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Effect of Boron Waste on Mechanical and Durability Properties of Concrete

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Abstract

It was emphasized that 8% of the CO_2 emission that causes global warming occurs during cement production. In addition, the consumption of natural resources used in cement production increases due to the increase in cement demand with the widespread use of concreting. In this context, studies on alternative binders continue in order to reduce cement production and consumption. Today, besides the use of mineral additives such as blast furnace slag, fly ash and silica fume as binders in concrete mixtures, studies on the disposal and sustainability of other industrial wastes are continuing. One of the wastes generated in the industry is formed in boron plants. It is important to evaluate boron wastes especially in Turkey, which has significant boron reserves in the world. Studies on the use of boron wastes in concrete mixtures have recently become widespread. In this study, a comprehensive literature search was conducted on the effect of boron waste usage on the fresh state, mechanical, durability and radiation absorption properties of cementitious systems. In this context, with the increase in the use of boron waste and the increase in its usage rate, the radiation absorption properties of cementitious systems, their resistance to high temperatures and freeze-thaw have improved. However, with the addition of waste, the fresh water requirement and setting time of the mixtures increased. In terms of compressive strength, if the boron wastes in the mixtures exceed 10%, their compressive strength generally decreased.

Keywords

Boron waste, cementitious systems, sustainability, strength, durability performance

1. INTRODUCTION

Cement production exceeds 4 million tons per year due to the increase in the need for concrete in infrastructure and residential constructions [1]. Since cement production, which is responsible for 8% of the CO_2 emitted in the world, causes important problems such as global warming and climate change, the use of sustainable binder materials, which are less harmful to the environment, is being investigated in many countries to reduce cement consumption [2]. In this direction, the use of different mineral additives such as fly ash, silica fume and blast furnace slag by substituting and activating them instead of cement has become widespread in concrete production [3,4]. In addition to these materials, the use of different types of materials in concrete, which is a waste of a production process and creates environmental and storage problems, continues to be investigated [5]. In addition to the fact that the materials to be used do not adversely affect the fresh state properties, mechanical properties and durability performance of the concrete, the fact that it provides benefits for its performance

increases its preferability [6]. In addition, the increase in energy demand with the developing technology and the construction of nuclear power plants for this purpose, the high strength and durability performance of concrete

mixtures, as well as radiation absorption and nuclear waste storage expectations are increasing. In this context, boron ore has recently gained an increasing momentum among the materials that can be used for the improvement of some properties of concrete and radiation absorption.

Boron is used in various areas of industry such as agriculture, cleaning, ceramics, glass, insulation, health and energy. Turkey has the largest boron deposits worldwide with a share of 72% (851 Mt) in terms of B_2O_3 content [7]. During the processing of boron in industrial areas, about 400000 tons/year of different types of solid boron-containing wastes are generated [8]. Excessive amounts of waste cause serious environmental and costly storage problems [9]. The B_2O_3 content in these wastes usually ranges from 11% to 25% by weight, depending on the starting minerals and processing conditions. Turkey's commercial boron ores are colemanite, tincal and ulexite. After colemanite is extracted and concentrated in the concentrate plant, it is reacted with sulfuric acid to obtain boric acid. Concentration plant waste is called colemanite concentrator waste and boric acid plant waste is called borogypsum. Colemanite concentrator waste and borogypsum contain approximately 6.3% and 4.2% B_2O_3, respectively. It was declared that the amount of CO_2 released during the heat treatment of boron-derived wastes is lower than in cement production. The loss on ignition value of boron-derived wastes below 1000°C obtained by XRF analysis is about 30% by weight due to the removal of both H_2O and CO_2, while a higher amount of CO_2 is released (the cement content is 57.5% by weight on average) during conventional cement production [10]

Kök et al. [11] stated that boron wastes provide protection against gamma rays and thermal neutrons in concrete mixtures. In addition to this feature, it was stated that geopolymerization accelerated and compressive strength increased by adding geopolymer to concrete mixtures for the immobilization of nuclear boron wastes [12]. In this context, recent studies on the effects of boron use on the radiation absorption, fresh state properties, compressive strength, freeze-thaw and high temperature resistance of concrete are presented in this study.

Radiation absorption properties

The photon attenuation coefficient is an important parameter for characterizing the penetration and diffusion of X- and gamma-rays in multi-element materials. Photon attenuation coefficients are essential in a variety of nuclear science, technology and medical applications. The mass attenuation coefficient, l/q, is a measure of the average number of interactions between the photons and the matter occurring per unit area thickness of the substance under investigation [13]. The fact that radiation can be harmful has led to the development of a wide variety of shields to protect against it [13]. A larger amount of shielding material is required for nuclear radiation shielding. Therefore, examining the radiation flux propagation in shielding materials is a basic requirement for shield design [13]. Good shielding should have a high absorption cross section for radiation and at the same time, the irradiation effects on its mechanical and optical properties should be small. The mass attenuation coefficients depend on the chemical composition of the absorbing material and the energy of the gamma rays. In this context, the use of materials such as waste boron in nuclear shield concrete mixtures should be investigated. The radiation permeability of the concrete produced by using boroplaster and colemanite concentrator wastes for 59.54 and 80.99 keV gamma rays was measured and it was investigated whether adding these boron-containing wastes to cement as a protective material would be beneficial. It was emphasized that both boron waste types are effective in preventing radiation transmission. As a result, it was reported that boron wastes add gamma ray shielding properties to the cementitious mixture [13]. Many studies were conducted on the radiation shielding properties of concretes containing colemanite and ulexite as aggregate additives. In the study carried out by Oto and Gür [14], the mass attenuation coefficients of concrete containing ulexite and ulexite concentrator wastes were examined, and it was declared that both materials could be used for gamma ray protection at concentrations below 10%. Binici et al. [15], the linear absorption coefficient increased from 0.07 cm^{-1} to 0.24 cm^{-1} by adding 10% waste barite to the control mixture. By adding 0.25% and 0.75% colemanite to the control mixture, the said value was measured as 0.08 cm^{-1} and 0.05 cm^{-1}. As a result, it is seen that barite wastes are effective in preventing radiation transmission, the said effect is low for colemanite and these materials are promising materials for gamma ray shielding in mortars. It was observed that the linear absorption coefficient increased from 0.24 cm^{-1} to 0.69 cm^{-1} with the increase in barite usage rate from 2.5% to 10%. It was emphasized that high amounts of barite (90%) containing $BaSO_4$ are effective in shielding gamma rays.

Effect on fresh state properties

The use of boron-derived materials in cementitious systems significantly affects the fresh state properties of the mixtures. The increase in boron waste replacement ratio from 1% to 7% in mortar mixtures containing boron waste adversely affected the fresh state properties of the mixtures and increased the need for water reducing admixtures for adequate workability [16,17]. It was stated that the use of boron waste generally causes an increase in rheological parameters (viscosity and shear stress). It was reported that the mixture containing 1.0% boron waste behaves similarly to the reference mixture in terms of some rheological parameters, but with the said ratio increasing to 1.5, 2.0, 2.5%, the static and dynamic shear stress and plastic viscosity values together

exceed the reference mixture values over time [18]. In addition, the increase in colemanite waste content in mortar mixtures negatively affected the time-dependent slump flow performance. Similar to the slump-spreading behavior in terms of rheological properties, an increase was observed in all three of the viscosity, static and dynamic shear stress values with the use of waste boron. It was declared that this increase has become more evident especially with the use of waste boron at high rates. Similarly, in another study, the slump values decreased from 220 mm to 200 mm when the water/binder ratio was 0.6, and from 40 mm to 8 mm when the water/binder ratio was 0.5, with 15% waste boron replacement in the control mixture [19]. It was observed that the effect of boron on fresh state properties is greater in mixtures with low water/binder ratio. It was reported that this negative effect is due to the high loss on ignition of the waste boron and the clay minerals in it and to increase the friction between the cement grains [17].

Erdoğmus [20] and Gencel et al. [21] suggested that the setting times increase with the increase in the amount of waste boron and the effect of the B_2O_3 component in cementitious systems. Borates such as soluble boric acid react with the calcium compounds of Portland cement and cause the formation of insoluble calcium borate compounds such as $2CaO{\cdot}3B_2O_3{\cdot}8H_2O$ and $CaO{\cdot}B_2O_3{\cdot}6H_2O$ under high pH conditions [12]. These calcium borate compounds accumulate on the surface of cement particles and slow down or prevent hydration reactions [16]. In this context, the setting of portland cement is delayed and its early age mechanical properties are weakened [22].

Shen et al. [23], it was reported that borax significantly delays the early hydration and setting time of calcium sulfoaluminate (CSA) cement containing different proportions of gypsum. It was reported that the use of an appropriate amount of borax as a retarder is beneficial for the development of advanced age strength of CSA cements. While the early strength of borax mortars decreased in mixtures containing 5% gypsum, improvements were observed in the mechanical properties of mixtures containing 15% and 20% gypsum. It was emphasized that this is due to the fact that borax prevents the hydration of ye'elimit and the depletion of plaster in 1 day. It was stated that the effect of borax on ettringite formation is related to the gypsum content of CSA cements.

It was stated that the use of boric acid and borate is a solution to the problem of magnesium phosphate cement reacting too quickly. Borax is the most commonly used set retarder and its retarding effect is related to the ratio of boron to magnesium [24]. For example, it was stated that 5% and 10% borax magnesium phosphate cement pastes reach their final setting temperature in 18 minutes and 22 minutes, respectively [25]. It was found that boron-containing compounds can effectively control the reaction rate of magnesium phosphate cement [26]. Magnesium phosphate cement material has good interfacial bonding properties. With the application of borax, the reaction rate of magnesium phosphate cement with extremely high early strength becomes controllable. Thus, the possibility of emergency repair and reinforcement of magnesium phosphate cement for roads, airport runways, bridges and military repair materials was also investigated [27].

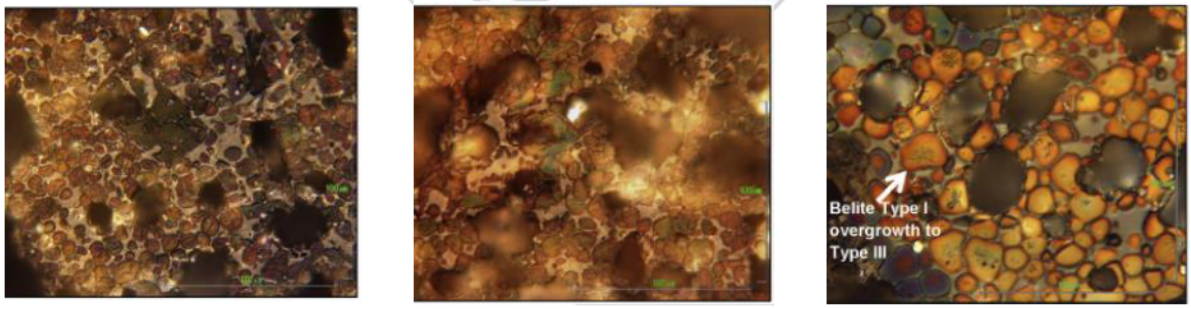
Effect on strength properties

Boron waste materials affect the setting times of cementitious systems as well as the mechanical properties of early and advanced ages. Final compressive strength in cementitious systems containing waste boron is also relatively low (2 ± 0.3 MPa) due to the delay of setting of calcium borate compounds. Fabian et al. [28], the use of cementitious mixtures for the disposal of nuclear waste was investigated. As a result of the study, it was emphasized that the leakage of nuclear wastes is less with the increase in the compressive strength of concrete mixtures. In addition, it was stated that the 28-day compressive strength value decreased from 42.57 MPa to 37.51 MPa with the increase of boron waste amount (boric acid) from 50 g/l to 250 g/l. It was declared that the use of geopolymer waste forms is more beneficial than cement waste forms for the immobilization of radioactive wastes [29]. In general, geopolymers exhibited higher compressive strength than cement, despite shorter curing days with the same waste load [30]. According to the Japan Atomic Energy Agency, metakaolin-based geopolymers are superior to portland cement for immobilization of sewage sludge ash containing 137Cs [29]. It was determined that the amount of radioactive cesium (<0.1%) leached in geopolymers is approximately 300 times lower than that in cement [29]. Kim et al. [12], a phosphate-based geopolymer waste storage mixture was investigated for the immobilization of radioactive boron waste. All geopolymers met the landfill acceptance criteria (3.45 MPa), despite the short setting time. It was declared that curing at high temperatures (at 60 and 90°C) compared to room temperature increases the 7-day compressive strength of geopolymers and the maximum compressive strength is obtained in a geopolymer containing 40% by weight waste boron. XRD, MAS NMR and SEM analysis results of the geopolymer with the highest compressive strength and the reaction product prepared under high temperature curing conditions showed that the crystalline phases in boron waste were transformed into amorphous boron phosphate phase and 4-coordinated boron was included in the geopolymer structure. Therefore, high temperature curing conditions lead to direct precipitation of the amorphous boron phosphate phase and accelerated geopolymerization, resulting in higher compressive strength as the boron waste content increases. These results show that phosphate-based geopolymers are promising waste

form candidates for the immobilization of waste boron and other radioactive wastes, which are difficult to immobilize using conventional cement or alkali-activated materials. However, it was observed that the 7-day compressive strength of geopolymers decreased as the amount of waste boron in the mixture increased. It was assumed that the consumption of an acidic activator by the boron phosphate reaction results in low compressive strength.

The effect of the use of waste boron materials obtained from the concentrated borax unit (CW1) and borax pentahydrate unit (CW2) in mortars on the strength was investigated by Özdemir et al. [31]. These waste borates were used in mortar mixtures with 1, 2.5, 5 and 10% cement substitutes. Mortar containing 1% CW1 has a higher 28-day compressive strength than the control mixture. However, the mechanical properties of the mortar were adversely affected as the use of waste boron and the rate of B_2O_3 in waste boron increased. Mortar mixes containing up to 5% CW1 (B_2O_3=22.48%) and 10% CW2 (B_2O_3=8.23%) met TS24 (1985) standard minimum compressive strength requirements (32.5 MPa). As a result, it was suggested that these wastes can be replaced by cement up to 5% and 10%, respectively. Similar results were found by Gencel et al. [21] has also been observed. Durgun et al. [17], the 28-day compressive strength of all samples containing 1, 3, 5 and 7% waste boron was lower than the control sample. While the difference between the control sample and the waste boron added samples was closed in the 56-day samples, the sample containing 5% waste boron had a higher compressive strength than the control sample. In another study, it was observed that there was a decrease in early strength in mortar mixtures with boron waste substituted for natural gypsum compared to the control sample. However, setting times were not significantly affected [32].

The use of boron wastes in different types of cementitious systems was studied by various researchers. In a study, the incorporation of boric acid and boron oxide from boron waste into raw meal for the production of belite cement and the effect of boron as a part of clinker on belite crystals were investigated. Three types of clinker were prepared by adding 1.5% by weight boric acid (BC_BA1.5) and 1.0% (BC_BW1) and 6.5% (BC_BW6.5) boron waste in the whole mixture. According to the development of free lime content and microstructure of the clinkers, firing was carried out at 1330°C, 1350°C and 1310°C for BC_BA1.5, BC_BW1 and BC_BW6.5, respectively. In Figure 1, optical microscope images of clinkers formed at 1320°C firing temperature are given. Boron addition helped stabilize the upper belite polymorphs as well as lowering the clinkerization temperature. As a result of the study, with the addition of 1% and 1.5% boron, the 28-day compressive strength values were declared to be 60 MPa and 52.7 MPa, respectively. It was emphasized that the increased molten phase caused the deterioration of hydraulic properties due to the formation of higher amounts of C_5BS in the clinker. The 28-day compressive strength varied depending on the type of belite crystals in the clinker rather than the content. The results show that although the increase in the use of boron oxide in certain cements affects the strengths somewhat negatively, it may be beneficial in terms of lowering the firing temperature of the cement in the kiln [7].



(a) (b) (c)

Figure 1. Optical microscope images of clinker formed at 1320°C firing temperature: a) BC_BA1.5, b) BC_BW1 c) BC_BW6.5 [7]

In some studies, boron wastes were calcined at high temperature and used to improve their properties. In a study, ground heat-treated (1 hour 800 degrees 10°C/min) boron-derived wastes with different particle sizes (larger, closer and smaller median particles) than Portland cement were used instead of Portland cement at a rate of 25% by weight. In addition, the compositions and microstructures of the hydration products were found to be similar for all mortars. However, it was stated that the strength of 7- and 28-day cement mortars does not always increase with the reduction of particle size of ground heat-treated boron wastes. Compared to cement, coarser ground heat-treated boron wastes resulted in a high strength loss as expected (42.5% reduction). However, the smaller median particle size of ground heat-treated boron wastes reduced the strength of the mortars due to the waste's non-reactivity and lower packing density. Ground heat-treated boron tailings with a median particle size

comparable to that of cement reduced the strength to only 6.3% of that of the reference mortar. It was associated with a higher particle size range of ground heat-treated boron wastes, which provided the same packing density as the reference mortar due to the filling effect. Environmentally friendly cement mortars can be produced using boron wastes, which cause serious environmental problems in terms of compressive strength and costly storage problems, by replacing 25% by weight of Portland cement with ground heat-treated boron wastes. In summary, not only the median particle size of cement substitutes, but also the particle size range plays a critical role in the filler density and therefore the strength of cement mortars. Therefore, the use of non-reactive fillers with a slightly wider range of particle size may be beneficial for other studies focused on increasing the strength of reinforced concrete mortars. Data obtained from microstructural analysis and compression test showed that ground heat treated boron wastes can be replaced with cement at a rate of 25% by weight for 1 hour at 800°C [10].

In addition, recently, it has become widespread to investigate the usability of boron wastes in alkali-activated geopolymer systems. In the study conducted by Yön and Karataş [33], self-compacting alkali-activated mortar samples were produced using 0-25% boron waste and granulated blast furnace slag. The samples were cured at 60 and 100°C for 3 days. As a result, it was determined that the mechanical and durability properties of the samples decreased with the high use of boron waste in the mixtures. According to this study, it was suggested that up to 10% boron waste can be used in self-compacting alkali-activated mortar in terms of both mechanical and durability performances. In particular, it showed that the addition of waste boron caused greater sensitivity to the curing temperature at 100°C in compressive strength compared to 60°C. It can be said that the main reasons for low compressive strength for waste boron samples are increased water demand for waste boron content, low CaO: SiO_2 and SiO_2: Al_2O_3 and slowing of binder hydration. The presence of calcium aids in curing by improving CSH and CASH and coexists with NASH in the geopolymer matrix [34]. Also, the increase in curing temperature (from 60°C to 100°C) affects the C–S–H gel, which causes the self-compacting alkaline active mortar to be porous with many cracks [35]. Therefore, high amount of calcium activated binder in NH-NS showed lower strength at 100°C [33]. Similarly, in the study examining the effect of waste boron replacement ratio (0, 10, 20, 30, 40%) on the compressive strength of alkali-activated mortars, it was declared that the mixture containing 90% metakaolin and 10% waste boron showed the best performance [36].

Effect on freeze-thaw resistance

The effect of boron wastes on the freeze-thaw resistance of cementitious systems was investigated. In the study carried out by Erdoğmuş [20], the effect of samples containing 6% silica fume and 2-12% colemanite on the dissolution resistance was investigated. While the strength loss was 14.9% in the mixture without colemanite waste after freeze-thaw cycles, it was reported that this ratio was 14.5-28.3%, respectively, in mixtures containing 2-8% colemanite. For 10% and 12% ratios, it was stated that the said ratio exceeded the 30% strength loss limit specified in ASTM C666 with 39.4% and 53.3%. Similar results were also observed in the study conducted by Topçu and Boğa [37]. It was stated by the researchers that the decrease in the flexural strength of the mortar sample containing 3% and 10% waste colemanite after freeze-thaw was 10% and 20%, respectively.

The effect of using boron wastes (0-25%) in granulated blast furnace slag and self-compacting alkali activated mortar on permeability and freeze-thaw resistance was investigated. It was reported that with the increase in boron waste utilization rate (5, 10, 15%), the sorptivity coefficient and porosity increased, the pores became more independent, and during the freeze-thaw cycles, the external liquid entered the pores of the mortar more easily, causing the sample to disperse [33]. Similar results were observed by Erdoğmus [20]. In a study examining the 1-year durability performance of alkali-activated mortars containing 90% metakaolin and 10% boron waste, it was declared that the freeze-thaw resistance was appropriate [38]. It was stated by the researchers that the main reason for this is that the compact structure of the matrix is resistant to freezing and thawing effects and provides a good degree of adhesion.

Effect on high temperature resistance

The strength of concrete after exposure to high temperatures is important in terms of durability performance. It was declared that the high temperature resistance is not adversely affected by the use of waste boron [36]. It was declared that the strength preservation performance of concrete mixtures containing 1, 3 and 5% colemanite wastes is better after being exposed to 200-1000°C [39]. In addition, it was declared that the cement mixture on which the pumice aggregate in lightweight concrete mixtures is coated contains colemanite and that the high temperature resistance of lightweight concrete samples improves with the increase of this content [40]. With the increase in temperature, the enlargement of the sample pore structure and the water evaporation in the matrices increase. With the loss of mass, additional voids are formed. These additional gaps are the reason for the decrease in ultrasonic pulse velocity results. Dehydration and moisture loss in samples produced at high temperatures lead to weight loss and microstructure damage. The colemanite substitution reduced the weight loss

after high temperatures. According to the results, up to 10% substitution of colemanite resulted in an improvement in strength properties and ultrasonic transmission rate test results, and this amount of improvement decreased at higher substitution rates [41].

CONCLUSION

The following results were obtained when the use of boron waste in cementitious systems and the effects on fresh state, mechanical properties and durability performance were examined together.

- It was understood that boron wastes are promising materials in terms of benefiting the properties of cementitious systems by preventing radiation transmission and shielding gamma rays.
- The water requirement and setting time of cementitious systems increased with the use of boron waste and the increase in its usage rate.
- When the use of boron waste exceeds the order of 10%, their compressive strength generally decreases. In this context, it was understood that more suitable results can be obtained by using the waste when calcining at high temperatures.
- It was understood that the freeze-thaw resistance decreases as a result of the increase in the porosity of the cementitious systems with the use of boron.
- It was declared that boron waste improves the resistance of cementitious systems against high temperatures.

Data Availability Statement

All data, models, and code generated or used during the study appear in the submitted article.

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Conflicts of Interest/Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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