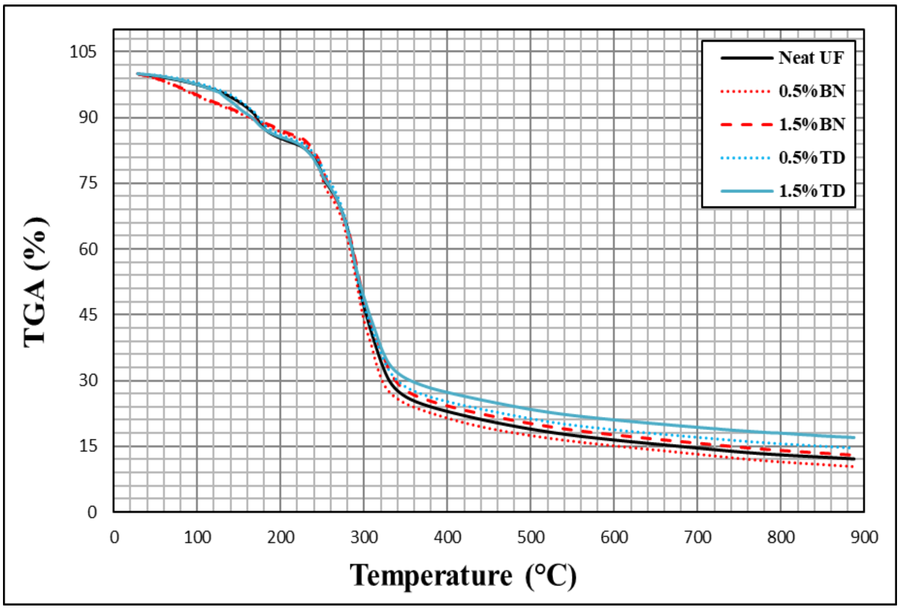

Figure 3. Thermogravimetric analysis of the urea formaldehyde with the nano-fillers.

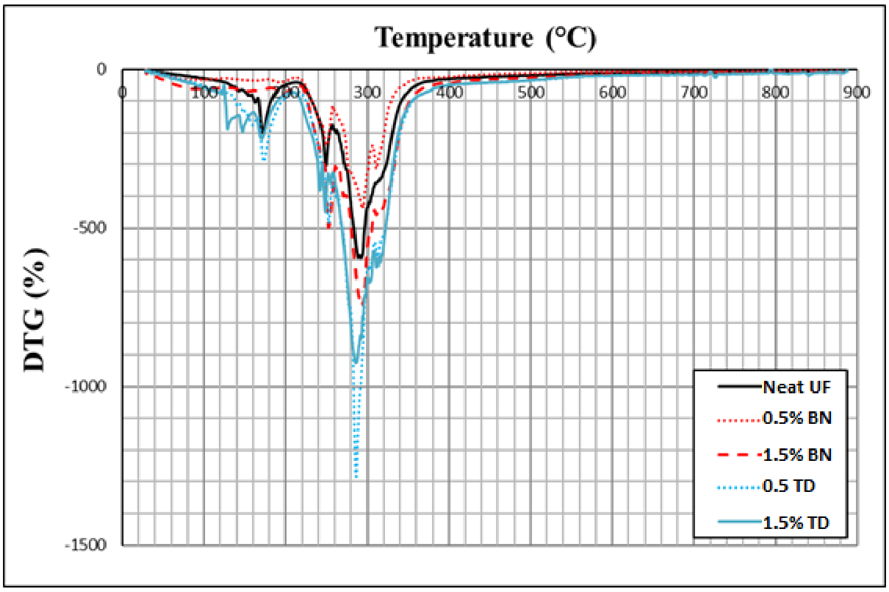

Figure 4. Derivative thermal gravimetry of the urea formaldehyde with the nano-fillers.

3.4. SEM images of the urea formaldehyde with the nano-fillers

The polymer matrise generally were reinforced with various nano particle such as nanoclays, TiO2, Al2O3, etc. to obtain good properties such as larger surface area, high mechanical properties or smoother surface, etc and the reinforcing effects the morphological characterization of the matrix. In this study, SEM spectroscopy was used to examine the morphological chages in the adhesive. SEM images of the urea formaldehyde with nano boron nitride (BN) and nano titanium dioxide (TD) were shown in Fig. 5.

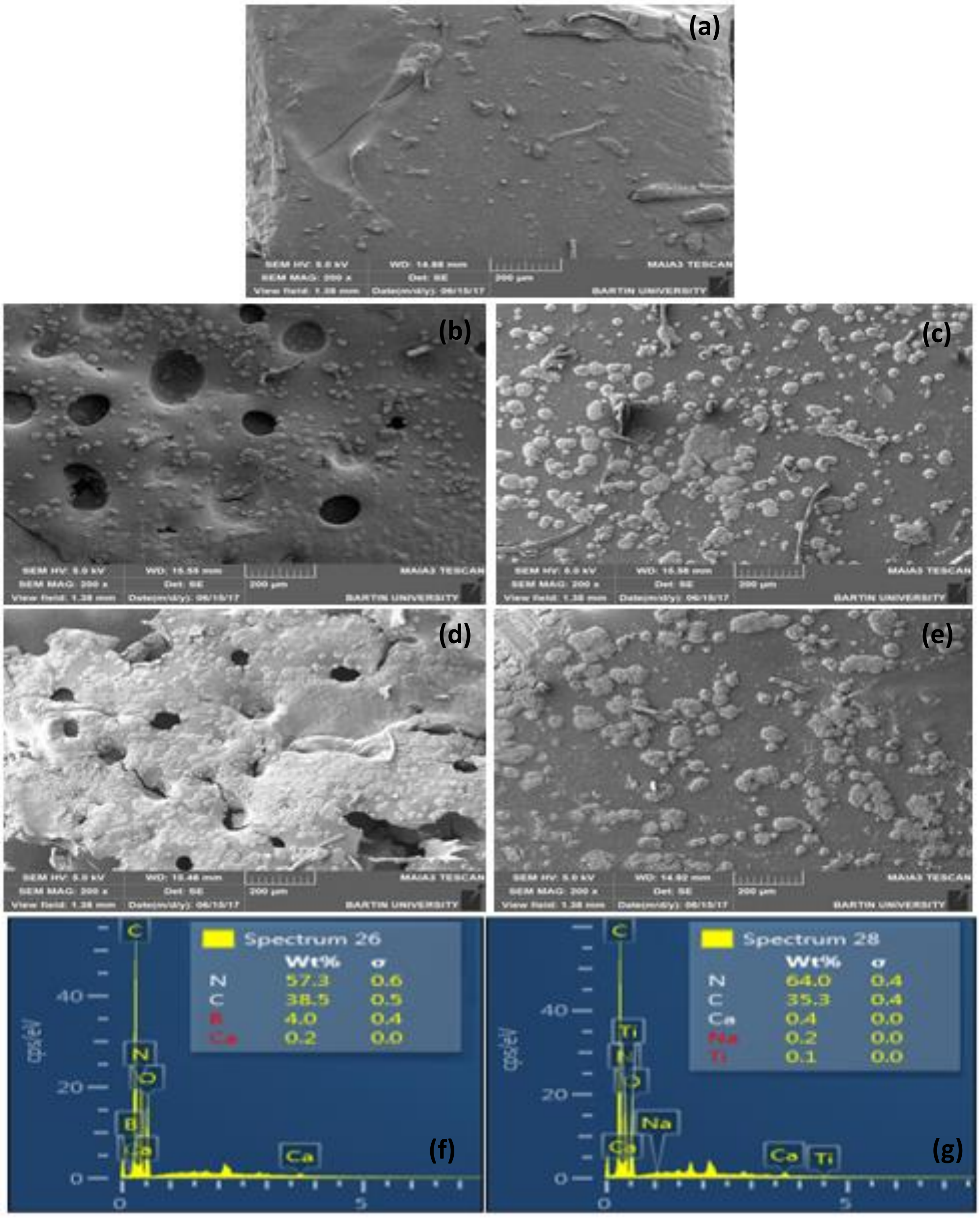

Figure 5. SEM and EDAX images of the urea formaldehyde with the nano-fillers. (a) neat polymer matrix, (b), (d) and (f) the samples with 0.5 and 1.5% BN, and (c), (e) and (g) the samples with 0.5 and 1.5% nano-TiO_2.

According to the SEM images, both nano-TiO_2 and BN were well dispersed in the urea formaldehyde, but some aggregates were found in the urea formaldehyde, and also porous structures with the addition (0.5%) of both TiO_2 and BN in the matrix polymer were determined. Both nano-TiO_2 and BN were determined on the surface of the urea formaldehyde with scanning of EDAX.

4. Conclusions

Physical and mechanical properties of the experimental boards generally satisfied the requirements for fiberboard defined in the related standards in results and discussion section. The best WA and TS values were found in the 0.5% BN added experimental boards. TS and WA did not significantly change with increasing nano particle contents. However, mechanical properties were affected significantly by means of added nano particle in adhesives. According to the thermogravimetric analysis, thermal properties of the urea formaldehyde with nano-fillers were determined to be similar, and the best thermal properties was found to be for the urea formaldehyde with BN. Some aggregates and holes in the structure of the urea formaldehyde with nano-fillers were determined. As result, the addition of the nano-fillers to the urea formaldehyde has a positive effect on the general properties of the fiberboards.

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