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TECHNO-ECONOMIC ANALYSIS OF ZINC BORATE PRODUCTION FROM ZINC OXIDE AND BORIC ACID

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Abstract: The synthesis of zinc borate, 2ZnO3B₂O₃3.5H₂O, was investigated via reaction of solid zinc oxide with boric acid aqueous phase in the existence on seed crystals. Commercially important type of zinc borate was synthesized using the 4.7 M boric acid concentration and B2O3 to ZnO molar ratio of 2 at 90°C, at 900 rpm and for 4 hours reaction time in a stainless-steel reactor. The amount of seed crystals was 1.5% of the boric acid initially used. At the end of each experiment, X-ray diffraction (XRD) instrument as well as Fourier-Transform infrared spectroscopy (FTIR) instrument were carried out for identifying final products. A continuous process was developed with SuperPro Designer software. The design was derived from experimental and literature data. A technoeconomic analysis of the zinc borate synthesis process was carried out for different annual plant capacities (6,000-12,000t). Unit prices of raw materials and final product were obtained from the literature and the market as US\$600/ton boric acid, US\$2,200/ton zinc oxide and US\$4,500/ton zinc borate. The fixed investment cost of the zinc borate production plant with capacity of 10,000 ton/year was found to be US\$22,780,000 and the operating cost was found to be US\$26,273,000/yr. The payback time was determined as 1.93 years from the economic analysis of the process.

Keywords: Zinc borate, Economic analysis, SuperPro designer, Process development, Production cost

1. Introduction

Zinc borates are important borate salts that can be produced from the reaction of boron compounds e.g., borax, boric acid and zinc compounds e.g. zinc oxide and zinc sulfate (Cui et al., 2012). Although there have been a number of zinc borates synthesized in the literature, $2ZnO·3B₂O₃·3.5H₂O$ is commercially important borate compound produced in large quantities in the world (Roskill, 2010). Although the zinc borates of $2ZnO·3B_2O_3·3H_2O$ and $2ZnO·3B_2O_3·3.5H_2O$ are same compounds, there has been a controversial case in naming of those compounds in commerce until the appearance of Schubert's article which has proven that zinc borate has oxide chemical formula and closed formula as following $2ZnO·3B₂O₃·3H₂O$, $Zn[B₃O₄(OH)₃$, respectively (Schubert et al., 2003). Zinc borate of $2ZnO·3B₂O₃·3.5H₂O$ is going to be called as just zinc borate hereafter in this manuscript. It is widely used in the production of fire-resistant products e.g., cables, fabrics, electronic parts, paints in an automobiles and aircrafts (Mergen et al., 2012). Those products are mainly manufactured using polymers, such as nylon, PVC, polyester, and ABS (Engin, 2009; Ting et al., 2008). A fireretardant additive should be thermally stable during the processing of the polymers in the extruder. Zinc borate of $2ZnO·3B₂O₃·3.5H₂O$ is most frequently used as a polymer

additive due to its comparatively higher dehydration start at 290°C (Çakal et al., 2020). The ability of zinc borate to retain dehydration water up to 290°C enables its use in a high temperature polymer processing (Schubert, 2003). Since it does not have toxic properties during thermal processing, special methods are not required during its addition to polymers. The extrusion temperature varies depending on the percent of zinc borate used, the type of polymer and other additives (Baltacı, 2010). Since zinc borate is used as a flame retardant, it reduces the flammability properties of the polymer and increases its decomposition resistance (Bardakçı, 2009). Therefore, polymers containing zinc borate could be processed at higher extrusion temperatures up to 290°C.

Zinc borate is produced by arranging the molar ratio of ZnO to B_2O_3 and reaction conditions e.g., reaction temperature and time, boric acid concentration and stirring rate (Eltepe et al., 2007). The boric acid mixture reacts with solid zinc oxides at 90°C and atmospheric pressure. Sawada and his colleagues produced zinc borate in a two-step process: in the first stage, solid boric acid and zinc oxide were added into an aqueous boric acid solution at 60° C and were mixed during 1.5 hours. Then, that mixture was then agitated for a further 4 hours at 90°C. In their experiment, Sawada and his coworkers used zinc borate as seed crystals. The amount

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of the seed crystals is 1.5 wt. % of the boric acid initially used. The characterization study of the produced materials was carried out by XRD. It was determined that 2ZnO. 3B2O3. 3H2O zinc borates were obtained in the experiments using seed crystal (Sawada et al., 2004). An 85L reactor was utilized to examine the large scale at which zinc borate can be manufactured through the reacting zinc oxide with a boric acid solution at 85°C (Kılınç et al., 2010). Particle size is one of the most important properties of materials used as additives. Therefore, Shete et al. investigated the particle size of the final product and the conversion parameters of zinc oxide affecting the formation of zinc borate in the synthesis of zinc borate using zinc oxide and boric acid starting materials. In the experiments; agitation speed, impeller type, particle size of zinc oxide, temperature, initial boric acid concentration were studied to determine the effects of mixing. From the experimental results, it was seen that the mixing parameters influenced the particle size of zinc borate. It was determined that with the increase in the average initial size of zinc oxide, the time taken for the conversion of zinc oxide to zinc borate increased. The increase in reaction time with the increase in particle size proved that the reaction is a surface controlled reaction. (Shete et al., 2004).

The widespread use of zinc borate as a flame retardant in recent years is one of the factors encouraging the increase in the production capacity of zinc borate. Due to the increasing demand for zinc borate use and changing market conditions, it is necessary to increase the number of zinc borate production facilities and the capacity of existing production facilities. There are several companies which produce zinc borate in thousands of metric tons for more than four decades in the world. Although Türkiye has the highest boron minerals, the most of the boron products, such as zinc borates were not produced commercially until the announcement of Akdeniz-Chemson Inc. which launched its zinc borate plant with the capacity of 3,000 t/y in 2021 (Rais, 2021). Process modelling, using data from laboratory experiments and from the markets, can usually improve performance of the process, purity of the product and identify effective inputs for the process (Kwan et al., 2018). A sustainable synthesis of boric acid by reacting the solid colemanite with sulphuric acid was examined using the SuperPro Designer program, taking into account the cost of treating the waste and processing the raw material. The evaluation of zinc borate $(2\text{Zn}0.3\text{B}_20_3.3.5\text{H}_20)$ synthesis from zinc oxide reaction with boric acid was not examined considering raw material availability and transport advantages in Türkiye (Gönen et al., 2022).

A continuous process was selected for zinc borate production and its flowsheet having reactor and other unit operations (filtration, drying and milling) was designed in this program. Reactions taking place in the reactor, parameters of separation units, raw material and product properties obtained from laboratory experiment and from the literature (Eltepe et al., 2007; Sawada et al., 2004; Gönen, 2009) were used in the SuperPro Designer program. The objective of this investigation in this study was to develop a process for zinc borate (2ZnO. 3B2O3. 3.5H2O) reaction which was based upon between zinc oxide with boric acid. SuperPro Designer Program (version 9.0) was applied to conduct the technical and economic analysis of this process.

2. Materials and Methods

2.1. Materials Used in the Experiments

Zinc oxide (99.0%, wt.) obtained from Merck Inc. and boric acid (99.92%, wt.) obtained from Eti Mine Inc. were used in zinc borate production. Commercial zinc borate $(22n0.3B_2O_3.3.5H_2O)$ obtained from Eti Mine Inc. was utilized as seed crystals in reaction. EDTA, NaOH, NH4Cl, Phenolphthalein, Methyl Orange, HCl (37%, Merck) and Mannitol from Sigma-Aldrich were used in the titration of the final product. Deionized water was used in all experiments and analyses.

2.2. Zinc Borate (2ZnO. 3B2O3. 3.5H2O) Production

In this study, zinc borate was synthesised through a reacting zinc oxide with boric acid as described in Equation 1 in a stainless steel reactor with an electromagnetic mixer at the laboratory scala.

$$
2ZnO_{(s)} + 6H_3BO_{3(aq)} \rightarrow 2ZnO \cdot 3B_2O_3 \cdot 3.5H_2O_{(s)} + 5.5H_2O_{(l)} \tag{1}
$$

Design of the experiment consisted of a stainless steel reactor with a magnetic drive unit, surrounding aluminum heater unit, a split-ring valve, thermostate, manometer, needle valve and rupture disc mounted on the reactor cover. A four bladed, turbo impeller was fitted to the reactor.

The experiments were carried out in a stainless steel reactor which was run at 90 °C during 4 hours and molar ratio of B_2O_3 to ZnO equal to is 2.0, a boric acid concentration that is 4.7 M (Gönen, 2009) and a stirring rate of 900 rpm. Initially, 50 ml of $H₂O$, a stoichiometric amount that is boric acid (H_3BO_3) as well as zinc oxide (ZnO) were added to reactor. After closing the reactor lid, the mechanical stirrer was started and a temperature controller was adjusted to the desired reaction temperature 90 °C. A second experiment was done with same condition but zinc borate amount of 1.5% boric acid used was added as seed crystal.

Boric acid was used in excess to consume all the zinc oxide used in the reaction. Zinc oxide is not soluble in an aqueous phase. Therefore, it was difficult to separate the unreacted zinc oxide from the solid product (zinc borate). The precipitated solid products were separated by vacuum filtration. Then, it was washed deionized water to free the unreacted boric acid from the final reaction solution when the reaction was completed. The boric acid content of the maternal solution and the Zn, B₂O₃ content of the synthesized solid samples were evaluated by analytical titration at the end of each experiment. An oven at 105 °C was used to dry the wet cake obtained from the filtration for five hours. X-ray diffractometer (Bruker D8 Advance Twin-Twin) was used to analyze the crystal structures of the formed powders with CuKα radiation at 45 kV and 40 mA. The absorption spectra of KBr pellets prepared by mixing 2.0 mg of sample and 100 mg of KBr in an agate mortar and pressing the mixture under certain pressure were obtained using Fourier transform infrared (FTIR) spectrophotometer (Jasco FT/IR 4700).

3. Results

3.1. Product Characterization

An overconcentration of boric acid was utilised to synthesise zinc borate, because boric acid is soluble in an aqueous phase (Gönen et al., 2011). In these experiments, the boric acid concentration was first adjusted to 4.7 M. Then, the quantity of zinc oxide which corresponds to a molar ratio of B_2O_3 to ZnO equal to 2.0 was added into the reactor. Experiments were carried out in the absence and in the presence of a seed crystals to synthesise zinc borate. Under these experimental conditions, FTIR spectra of final solid powders are shown in Figure 1 and XRD patterns are shown in Figure 2.

Figure 1. FTIR spectrum of (a) zinc borate sample synthesized at 90°C for 4 h without seed crystals (b) zinc borate sample synthesized at 90°C for 4 h including seed crystals.

The characterization study of the synthesised zinc borate powders was performed via Fourier Transform Infrared (FTIR) spectroscopy. Both spectra in Figure 1 were obtained in the 4000-400 cm-1 wavenumbers and are completely different each other. The "O-H" groups present in the structural of zinc borate with the formulae 2ZnO. 3B2O3. 7H2O are represented by the broadening peak at 3600 cm^{-1} to 2800 cm^{-1} in the Figure 1.a. The spectrum peak placed into 1370 cm-1 indicates asymmetric stretching vibrations of the BO3 structure. Both peaks at 1053 and 997 cm-1 wavenumbers indicate the asymmetric stretching vibration of BO4 bonding (Gönen, 2009). The symmetrical stretch motions of the BO3 and BO4 units were differentiated from the peak at 907 and 822 cm-1 wavenumbers respectively (Jun et al., 1995). It was concluded that reaction boric acid with zinc oxide in the absense of a seeding crystals produced zinc

borate of 2ZnO. 3B2O3. 7H2O at 90°C over a reaction time 4 hours. FTIR spectrum of zinc borate sample synthesized using seed crystals is shown in Figure 1.b. Both strong peaks at wavenumbers 3438 and 3187 cm-1 belong to the vibration of O-H groups, which is the fingerprint associated with zinc borate having formula 2Zn 0.3B_2 $0_3.3.5\text{H}_2$ 0 (Eltepe et al, 2007). The peaks at these wavelengths can be seen in Figure 1.b. In addition, of these peaks in Fig.1.b, the one at 3200 cm wavelength is the characteristic peak belonging to the ZnO. B203 group (Eltepe, 2004; Gönen, 2009). The peaks at wavenumbers of 1363 cm-1 and 1312 cm-1 represent asymmetric stretching vibration of $BO₃$ groups, and the peaks at wavenumbers of 1175 and 1059 cm-1 represent asymmetric stretching vibration of BO4 groups. Others at 974 and 745 $cm⁻¹$ correspond to symmetrical BO₃ and BO4 (Jun et al., 1995). When FTIR spectra shown in Fig. 1.a and Fig.1.b are compared it is seen that they have different chemical structures. The desired zinc borate has the chemical structure $22n0.3B_2O_3.3.5H_2O$ and was synthesized at 90°C and 4h reaction time using seed crystals.

Figure 2.a and Figure 2.b show XRD patterns obtained from zinc borate samples prepared in the absence and presence of seed crystals at 90°C and 4h reaction time, respectfully. The principal peaks in the X-ray diffraction diagram for the powdered zinc borate sample shown in Figure 2.a. are at 10.59°, 12.95°, 20.83°, 21.20°, 28.10°, 30.10°, 31.0° and 33.25° 2θ. On the basis of the correspondence of these main peaks, the solid product obtained in the absence of seed crystals could be zinc borate whose chemical formula is 2ZnO∙3B2O3∙2H2O (Gönen, 2009). However, the main signals in the XRD pattern from the zinc borate sample shown in Fig. 2.a. has peaks at 18.0°, 20.60°, 21.7°, 22.5°, 23.7°, 24.1°, 27.5° and 28.7° 2θ values By looking at the XRD pattern in Fig. 2.a., it can be said that the synthesized zinc borate 4h reaction time without seed crystals has a different chemical structure from the zinc borate composed by 2ZnO∙3B2O3∙3.5H2O. Referring to the XRD diagram of zinc borate recorded by Sawada and co-workers and JCPDS 32-1464 database, the zinc borate which was gained in 4.7 molar boric acid concentration in the absence of seed crystals is zinc borate with a type 2ZnO∙3B2O3∙3.5H2O (Sawada et al., 2004).

Figure 2. XRD patterns acquired from (a) zinc borate sample synthesised at 90°C for 4 hours without seed crystals (b) zinc borate sample synthesised at 90°C for 4 hours with seed crystals.

3.2. Techno-Economic Analysis of Zinc Borate Production Process

3.2.1. Process Description

The zinc borate production process was designed in SuperPro Designer program (Version 9.0) as shown in Fig. 3. The process consists of blending tank, reactor, filtration unit, washing unit, drying unit, milling and powder storage tank. The process starts with the preparation of a boric acid solution in the blending tank, where raw boric acid and water are mixed together with a recycle stream from the filtration unit. To obtain the desired zinc borate, 2ZnO. 3B2O3. 3.5H2O, the boric acid concentration was then precisely adjusted to the specific one required. Filtrate from washing unit was directly recycled to the reactor. Zinc borate cake from the belt filter is fed to the fluidized bed dryer where hot air at 105°C is used to remove the water from the zinc borate less than 0.5% by wt. Finally, dried zinc borate is stored in a silo for being packed.

The chemicals used in the zinc borate production plant, the reactions in the reactor and the conversion value were entered into the SuperPro Designer program as data. The units used in the process were determined and the units were connected to each other with pipe connections. The parameters of the units (temperature, reaction time, solid/liquid ratio) were defined in the program. In addition, labour costs, costs of raw materials, unit prices of the final products produced and the utilities used were defined to the program by making use of market conditions. Here, the values in the database of the program were used for the prices of some utilities (such as air price, waste treatment cost, steam cost and operator price).

The properties and unit prices of the chemicals and final products used in the production process are shown in Table 1. These data were obtained from the market. In the SuperPro Designer program all prices were entered in US\$. For this reason, all costs and outputs in the project are given in US\$.

Since the process is designed in a continuous mode, two reactors were operated in staggered mode to achieve the required reaction time of four hours. The 2023 data was used to calculate the economic assessment variables for the whole project. The building of the facility starts in 2023 and lasts for 15 months. The commissioning period was set at 4 months and the project lifetime was set at 15 years. An inflation rate of 4% and an annual interest rate of 7 % were used in the economic analyses.

The economic The total capital investment was determined by summing total direct cost (TDC), total indirect cost (TIC), contractor's fee & contingency (CFC), working performance of the zinc borate production process was investigated through evaluation of total capital investment, operating expenses and income generation. The total capital investment was determined by summing total direct cost (TDC), total indirect cost (TIC), contractor's fee & contingency (CFC), working capital and startup costs cost as presented in Table 2. To calculate the total capital investment and operating costs, the factors determined by SuperPro Designer were multiplied by the purchase cost (PC) of the main equipment in the plant. The total direct cost (TDC) includes the construction of the facility, comprising the purchase of equipment (PC), installation of equipment, process piping and other related costs. Total indirect cost (TIC) comprises engineering and supervising and constructing costs.

Equipment size and the number of equipment needed for each capacity have been computed from mass and energy balances for the zinc borate synthesis reaction. The increase in the capacity of the zinc borate plant has a significant effect on the number of equipment used in the process. As a function of capacity, the purchase price of the required process equipment has been on the rise. The costs of these items are estimated by multiplying the equipment purchase cost by the coefficients indicated in Table 2. e.g. process piping is seen as 0.35 times the cost of the equipment.

Annual operating costs are computed by summing total variant production costs, fixed costs, factory overheads and general expenses, and details of the production cost parameters are given in Table 3. As the plant is assumed to be very close as far as raw materials and markets are concerned, logistics costs have not been included in the operating cost calculations. Data from the integrated module in SuperPro Designer Program and from the literature were used for the mass and energy balance calculations (Peters et al., 2003). The annual operating cost for the zinc borate production process was calculated based on raw materials, labor, facilities, laboratory and utilities.

Table 2. Total capital investment (US\$M) required for a zinc borate plant

Table 3. Parameters for annual operating cost

3.2.2. Techno-Economic Analysis (TEA)

Zinc borate production flowsheet was designed in SuperPro Designer program as shown in Figure 3. The operating mode was set to be 7,920 operating hours per year for production of zinc borate. The data obtained from experimental study (reaction time, solid liquid ratio,

boric acid concentration, solubility and etc.) and from the literature (molecular weights, heat of reactions, heat capacities and etc.) were entered into the program. As the boric acid was used in excess to reach higher conversion values, the amount of boric acid separated from the product was recycled to the reactor.

Figure 3. Flowsheet of Zinc Borate Production Process

A percentage of the recycle stream is purified to prevent the accumulated boric acid and to remove the water formed in the reaction and that used in the filtration unit. SuperPro Designer 9.0 software was used for mass and energy balance calculations.

The economic performance for the zinc borate plant was investigated by determining the total capital cost, operating cost and revenue generation for different production capacities ranging from 6,000 t/y to 12,000 t/y. Table 2 summarizes the total investment for a zinc borate process calculated by SuperPro Designer program. As shown in Figure 4, the total capital investment of the zinc borate production process varies between US\$21.4 and US\$26.77 M depending on the capacity of the production plant (6,000-12,000 t/yr). It can be seen from Figure 4, total capital investment increases as production capacity rises. On the other hand, the production cost per ton decreases as the production capacity increases.

Figure 4. The Impact of Plant Capacity on Total Capital Investment and Production Costs.

Table 4. Payback time and revenues for different plant capacities

Plant capacity	Revenues	Payback time
(t/y)	(US\$M/yr)	(vr)
6,000	27	3.24
8,000	36	2.18
10,000	45	1.93
12,000	54	1.89

The payback time (PBT) and net present value (NPV) equations were used to assess the financial viability of the process (Peters et al., 2003). The payback time is described in equation 2. The payback time has been computed using the revenues for each capacity and presented in Table 4.

Payback time (years) =
$$
\frac{\text{Total Investment}}{\text{Net Profit}}
$$

\n(2)

The payback time was found out to be 3.24 years for the 6,000 t/yr capacity and when the factory capacity is upgraded to 10,000 t/yr, the payback time decreases to 1.93 years. As the capacity of the zinc borate production facility increases, there is no significant change in the payback time. Therefore, a production capacity of 10,000 t/yr was selected to investigate the other parameters listed in the TEA. For an annual production capacity of 10,000 ton zinc borate, the total capital investment was found to be US\$25.86 M. The operating cost was calculated as US\$26.4 M.

The net present value is a measurement of economic viability used to determine whether an investment project is profitable or not. It can be used to make comparisons between the economic viability of different investments. It can be estimated by means of equation 3. In this formula, i is the rate of interest, NCF is the cashflow for year k and N is the lifetime of the project in years.

$$
NPV = \sum_{k=1}^{N} \frac{\text{NCF}}{(1+i)^k}
$$
 (3)

The total direct plant cost for a 10,000 t/yr capacity is approximately US\$12,380,000. The total cost of the plant is US\$19,810,000 when the indirect engineering and construction costs (US\$7,430,000) are added to the direct plant costs. The total of the contractors' fees and the contingency costs is US\$2,970,000. The direct fixed capital cost for this zinc borate production process was calculated to be US\$22,780,000 and is given as Table 1. The SuperPro Designer program database and literature data are used to determine the percentage cost contribution of each equipment for the FCI (Rais, 2021). Annual operating cost of zinc borate production that was calculated by using SuperPro Designer program is presented in Figure 5. The plant producing 10,000 ton of zinc borate has the total annual operating cost of US\$26,273,000. Raw materials, especially zinc oxide, have the largest contribution in the operating cost. Consumables, waste water treatment, transportation, advertising, running royalties, disposal of failed product were excluded in the estimation of annual operating cost as pointed out in Figure 5.

Figure 5. Annual operating cost.

Raw materials are the most important contributor to the cost of producing of zinc borate, accounting for 59%. The cost of zinc borate production for a 10,000 t/yr capacity was calculated as US\$2.63/kg using Superpro Designer 9.0 software and a purchase cost of US\$600/ton of boric acid. The purchase cost of boric acid was identified as an important component of the raw material cost as it is consumed three times more based on stoichiometry. To evaluate the impact of boric acid price on zinc borate manufacturing costs, boric acid prices ranging from US\$600 to US\$3,000 per ton were entered into Superpro Designer 9.0 for a 10,000 t/yr zinc borate production capacity.

Figure 6 shows the impact of boric acid purchase price on zinc borate production cost. As can be seen in Figure 6, as the price of the boric acid mineral used in the zinc borate production process rises, the cost of producing zinc borate also goes up. According to the information obtained from market research, the price of zinc borate varies between US\$2,000 and US\$4,700/ton. If boric acid mineral is purchased at a price of \$2,400/ton and above, the zinc borate production plant cannot make a profit. In order to keep zinc borate production cost between market values, the cost of boric acid mineral should be kept below US\$2,400/ton. The net present value of the project considering 7% interest rate was calculated as US\$68,180,000. Gross profit was calculated as US\$18,747,000/yr deducting annual operating costs from annual revenues. 40% income tax rate was used in the calculation of net profit.

Figure 6. Zinc borate production costs as a function of boric acid price.

4. Conclusion

Using zinc oxide, boric acid and seed crystals, zinc borate was synthesized in this work. XRD as well as FTIR analyses were carried out on the final product obtained at the end of the experiment. The results show that it is possible to produce the desired product zinc borate using seed crystals amount of 1.5 % boric acid initially used and for the reaction time of 4 hours. Zinc borate in the desired form $2Zn0.3B_2O_3.3.5H_2O$ could not be synthesised without using the seed crystals. Production process of zinc borate (2ZnO·3B2O3·3.5H2O) was designed using SuperPro Designer program. The total capital investment (TCI) of the zinc borate production plant was estimated to be in the range of US\$21.4 M and US\$26.77 M depending on the production capacity from 6,000 to 12,000 t/yr. Raw material costs $(59%)$ are the most important factor contributing to Total Capital Investment (TCI). The detailed techno-economic analysis of zinc borate production process was performed for 10,000t annual zinc borate capacity. The unit production cost of zinc borate was calculated as US\$2.63/kg by using SuperPro Designer 9.0. Purchase price of raw materials zinc oxide and boric acid were obtained from market as US\$2.2/kg and US\$0.60/kg, respectively. Selling price of zinc borate was used as US\$4.5/kg in the process. The fixed and operating costs of designed zinc borate process are calculated as US\$22,780,000 and US\$26,273,000/yr. The payback time was determined as 1.93 years from the economic analysis of the process. This is a preliminary study for the zinc borate production. In future, the process will be re-analyzed for different scenarios considering waste water treatment, consumables, transportation, and different price of raw materials.

Author Contributions

The percentages of the authors' contributions are presented below. All authors reviewed and approved the final version of the manuscript.

C=Concept, D= design, S= supervision, DCP= data collection and/or processing, DAI= data analysis and/or interpretation, L= literature search, W= writing, CR= critical review, SR= submission and revision, PM= project management, FA= funding acquisition.

Conflict of Interest

The authors declared that there is no conflict of interest.

Ethical Consideration

Ethics committee approval was not required for this study because of there was no study on animals or humans.

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