Properties of Boards Prepared from Mixture of Mineral Adducts with Bark and Cone of Calabrian Pine (*Pinus brutia*)

ABSTRACT

Olivine and dolomite addition with some formulations clearly retarded the water absorption change in both bark (Type 1 boards) and cone (Type 2 boards) based experimental panels. However, the lowest water absorption value of 15.20% was found in dolomite contain (40:60, dolomite-bark chip, w/w, %) sample which show approximately 169.6% lower than olivine contain sample at similar proportions. However, it is important to note that Type 1 boards show considerably lower water absorption than Type 2 boards at similar experimental conditions. The boards which produced with 10-, 20-, and 30% dolomite and 90-, 80-, 70% bark chips (XBI, XBII and XBIII) proportions, show higher thickness swelling (TS) than control (B0) while rest of show lower TS than control samples, regardless of type of mineral or experimental conditions. The highest TS improvement of -79.1% was found with sample of YBV which produced with equally (1:1, w/w) proportions olivine and pine cone chips in matrix. It is noticeable that dolomite addition to Type 1 boards are usually effects higher IB values than olivine added samples while the highest IB value of 0.72 MPa was found with sample of XBV. Moreover, only sample of XBII (20% dolomite and 80% pine bark chip in boards proportions, w/w) show higher MOR strength value (3.26 MPa) than control (B0: 3.63 MPa), regardless of mineral proportions. In Type 2 boards, all experimental panels that prepared with two different mineral addition at five loading levels show lower MOR values than control (C0: 6.59 Mpa). It is also realized that both Type 1 and 2 type experimental panels show lower elastic property (MOE) than control samples (B0: 1129.3 MPa: C0: 820.1).

Keywords: Dolomite, olivine, Calabrian Pine bark, Calabrian Pine cone, mechanical properties, biocomposites

1. INTRODUCTION

In these days, requisition for natural materials in composite production has been incrementing incredibly due to urbanization and expeditious industrial development. However, bio-composites have conventionally been achieved by mixing bio-based raw materials with various synthetic binders. Some common of those are mainly particleboard, fiberboard and plywood in different proportions as per design strength requirement [1-2]. Despite many advantages of those materials, there is also some important drawback which are low fire, heat-, and moisture resistance [1-5]. In these respects, environmentally friendly bio-composite manufacturing has long been employed to alter undesirable lignocellulosic properties [4-5]. Nevertheless, the use of synthetic additives has major downsides, specifically the adverse environmental impacts associated with their production, their relatively high cost, and huge energy consumption during their production. In recent years sizably voluminous greenhouse gas emission (mainly CO_2) and high energy required in the production of wood products, causing a severe difficulty on ecology [6]. Considering the negative impacts of using petroleum-based additives (i.e hydrophobic substances, reinforced elements), industries and researchers are seeking alternative cost-effective, and durable elements to replace or minimize the use of traditional additives in composite manufacturing process [7-8]. The most appropriate way to overcome the environmental impact is the effective utilization of alternative additives or lignocellulosic waste materials in matrix structure. The benefits of such materials in composite are documented by various researchers [9-11]. In recent studies, unconventional wastes such as forest residues of bark and cones have been utilized in greener material production [12-13]. Several literature interpretations reported the potential of pine cone and bark as tree residue using in composite and paper making [12-14]. However, there has been an increasing interest in the use of non-traditional additives to improve the engineering properties of bio-composites. As a result, a variety of novel or improved sustainable materials has been proposed and well documented in the literature. Among the diverse these additives, minerals, including cement, gypsum and magnesia have been investigated for their utilization in various type material manufacturing experiences [15-17]. It has been proposed that some minerals are capable of forming stable structures with strength improvement properties after reacting with the wood in the presence of water. Dolomite is a mineral formed by the combination of calcium (Ca) and magnesium (Mg) in limestone ($CaCO_3.MgCO_3$) [5]. It is mostly utilized for production of iron- steel, ceramics, paints, fertilizer, glass, cement and bricks [5]. Olivine is classified as a rock-forming mineral. Its chemical formula is (Mg2+, Fe2+).2SiO_4 and usually found in crystal aggregates [5]. The various literature studies revealed that the utilization of dolomite as a binder and granular material (fine and coarse) in cement concrete and other applications influences their property. Various studies already exist on the utilization of dolomite- and olivine waste as material substitute in metallurgy and construction industry [5, 17-20].

Although there is some literature reports that improve many properties of lignocellulosic matter when used in composite matrix together. But there are very limited studies on the use of olivine and dolomite in the forest products industry. However, limited to no study has been undertaken to evaluate the effect of non-traditional additives such as; olivine and dolomite and their combination with mixing lignocellulosic in bio-composite manufacturing [18-20]. The high resistance properties of olivine and dolomite against abrasion and fire could be a potential material for bio-composite manufacturing. Therefore, the objective of this study was to examine selected mechanical performances of experimental panels manufactured from olivine and dolomite mixed with pine tree residues of cone and bark particles.

2. MATERIALS AND METHODS

Calabrian pine tree (*Pinus brutia* Ten.) residues of bark and cones were obtained from Isparta region in Turkey. The pine cone and barks were turned to chips at laboratory type hammer mill. The chips were screened to 1-3 cm particles and dried in the oven at 105 (± 3 ^oC) until they reached 2-3% moisture content. The bonding agent employed was urea-formaldehyde resin supplied by a commercial operated particleboard plant, in Turkey. It has 65% solid content and 20% ammonium chloride hardener, utilized as received. The glue was applied 10% and hardener was 1% in the test samples by weight based on oven dry material. The minerals of olivine and dolomite were received from a company that operated in Isparta-Aksu mining site, in Turkey. Metal mold plates with the dimensions of 400 x 400 x 10 mm was used to prepare the board paste. Then it were pressed for 5 minutes under 2.5 N/mm^2 at 170-180 ºC with laboratory type electrically heated press. Samples were kept between metal plates after the end of pressing process and then climatized. The experimental panels were conditioned at 23 ºC and 65% relative humidity and samples were cut to determine the IB (Internal Bond), MOE and MOR (Modulus of Elasticity and Rupture), TS (Thickness Swelling after two- and 24-hours' immersion in water) and The Water Absorption (WA, %), in accordance with TS EN 310 (1999), TS EN 319 (1999) and TS EN 317 (1999), ASTM D 1037, standards, respectively.

Some code number and abbreviations were established throughout the study, the code number established for experimental boards are; **X:** Dolomite, **Y:** Olivine, **B:** Bark chip in mixture (Type-1 boards), **C:** Cone chip in mixture (Type-2 boards), **X-/YI, II, III, IV and V:** Dolomite and olivine proportions in mixture (w, gr, %) of 10-,20-,30-,40- and 50%, respectively.

3. RESULT AND DISCUSSION

Water absorption and thickness swelling tests are popular tests for evaluating the durability through water transportation mechanisms via pores in composites. Tables 1 summarize water absorption (WA, %) properties of boards produced by adding various proportions of dolomite and olivine to Calabrian pine bark- and cone chips in composite structure. Data of WA (%) in Table 1 show that olivine and dolomite addition with some formulations clearly retarded the absorption change in both type boards. It can be seen that olivine is proved to be considerably more effective in reducing the water absorption with samples of YBI-III and YCI-II while dolomite show lowering absorption properties in samples of

XBIV-V and XCIII-V, at similar experimental conditions. However, the lowest absorption value of 15.20% was found with sample of XBIV which indicates approximately 169.6% lower value than counterpart sample of YBIV. It is also notable that bark Type-1 boards show considerably lower water absorption than Type-2 boards at similar experimental conditions. This is probably due to higher flexibility and easier arrangement of bark chips rather than cone chips when mixed with minerals in network matrix structure.

Table 1. The water sorption (%) properties of experimental boards.

Boards	X	Y	Δ from each other (X or Y, %)
Mineral- pine bark based panels			
B0	36.73	36.73	0
XBI-YBI	30.58	19.33	36.8 (-Y)
XBII-YBII	40.49	25.16	37.8 (-Y)
XBIII-YBIII	49.74	27.70	44.3 (-Y)
XBIV-YBIV	15.20	40.98	169.6 (-X)
XBV-YBV	25.83	32.25	24.9 (-X)
Mineral-pine cone based panels			
C0	53.68	53.68	0
XCI-YCI	60.29	59.04	2.1 (-Y)
XCII-YCII	62.55	48.58	22.3 (-Y)
XCIII-YCIII	46.18	75.39	63.3 (-X)
XCIV-YCIV	36.37	37.89	4.1 (-X)
XCV-YCV	30.69	59.19	92.9 (-X)

Figure 1 depicts the water absorption (WA, %) and differences (Δ%) of boards from controls (B0-C0), produced by adding various proportions of dolomite and olivine to pine bark- and cone chips in composite structure. In general, for bark-based (Type-1) experimental panels, it can be seen that samples of XBII, XBIII and YBIV show 10.3%, 35.4% and 11.5% higher WA properties than controls while others show in range of -58.7% (XBIV) to -12.2% (YBV) lower WA properties than controls. For cone-based (Type-2) boards, samples of XCI, YCI, XCII, YCIII and YCV samples show 12.3%, 9.9% and 16.5%, 40.4% and 10.2% higher WA properties than controls while others show in range of -542.8% (XCV) to -9.5% (YCII) lower WA properties than control samples. Although it is not easy to correlate WA properties of boards with panel mixture proportions, but there is considerably lowering WA properties observed at certain conditions.

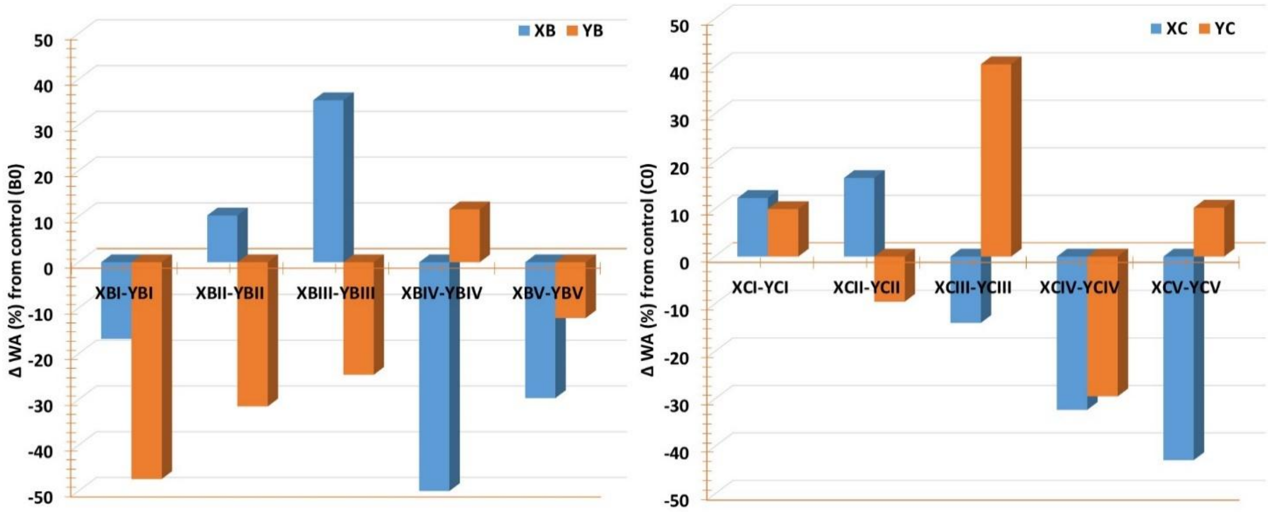

Figure 1. Water absorption (Δ%) difference properties of experimental boards from controls

Table 2 shows the comparative thickness swelling (TS, %) properties of boards after 24 h. immersion in water. It is also noticeable that samples of XBI-III show higher TS values than control sample (B0: 11.73%), all other panels show lower TS values than controls, regardless of experimental conditions or preparation formulations. However, except sample of XBIV which show approximately 59.9% lower TS than counterpart olivine added samples (YBIV), in all other olivine formulated samples show in range of 1.3% (YCII) to 73.9% (YCI) lower TS than dolomite formulated at similar conditions. This is probably due to higher compability of olivine with both pine bark- and cone in matrix structure.

Table 2. The thickness swelling (%) properties of experimental boards.

Boards	X	Y	Δ from X-Y (%)
Mineral- pine bark based panels			
B0	11.73	11.73	0
XBI-YBI	12.94	6.47	50 (-Y)
XBII-YBII	17.95	5.48	69.5 (-Y)
XBIII-YBIII	16.92	8.76	48.2 (-Y)
XBIV-YBIV	5.16	8.25	59.9 (-X)
XBV-YBV	6.79	2.46	63.8 (-Y)
Mineral-pine cone based panels			
C0	21.44	21.44	0
XCI-YCI	20.15	5.25	73.9 (-Y)
XCII-YCII	15.93	15.73	1.3 (-Y)
XCIII-YCIII	13.51	8.24	39.1 (-Y)
XCIV-YCIV	16.61	12.55	24.4 (-Y)
XCV-YCV	13.66	6.06	55.6 (-Y)

Figure 2 shows the thickness swelling differences (Δ%) of boards from controls (B0-C0), made by adding various proportions of dolomite and olivine to red pine bark- and cone chips in composite structure. However, except samples of XBI-III which show higher TS than control (B0), all other bark- and cone-based samples show lower TS values than control samples, regardless of type of mineral additive or proportion in panel structure. The highest TS improvement of -79.1% was found with sample of YBV, followed by samples of YCI (-75.5%) and YCV (-71.7%). It is notable that except sample of XBIV which show higher swelling retardant than counterpart sample of YBIV (-56.1% vs -29.7%), olivine addition in to all bark- and cone-based samples show lower TS property under similar additive proportions and manufacturing conditions than dolomite added samples. It is clear that the marked effect of lowering TS with both mineral addition to cone and bark type boards which is noticeable as shown in Table 2 and Figure 2.

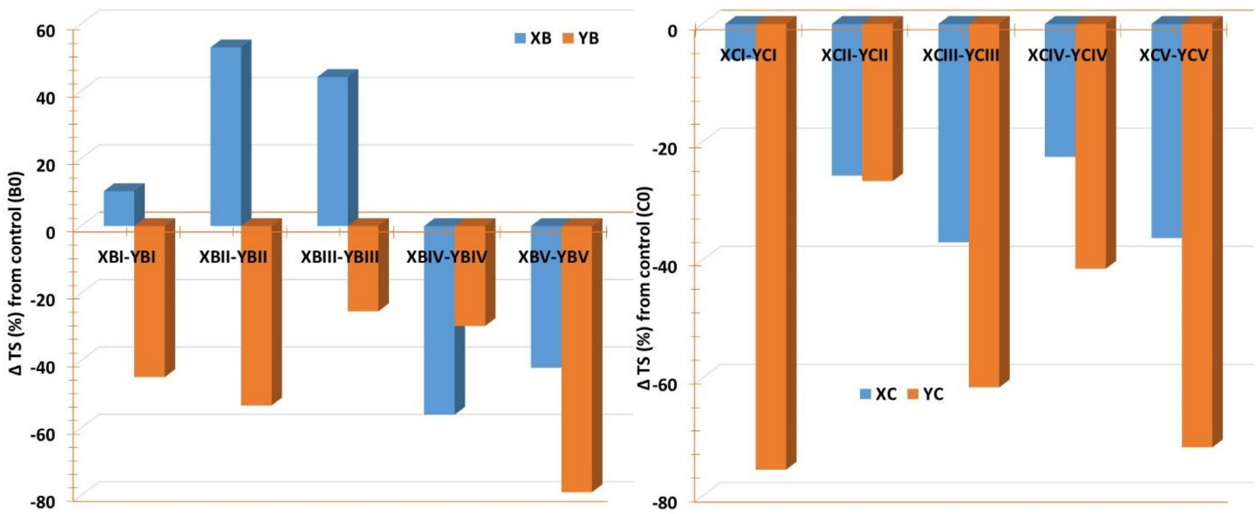

Figure 2. Thickness swelling (Δ%) difference properties of experimental boards from controls

Average values of IB, MOR, and MOE values obtained from two type experimental panels which produced with various proportions with mineral additives are summarized in Table 3. As it seen in Table 3, pine cone control sample show higher IB (B0: 0.36 MPa, C0: 0.90 MPa) and MOR strength properties (B0: 3.63 MPa, C0: 6.59 MPa) while control bark panel's MOE value is higher (B0: 1129.3, C0: 820.1 MPa).

For bark-based (Type-1) boards, dolomite addition is usually affecting higher IB values than olivine added samples while the highest IB value of 0.72 MPa was found with sample of XBV. However, there is not a clear correlation was found between mineral type of proportions for MOR and MOE properties. The highest MOR value of 3.26 MPa was found with sample XBII which is approximately 59.2% higher than counterpart olivine added sample (YBII: 1.33 MPa). It is also noticeable that only sample of XBII show higher MOR value than control (B0: 3.63 MPa), regardless of mineral formulations. All mineral added bark type boards show lower MOE values than control sample (B0: 1129.3 MPa). The olivine added samples of YBI and YBII show %9.5 and 60.1% higher MOE values than counterpart dolomite added samples, respectively. However, increasing dolomite content in bark-based panels (XBIII- XBV) show higher MOE values than olivine samples which prepared similar conditions.

For cone-based (Type-2) boards, only samples of XCII, YCII and YCIII show higher IB properties than control sample (C0: 0.90 MPa). The highest IB value of 1.32 was found with sample YCII, followed by YCIII (1.10 MPa) and XCII (0.97 MPa) in that order. It is notable that olivine is more effective for higher IB property of experimental board up to 30% (w/w, %) addition level (YCI-YCIII) while dolomite is more effective in 40 and 50% addition level in panel formulations (XCIV and XCV). However, all experimental panels that prepared with two mineral addition at five loading levels show lower MOR values than control (C0: 6.59 MPa), regardless of experimental conditions. The lowest MOR value of 1.90 MPa was found with sample of XCIV. Similar trend was also found for MOE values that all experimental panels which prepared with mineral additions show lower MOE than control (C0: 820.1 MPa). It is also notable that except sample of YCV which prepared with 1:1 olivine/pine cone chip mixture (w/w), all other samples which prepared with dolomite show higher MOE properties than olivine added samples at similar conditions.

Table 3. The mechanical strength properties of experimental boards.

Samples	IB (MPa)			MOR (MPa)			MOE (MPa)		
	X	Y	Δ from X-Y (%)	X	Y	Δ from X-Y (%)	X	Y	Δ from X-Y (%)
Mineral-red pine bark-based panels									
B0	0.39	0.39	0	3.63	3.63	**0**	1129.3	1129.3	0
XBI-YBI	0.50	0.32	36 (+X)	4.42	2.29	**48.2 (+X)**	740.3	810.7	9.5 (+Y)
XBII-YBII	0.58	0.49	15.5 (+X)	2.37	2.39	**0.8 (+Y)**	449.4	719.4	60.1 (+Y)
XBIII-YBIII	0.57	0.29	49.1 (+X)	3.26	1.33	**59.2 (+X)**	609.4	458.6	24.7 (+X)
XBIV-YBIV	0.59	0.26	55.9 (+X)	1.76	1.19	**32.3 (+X)**	812.6	704.1	13.4 (+X)
XBV-YBV	0.72	0.38	47.2 (+X)	1.23	1.37	**11.4 (+Y)**	503.1	417.1	17.1 (+X)
Mineral-red pine cone-based panels									
C0	0.90	0.90	0	6.59	6.59	0	820.1	820.1	0
XCI-YCI	0.84	0.89	5.9 (+Y)	4.94	3.51	28.9 (+X)	601.7	519.6	13.6 (+X)
XCII-YCII	0.97	1.32	36.1 (+Y)	3.76	4.0	6.4 (+Y)	525.1	457.8	12.8 (+X)
XCIII-YCIII	0.72	1.10	52.8 (+Y)	1.92	2.04	6.3 (+Y)	471.1	614.9	30.5 (+Y)
XCIV-YCIV	0.87	0.74	14.9 (+X)	1.90	2.55	32.8 (+Y)	623.9	540.1	13.4 (+X)
XCV-YCV	0.70	0.69	1.4 (+X)	2.27	1.13	50.2 (+X)	603.6	634.6	5.1 (+Y)

Figure 3 shows the Internal Bond (IB) strength differences (Δ%) of boards from controls (B0 and C0), made by adding various proportions of dolomite and olivine to red pine bark- and cone chips in composite structure. However, except samples of YBI, YBII and YBIV which show -17.9%, -25.6% and -2.6% lower than control board (B0: 0.39 MPa), all other bark-based boards show higher IB values than control sample. The highest IB difference (improvement) of 84.61% was found with sample of XBV, followed by samples of XBIV (51.3%) and XBII (48.7%). It is clear that all dolomite addition in to all bark type panels show higher IB property under similar manufacturing conditions than olivine added bark samples. It is clear that the marked effect of increasing IB with dolomite addition to bark type boards which is noticeable as shown in Table 3 and Figure 3. It is noticeable that only cone-based boards of XCII, YCII and YCIII show higher IB values than control (C0: 3.63 MPa).

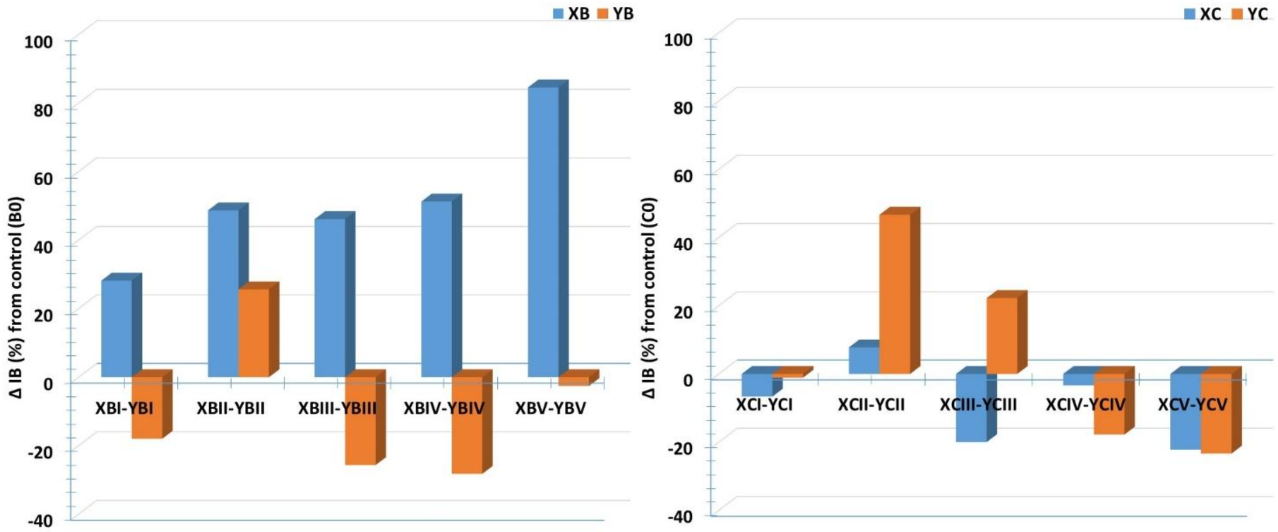

Figure 3. IB strength (Δ%) difference properties of experimental boards from controls

The bending strength (MOR) differences (Δ%) of boards from controls (B0 and C0), made by adding various proportions of dolomite and olivine to red pine bark- and cone type boards re shown in Figure 4. In general, except sample of XBI which show only 21.8% higher MOR value than control (B0), rest of experimental panels show lower MOR values than control samples (B0: 3.63 MPa, C0: 6.59 MPa), regardless of panel type and mineral additive proportions. The highest MOR reduction (-67.2%) was found for YBIV sample in bark type boards and YCIII (-70.9%) in cone type boards. It could be concluded that the addition of both mineral in top in cone bark- and cone type boards are not improving effects on MOR strength properties. Since the polymerization level and crystallinity of cellulose of bark and cone are less than wood [25], it may be suggested that when these used as reinforced element in matrix, could be lowering effects. Those may enables matrix mechanical properties to occur lower degree than wood-based systems. The results obtained in this study confirm this suggestions.

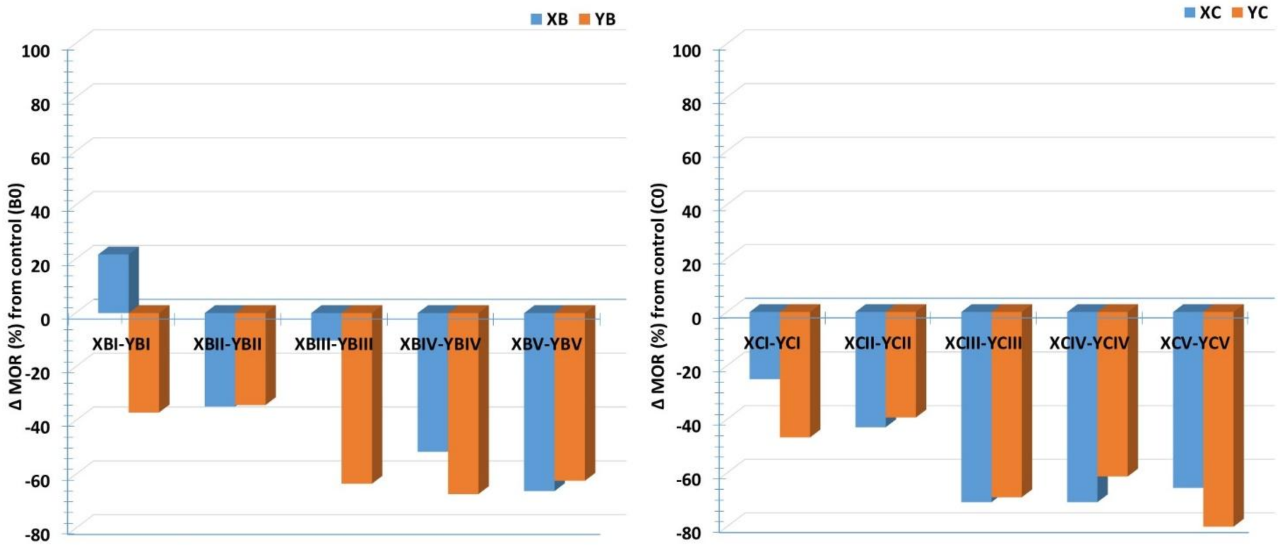

Figure 4. MOR strength (Δ%) difference properties of experimental boards from controls

The elasticity (MOE) property changes (Δ%) of boards from controls (B0 and C0), made by adding various proportions of dolomite and olivine to pine bark- and cone-based samples are shown in Figure 5. It can be clearly seen that all the samples show lower elasticity properties than controls (B0: 1129.3 MPa: C0: 820.1), regardless of formulation types and experimental conditions. The highest MOE reduction (-63.1%) was found for YBV sample in bark-based boards and YCII (-44.2%) in cone-based boards. It could be expected considering more rigid mineral addition to bark- and cone particles/chips in matrix network structures. But there is not any clear correlation was found between type of mineral additives and lignocellulosic type (cone or bark). It is also important to note that the MOE values was calculated lower than the standard value for all boards that red pine bark/cone chips and dolomite/olivine mixture, including the controls.

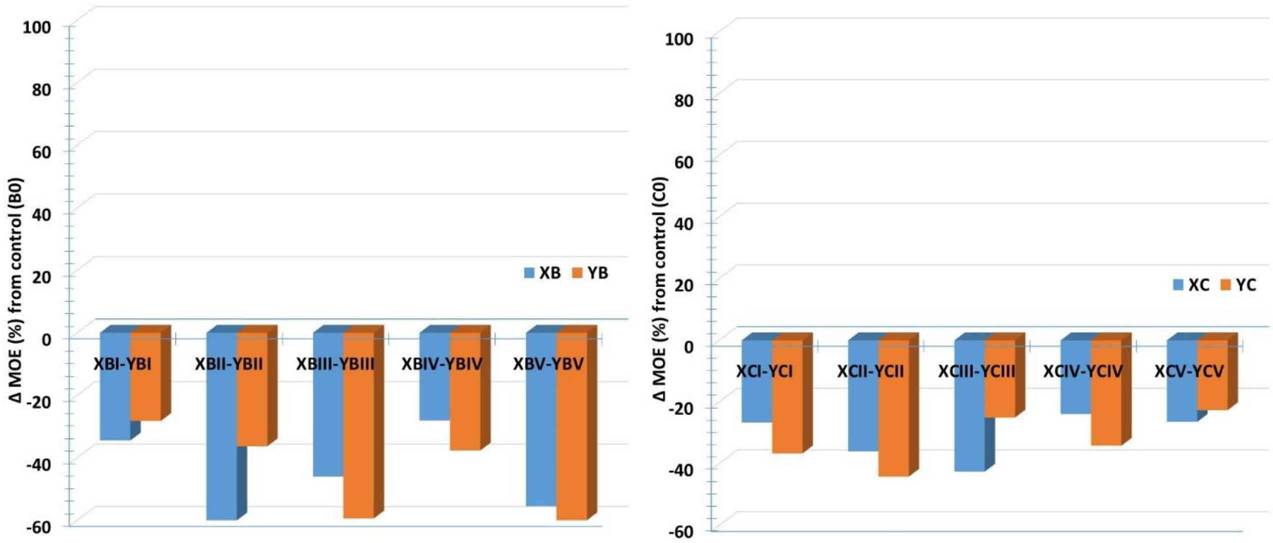

Figure 5. MOE (Δ%) difference properties of experimental boards from controls

4. CONCLUSION

Calabrian pine cone and bark chips were used for experimental boards manufacturing with various proportions of minerals of olivine and dolomite in the presence of urea-formaldyhde adhesive. It is important to note that olivine and dolomite as mineral adducts clearly retarded the water absorption properties of both type experimental boards. However, mineral-lignocellulosic based hybrid experimental samples show some comparable properties when used certain proportions. In certain conditions, dolomite and olivine as mineral adducts to lignocellulosic matrix could contribute positive effects on properties. These type of hybrid composites could be used as non-structural or lower strength needed places to meet demand on wood-based materials.

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