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# Stabilization of the Antioxidant Properties in Spray-Dried Microcapsules of Fish and Chia Oil Blends

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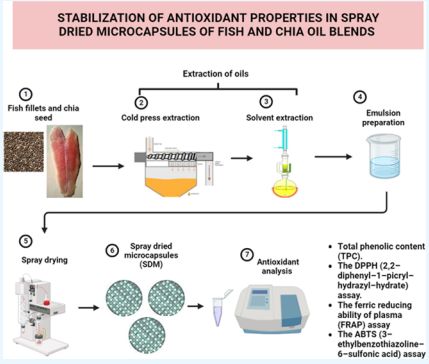
**ABSTRACT:** Even with healthy foods, there is still a need to protect the functionality during processing. The stabilization and enrichment of fish oil (FO) extracted from fish fillets using solvent extraction might make this healthy oil more available. FO was stabilized by mixing it with chia seed oil (CSO) at 50:50 at room temperature. The antioxidant properties of the blends were evaluated using the total phenolic content (TPC), free radical scavenging activity (DPPH), ferric reducing antioxidant potential (FRAP), and 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS) activities with FO and CSO as controls. The blends of FO and CSO increased the oxidative stability, while FO was the most susceptible to degradation. The stability and bioactivity of antioxidants against environmental factors were improved by using encapsulation. Response surface methodology (RSM) was used to optimize spray-drying operating conditions for spray-dried microcapsules (SDMs). The independent variables were the inlet air temperature (IAT), which varied from 125 to 185 °C; wall material (WM) concentration, which varied from 5 to 25%; pump speed (PS), which varied from 3 to 7 mL/min; and needle speed (NS), which varied from 3 to 11 s. The results indicated that the maximum antioxidant activity of SDM was obtained at 140 °C IAT, 10% WM, 4 mL/min PS, and 5 s NS, while the minimum value was obtained at 170 °C IAT, 20% WM, 6 mL/min PS, and 9 s NS. The IAT had a significant effect on the antioxidant activities, and the stability of SDMs was increased. These SDMs can be used in the formulation of food matrices due to their therapeutic and nutritional properties.




## 1. INTRODUCTION

The demand for long-chain polyunsaturated fatty acids (LCPUFAs), including $\alpha$ linolenic acid (ALA), eicosapentaenoic acid (EPA), and docosahexaenoic acid (DHA) of plant and marine origins for human consumption, which have effective health benefits, has been increasing.[1] These essential (E) LCFAs can be obtained by consuming the fish and chia seed oils, and are considered an important source of EFAs. Fish oil (FO) obtained from rohu fillets (*Labeo rohita*) lacks fat-soluble phenolic compounds, decreasing its antioxidant properties, typical of animal-based oils.[2,3] Omega-3 FAs can reduce free radical levels, although they are not considered to be strong antioxidants.[4] On the other hand, chia seed oil (CSO) is a good source of fat-soluble phenolic compounds that are natural antioxidants such as tocopherols, tetraterpenoids, plant sterols or phytosterols, chlorogenic acid, flavonoid, and flavonol.[5,6] The antioxidant activity of CSO focuses on neutralizing free radical damage in the human body. In addition to phytochemicals, CSO also contains omega-3 FAs that have cholesterol-lowering and anti-obesity activities.[7]

FO has a good amount of saturated FAs as palmitic and stearic acids,[8] while CSO has little saturated FAs.[9] Therefore, the blend of these two oils may lead to an omega-3-enriched oil with beneficial health effects.[10−13] However, the addition of omega-3-enriched oil to food products, especially in water-based products, is limited due to its low solubility and hydrophobicity.[14,15] LCPUFA-enriched oils are susceptible to lipid oxidation catalyzed by various environmental factors such as light, high temperature, and oxygen in the air.[16] Furthermore, controlling the delivery of omega-3-enriched oil using different optimal processing for different environmental conditions is also important to enhance their usefulness. Therefore, the optimal processing for different environmental conditions must be studied.

To improve the oxidative stability of an omega-3-enriched oil, microencapsulation can be used to control the release of drugs and other compounds for nutraceuticals and pharma-

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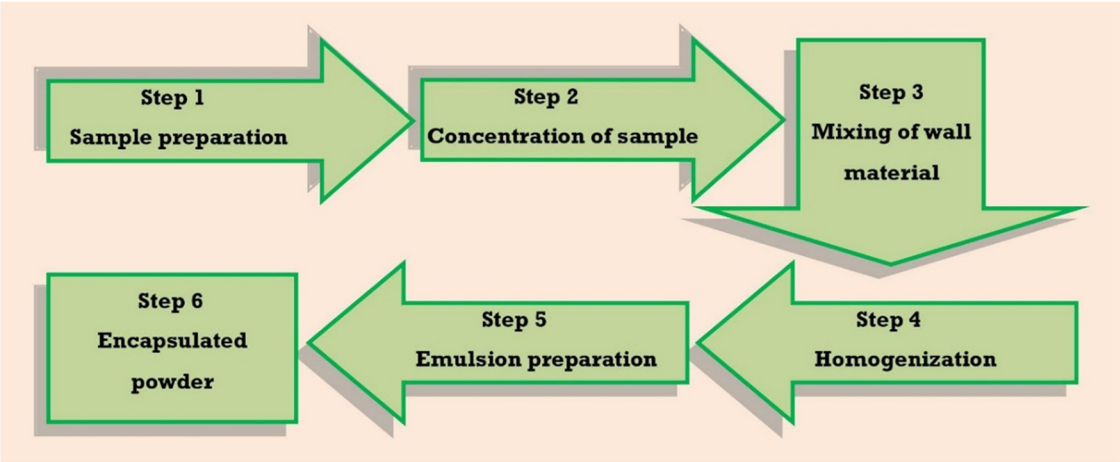


**Figure 1.** Schematic diagram of sample preparation for spray-drying.

ceuticals and also in food processing and preservation for multiple purposes such as the maximum retention of flavors and EFA, stabilization of antioxidants, and antimicrobial activity.[2,17] Microencapsulation is tailored to preserving phytochemicals, slowing down degradation processes, and preserving functional activity.[18] To prepare microencapsulated materials, spray-drying is an effective and often used method because it is a simple, rapid, cost-effective, continuous, scalable, automated, reproducible, and economical method for the production of a free-flowing powder from a liquid oil with both low- and high-temperature processing conditions.[19] Additionally, spray-dried powders can be used for a variety of purposes in food formulations on an industrial scale. It, therefore, is a way to achieve a controlled release of phenolic compounds.[20] The objectives of this study were to evaluate the effect of wall material (WM), pump speed (PS), needle speed (NS), and inlet air temperature (IAT) on the antioxidant properties of blends of FO and CSO spray-dried microcapsules (SDMs).

## 2. MATERIALS AND METHODS

**2.1. Raw Materials.** Chia seeds (Gazala's Pantry, Faisalabad, Punjab, Pakistan) and fish fillets (*L. rohita*) were purchased from a registered supermarket (Faisalabad, Punjab, Pakistan). All Sigma−Aldrich chemicals were purchased from a Local Scientific Stores, Faisalabad, Punjab, Pakistan. The raw materials were cleaned by removing any impurities and unnecessary materials. *L. rohita* of length 1 m and weight of 2 kg were used for the extraction of oil

**2.2. Processing of Oils.** *2.2.1. Extraction of CSO.* The CSO was obtained from chia seeds using a laboratory-scale screw oil press (model 6YL-550, Zhengzhou, Henan, China) according to Rahim et al.[13] Briefly, chia seeds were fed into a feed hopper at room temperature with the feed rate or auger rate already automatically adjusted. The movement of the screw in the shaft produced sufficient heat to denature the protein and increase the oil extraction. After extraction, the heavier impurities in the oil were removed using a sedimentation method (settling) at 25 ± 1 °C. The oil was then stored in screw-capped PVC bottles and kept in the dark at 25 ± 1 °C for a maximum of 1 week.

*2.2.2. FO Extraction.* A solvent extraction method was used for the extraction according to Rahim et al.[12] Briefly, 50 g of each fish fillet sample was dipped in a solvent mixture of methanol (100 mL) and chloroform (50 mL) and placed at room temperature for over the night. Then, the solvent mixture was evaporated using a rotary evaporator (LabTech, Model, EV311H−V, Hopkinton, Massachusetts) at a temperature of 50 °C, a pressure of 0.7 MPa, and a speed of 25 rpm.

**2.3. Spray-Drying of FO and CSO Blends.** *2.3.1. Emulsion Preparation.* Blends of FO and CSO (50:50) were prepared for spray-drying, as shown in Figure 1. The emulsions of these blends were prepared according to Samantha et al.,[27] with some modifications. The blends were poured into a 500 mL beaker and mixed by using a magnetic stirrer for 15 min at room temperature. For all FO and CSO blend formulations, 1% soy lecithin (w/w) was added slowly.[28] Gum Arabic (GA) and maltodextrin (MD) were used as the WM at 1:1 at 25 ± 1 °C. The emulsions were homogenized at 10,000 rpm for 10 min (FSH−2A, Jiangsu Jinyi Instrument Technology, Changzhou, Jiangsu, China).

*2.3.2. Spray-Drying.* A laboratory-scale mini spray-dryer (TPS−15, Toption, Shanghai, China) was used to make the SDMs using a central composite design (CCD). IAT, WM PS, and NS were in the range of 125−185 °C, 5−25%, 3−7 mL/min, and 3−11 s, respectively. The hydrostatic pump was used to pump the emulsions into the atomizer. The atomizer used compressed hot air. The spray-dried particles and gas were placed into a cyclone separator. The hot gas was exhausted through the exhauster, and the SDMs were collected in a glass collection tube. The SDMs were packed in zipper bags and stored at room temperature for a maximum of 1 week. Their antioxidant properties were measured immediately and after 1 week.

**2.4. Antioxidant Properties.** *2.4.1. Total Phenolic Content (TPC).* The TPC was measured using the Folin−Ciocalteu method with some modifications according to Singleton et al.[21] Oil samples (0.2 mL) were added into the sample tubes with 0.5 mL of Folin−Ciocalteu's reagent and mixed at room temperature. After mixing, the sample tubes were in the dark for 10 to 15 min. Then, 1 mL of saturated sodium carbonate solution was dissolved in the solution. The tubes were again placed in the dark for 30 min. The absorption was measured at 765 nm (Analytik Jena AG−Specord 200 Plus, Jena, Germany). The TPC was estimated using a gallic acid standard curve, assuming that the gallic acid was 100% pure.

*2.4.2. DPPH (2,2−Diphenyl-1-picryl-hydrazyl) Assay.* The antioxidant activity of oil samples was measured using the free


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radical scavenging capacity of DPPH according to the slightly modified version of Rebaya et al.[22] Briefly, 0.5 mg of oil was dissolved in methanol, and a 2 mL solution of 0.004% methanol was prepared for four or more concentrations of each prepared sample in a glass tube. Then, the prepared diluted solution was mixed with a 1 mm DPPH solution and incubated for 30 min in the dark at 25 °C. The absorbance was measured at 517 nm against methanol. The percentage of inhibition of DPPH radicals scavenging activity was measured using eq 1.

$$\text{inhibition (\%)} = \frac{\text{Ac} - \text{As}}{\text{Ac}} \times 100 \quad (1)$$

where Ac and As are the absorbances of the control and sample, respectively.

*2.4.3. Ferric Reducing Antioxidant Potential (FRAP) Assay.* The FRAP test was also used to determine the antioxidant properties of the oil samples. The Benzie and Strain[23] method was chosen with some modifications. Briefly, 5 $\mu$L of oil in a test tube and 180 $\mu$L of FRAP reagent were mixed at room temperature. The samples were incubated at 37 °C for 30 min, and their absorbance was measured at 593 nm at the beginning of the reaction and every 4 min. Vitamin C (ascorbic acid) was used as the standard curve. The FRAP value was determined using eq 2.

$$\text{FRAP}\left(\frac{\mu\text{g of AAE}}{\text{mL}}\right) = \frac{S_1}{S_0} \times \text{FRAP value of standard} \quad (2)$$

where AAE denotes ascorbic acid equivalent, $S_1$ is the change in the absorption of the oil sample from 0−4 min, and $S_0$ is the change of the standard solution from 0−4 min.

*2.4.4. ABTS (2,2′-Azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) Assay.* The ABTS assay was done according to Re et al.,[24] Henriquez et al.,[25] and Uluata and Özdemir[26] with slight modifications. Samples (20 $\mu$L) in a test tube at different concentrations were mixed with 2 mL of freshly prepared ABTS stock solution and stored in the dark for 16 h at 25 °C. The absorbance was measured at 734 nm. Butyl hydroxytoluene was used to prepare the standard curve. The ABTS values were calculated using eq 3.

$$\text{ABTS value (\%)} = \frac{\text{Ac} - \text{As}}{\text{Ac}} \times 100 \quad (3)$$

**2.5. Statistical Analysis.** The data was analyzed for their level of significance ($p \leq 0.05$) using the Stat−Ease (version 11.1.2.0, E Hennepin Ave, Minneapolis, MN) and Matlab software (version 7.5.0.338; R2007a, Natick, MA). The actual levels of spray-drying operating conditions were coded with −2, −1, 0, +1, and +2 and response factors were optimized with respect to the operating conditions. The linearity of the CCD for all of the analyzed parameters with their regression coefficient was used to estimate the quadratic effect of the independent factors. For the optimization of spray-dryer conditions, the response surface methodology (RSM) was executed to calculate the maximum value of response factors. Analysis of variance (ANOVA) was employed to check the ampleness of the CCD and significant terms in the model for response factors were found by ANOVA. The level of significance was assessed by the F-statistic intended from the obtained data.[29]

## 3. RESULTS AND DISCUSSION

**3.1. Antioxidant Properties of CSO, FO, and Their Blends.** Antioxidants are substances in foods that have the ability to inhibit oxidation, scavenge, and neutralize the activity of free radicals. The changes in the antioxidant properties of CSO, FO, and their blend are presented in Table 1. The results revealed that the CSO contained 16.1 ± 0.28 $\mu$g of GAE/mL of TPC, 34.92 ± 0.56% of DPPH, 151 ± 1.45 $\mu$g of AAE/mL FRAP, and 22.87 ± 0.31% of ABTS. Moreover, the antioxidant activity was found to be 6.13 ± 0.14 $\mu$g of GAE/mL of TPC, 3.91 ± 0.12% of DPPH, 970 ± 1.67 $\mu$g of AAE/mL of FRAP, and 3.78 ± 0.10% of ABTS for FO. In addition, FO, which was deprived of DPPH, when blended with CSO, now possesses DPPH activity and the same result was observed in TPC activity. Changes were also noted in the FRAP and ABTS assays. Da Silva Marineli et al.[30] showed a high antioxidant activity due to the presence of phenolic compounds in CSO. Furthermore, the results agree with a study using *L. rohita* fish skin oil, which showed FRAP (990 $\mu$g AAE/mL), 3.84% of DPPH, 6.50 $\mu$g GAE/mL of TPC, and 3.92% of ABTS.[10] Similarly, Ghosh et al.[3] reported that the antioxidant activity (TPC and FRAP value) of FO was inversely proportional to the processing factors. The TPC and FRAP values were increased by reducing the heating temperature and time. In addition, Ghosh et al.[31] stated that chia and fish skin oil were mixed in different proportions according to the treatment plan. The results indicated that the 2:1 ratio was better in terms of nutrition in antioxidant activity than the other ratios. Changes in the antioxidant activity of the oil blend were similar to those of Ngassapa et al.[32]

**Table 1. Antioxidant Properties of Crude Oils and Their Blend**[a]

| parameters | CSO | FO | blend |
|---|---|---|---|
| TPC ($\mu$g of GAE/mL) | 16.13 ± 0.28 | 6.13 ± 0.14 | 27.34 ± 0.39 |
| DPPH assay (%) | 34.92 ± 0.56 | 3.91 ± 0.12 | 91.23 ± 0.78 |
| FRAP assay ($\mu$g of AAE/mL) | 151 ± 1.45 | 970 ± 1.67 | 23.11 ± 0.35 |
| ABTS assay (%) | 22.87 ± 0.31 | 3.78 ± 0.10 | 15.23 ± 0.25 |

[a]CSO = chia seed oil; FO = fish oil; TPC = total phenolic contents; DPPH = free radical scavenging capacity; FRAP = ferric reducing antioxidant potential; ABTS = 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid.

**3.2. Antioxidant Properties of SDMs.** *3.2.1. Total Phenolic Contents (TPCs) of SDMs.* Phenolic compounds are quite vital components in omega-3 ($\omega$-3)-enriched oils, with oxidation−reduction properties responsible for their antioxidant properties. The hydroxyl groups in the oil are responsible for simplifying free radical scavenging. Gallic acid is used as the standard and total phenolic content (TPC), which is expressed as $\mu$g of GAE/mL.[33,34] The TPC of all 30 individual runs of the experimental design was estimated using the Folin−Ciocalteu process as represented in Table 2. The results of TPC revealed that the runs contained a considerable amount from 25.17 ± 0.24 to 26.33 ± 0.39 $\mu$g of GAE/mL as gallic acid equivalents, representing an approximate variation. The highest value of TPC was observed ($p \leq 0.05$) in spray-dried microcapsules (SDMs) at a low temperature (26.33 ± 0.39 $\mu$g of GAE/mL) and the lowest value of TPC was noticed ($p \leq 0.05$) in SDMs at a high temperature (25.17 ± 0.24 $\mu$g of GAE/mL). The surface plots of the TPC represent the mutual


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**Table 2. Optimization of Spray-Drying Operating Conditions for Antioxidant Properties of Spray-Dried Microcapsules (SDMs)[a]**

| | independent variables | | | | dependent variables | | | |
|---|---|---|---|---|---|---|---|---|
| spray-drying run | IAT (°C) | WM (%) | PS (mL/min) | NS (S) | TPC (μg of GAE/mL) | DPPH (%) | FRAP (μg of AAE/mL) | ABTS (%) |
| 1 ($C_1$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.77 ± 0.31$^{d}$ | 88.84 ± 0.58$^{e}$ | 20.79 ± 0.14$^{de}$ | 13.59 ± 0.15$^{d}$ |
| 2 ($C_2$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.75 ± 0.31$^{d}$ | 88.85 ± 0.58$^{e}$ | 20.78 ± 0.14$^{de}$ | 13.60 ± 0.15$^{d}$ |
| 3 | 155 (0) | 15 (0) | 5 (0) | 3 (−2) | 25.87 ± 0.32$^{cd}$ | 88.92 ± 0.58$^{de}$ | 20.97 ± 0.15$^{d}$ | 13.72 ± 0.15$^{d}$ |
| 4 | 155 (0) | 15 (0) | 7 (+2) | 7 (0) | 25.12 ± 0.24$^{gh}$ | 88.69 ± 0.57$^{f}$ | 20.67 ± 0.13$^{e}$ | 13.42 ± 0.13$^{e}$ |
| 5 | 140 (−1) | 20 (+1) | 4 (−1) | 9 (+1) | 26.11 ± 0.36$^{b}$ | 89.16 ± 0.60$^{c}$ | 21.30 ± 0.16$^{bc}$ | 13.88 ± 0.16$^{c}$ |
| 6 | 155 (0) | 25 (+2) | 5 (0) | 7 (0) | 25.66 ± 0.30$^{de}$ | 88.77 ± 0.57$^{ef}$ | 20.74 ± 0.14$^{e}$ | 13.49 ± 0.14$^{de}$ |
| 7 | 170 (+1) | 10 (−1) | 4 (−1) | 5(−1) | 25.53 ± 0.29$^{e}$ | 88.63 ± 0.56$^{fg}$ | 20.61 ± 0.13$^{ef}$ | 13.28 ± 0.12$^{f}$ |
| 8 ($C_3$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.78 ± 0.31$^{d}$ | 88.86 ± 0.58$^{de}$ | 20.77 ± 0.14$^{de}$ | 13.54 ± 0.14$^{de}$ |
| 9 | 140 (−1) | 10 (−1) | 4 (−1) | 5(−1) | 26.33 ± 0.39$^{a}$ | 89.45 ± 0.67$^{a}$ | 21.55 ± 0.18$^{a}$ | 14.16 ± 0.18$^{a}$ |
| 10 | 170 (+1) | 20 (+1) | 4 (−1) | 5(−1) | 25.44 ± 0.28$^{ef}$ | 88.53 ± 0.56$^{g}$ | 20.47 ± 0.12$^{fg}$ | 13.15 ± 0.12$^{fg}$ |
| 11 ($C_4$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.76 ± 0.33$^{d}$ | 88.87 ± 0.58$^{de}$ | 20.78 ± 0.14$^{de}$ | 13.61 ± 0.15$^{d}$ |
| 12 ($C_5$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.77 ± 0.33$^{d}$ | 88.85 ± 0.58$^{de}$ | 20.78 ± 0.14$^{de}$ | 13.59 ± 0.15$^{d}$ |
| 13 | 155 (0) | 5 (−2) | 5 (0) | 7 (0) | 25.59 ± 0.29$^{e}$ | 88.72 ± 0.57$^{f}$ | 20.70 ± 0.13$^{e}$ | 13.45 ± 0.13$^{e}$ |
| 14 | 140 (−1) | 10 (−1) | 6 (+1) | 9 (+1) | 26.15 ± 0.36$^{b}$ | 89.23 ± 0.64$^{bc}$ | 21.34 ± 0.17$^{b}$ | 13.92 ± 0.16$^{bc}$ |
| 15 | 170 (+1) | 10 (−1) | 6 (+1) | 5(−1) | 25.39 ± 0.27$^{f}$ | 88.46 ± 0.55$^{gh}$ | 20.40 ± 0.12$^{g}$ | 13.08 ± 0.12$^{g}$ |
| 16 | 170 (+1) | 10 (−1) | 6 (+1) | 9 (+1) | 25.28 ± 0.25$^{g}$ | 88.40 ± 0.55$^{h}$ | 20.34 ± 0.12$^{gh}$ | 13.01 ± 0.12$^{gh}$ |
| 17 | 170 (+1) | 10 (−1) | 4 (−1) | 9 (+1) | 25.32 ± 0.26$^{fg}$ | 88.34 ± 0.54$^{hi}$ | 20.27 ± 0.11$^{gh}$ | 12.92 ± 0.12$^{gh}$ |
| 18 | 140 (−1) | 20 (+1) | 4 (−1) | 5(−1) | 26.26 ± 0.37$^{ab}$ | 89.37 ± 0.66$^{ab}$ | 21.48 ± 0.18$^{ab}$ | 14.09 ± 0.17$^{ab}$ |
| 19 | 140 (−1) | 10 (−1) | 4 (−1) | 9 (+1) | 26.21 ± 0.37$^{ab}$ | 89.27 ± 0.54$^{i}$ | 21.39 ± 0.17$^{b}$ | 13.97 ± 0.16$^{bc}$ |
| 20 | 140 (−1) | 10 (−1) | 6 (+1) | 5(−1) | 26.16 ± 0.36$^{b}$ | 89.32 ± 0.65$^{b}$ | 21.43 ± 0.17$^{ab}$ | 14.04 ± 0.17$^{b}$ |
| 21 | 185 (+2) | 15 (0) | 5 (0) | 7 (0) | 25.35 ± 0.26$^{fg}$ | 88.43 ± 0.55$^{gh}$ | 20.75 ± 0.14$^{de}$ | 13.31 ± 0.13$^{ef}$ |
| 22 | 140 (−1) | 20 (+1) | 6 (+1) | 9 (+1) | 26.07 ± 0.35$^{bc}$ | 89.13 ± 0.60$^{c}$ | 21.26 ± 0.16$^{bc}$ | 13.83 ± 0.16$^{cd}$ |
| 23 ($C_6$) | 155 (0) | 15 (0) | 5 (0) | 7 (0) | 25.78 ± 0.31$^{d}$ | 88.84 ± 0.58$^{e}$ | 20.77 ± 0.14$^{de}$ | 13.60 ± 0.15$^{d}$ |
| 24 | 155 (0) | 15 (0) | 5 (0) | 11 (+2) | 25.92 ± 0.33$^{cd}$ | 88.98 ± 0.58$^{d}$ | 21.03 ± 0.15$^{cd}$ | 13.34 ± 0.13$^{ef}$ |
| 25 | 155 (0) | 15 (0) | 3 (−2) | 7 (0) | 25.98 ± 0.33$^{c}$ | 89.05 ± 0.59$^{cd}$ | 21.08 ± 0.15$^{cd}$ | 13.40 ± 0.13$^{e}$ |
| 26 | 170 (+1) | 20 (+1) | 6 (+1) | 9 (+1) | 25.17 ± 0.24$^{gh}$ | 88.22 ± 0.54$^{ij}$ | 20.13 ± 0.11$^{hi}$ | 12.84 ± 0.12$^{h}$ |
| 27 | 170 (+1) | 20 (+1) | 4 (−1) | 9 (+1) | 25.48 ± 0.28$^{ef}$ | 88.57 ± 0.56$^{g}$ | 20.54 ± 0.12$^{f}$ | 13.21 ± 0.12$^{fg}$ |
| 28 | 170 (+1) | 20 (+1) | 6 (+1) | 5(−1) | 25.24 ± 0.25$^{g}$ | 88.30 ± 0.54$^{hi}$ | 20.22 ± 0.11$^{h}$ | 12.88 ± 0.11$^{h}$ |
| 29 | 125 (−2) | 15 (0) | 5 (0) | 7 (0) | 25.95 ± 0.33$^{c}$ | 89.25 ± 0.64$^{bc}$ | 21.15 ± 0.16$^{c}$ | 13.95 ± 0.16$^{bc}$ |
| 30 | 140 (−1) | 20 (+1) | 6 (+1) | 5(−1) | 26.04 ± 0.35$^{bc}$ | 89.10 ± 0.59$^{cd}$ | 21.17 ± 0.16$^{c}$ | 13.77 ± 0.16$^{cd}$ |

[a]SDMs = spray-dried microcapsules; IAT = inlet air temperature; WM = wall material; PS = pump speed; NS = needle speed; TPC = total phenolic contents; DPPH = 2,2-diphenyl-1-picryl-hydrazyl; FRAP = ferric reducing antioxodant potential; ABTS = 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid; C1−C6 represents the central points of CCD; a−j means with different superscripts differ significantly ($p \leq 0.05$).

interaction between independent variables, as explained in Figure 2. Moreover, the differences in TPC of SDMs for the polynomial equation were found to be statistically significant in all experimental runs, as displayed in Table 3. The *F*-value and *p*-value of TPC were 5.94, indicating that the model was significant. Furthermore, the predicted $R^2$ value was 0.7203, which is close to the adjusted $R^2$ (0.7045), indicating no response transformation. The data were analyzed using CCD of the model regression equation of TPC in terms of both actual factor $R_1$ and coded factor $R_2$. The regression equation of TPC in coded and actual factors was used to make predictions about the dependent variables for given levels of each independent variable (Table 4). A similar study was carried out by Mohammed et al.[35] on the microencapsulation of black seed oil using the spray-drying method. In this research work, a CCD of RSM was used to optimize the processing conditions. Results of this research work indicated that the TPC in microencapsulated black seed oil ranged from 76.51 to 137.68 mg of GAE per 100 mL. According to the analysis of variance, three independent factors of spray-drying were significantly affected by the TPC of microencapsulated black seed oil powder. The TPC value of the microencapsulated black seed oil powder was reduced by increasing the temperature of operating conditions. Another research work reported that the TPC value of a spray-dried low-fat honey-based milk powder was significantly increased by reducing the temperature.[36] Furthermore, in another study, an increase in temperature reduced the TPC value of spray-dried propolis oil from 21.2 to 20.6 g GAE per 100 g.[37] Change in the TPC value of chia and fish skin oil blend was similar to the recent research work by Ghosh et al.[10] In addition, the phenolic content in chia seed oil (CSO), which is a good natural source of antioxidants to reduce the risk factor of various diseases, occurs due to the oxidative stress. Numerous research studies have concluded that the phenolic compounds present in essential oils have an important role in the prevention of many chronic diseases such as heart disease, inflammation, diabetes, and metabolic disorders.[38,39]

*3.2.2. Free Radical Scavenging Capacity (DPPH) Assays of SDMs.* Changes in the free radical scavenging capacity (DPPH) of spray-dried microcapsules (SDMs) are represented in Table 2. In this study, the maximum DPPH value of SDM was obtained at a low temperature of 140 °C and the minimum DPPH value was calculated at a high temperature of 170 °C. In fact, the inlet air temperature of spray-drying conditions also significantly affected the DPPH values. The results concluded that the DPPH values of SDMs were decreased by the increase in the temperature. Figure 3, indicating the contour plots,


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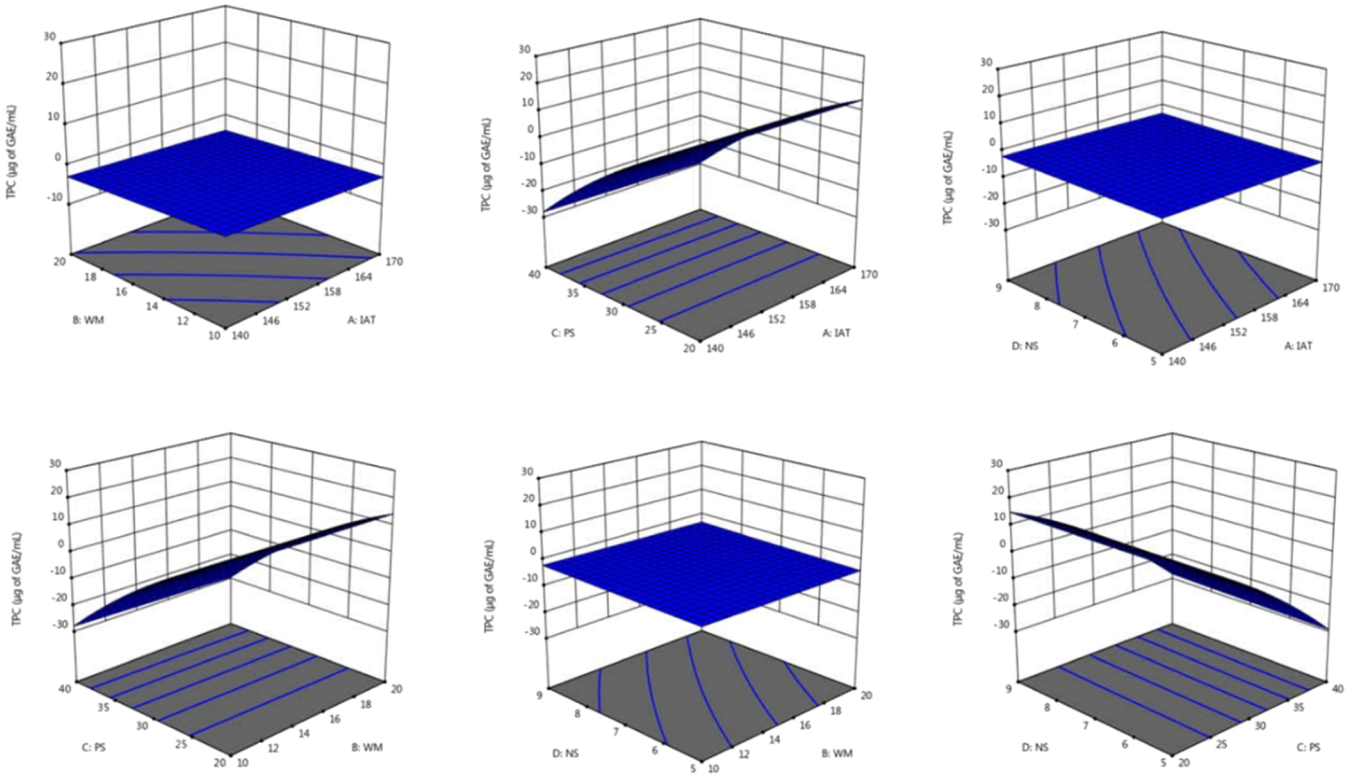


**Figure 2.** Interaction impact of spray-drying operating factors on the TPC.

**Table 3. Analysis of Variance (ANOVA) for Independent Variables' Effect on the Dependent Variable of Spray-Dried Microcapsules (SDMs)[a]**

| source of variation | | DF | TPC (µg of GAE/mL) | | DPPH (%) | | FRAP (µg of AAE/mL) | | ABTS (%) | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | MS | *p*-value | MS | *p*-value | MS | *p*-value | MS | *p*-value |
| model | | 14 | 0.2135* | 0.0007 | 0.2210* | <0.0001 | 0.2569* | 0.0050 | 0.2445* | 0.0005 |
| linear effects | IAT | 1 | $0.0102^{NS}$ | 0.6016 | $0.0072^{NS}$ | 0.5615 | $0.0164^{NS}$ | 0.6154 | 0.1120* | 0.5828 |
| | WM | 1 | $0.0086^{NS}$ | 0.6318 | $0.0167^{NS}$ | 0.3802 | $0.0322^{NS}$ | 0.4837 | $0.0349^{NS}$ | 0.3539 |
| | PS | 1 | $0.0518^{NS}$ | 0.2488 | $0.0001^{NS}$ | 0.9456 | $0.0051^{NS}$ | 0.7790 | 0.1605* | 0.2268 |
| | NS | 1 | $0.0044^{NS}$ | 0.7302 | 0.1112* | 0.4700 | 0.1122* | 0.6641 | $0.0148^{NS}$ | 0.5427 |
| interaction effects | IAT × WM | 1 | 0.1420* | 0.8156 | $0.0056^{NS}$ | 0.6070 | $0.0036^{NS}$ | 0.8135 | 0.2160* | 0.6969 |
| | IAT × PS | 1 | $0.0025^{NS}$ | 0.7956 | $0.0009^{NS}$ | 0.8364 | 0.2349* | 0.7832 | 0.1128* | 0.7916 |
| | IAT × NS | 1 | $0.0006^{NS}$ | 0.8969 | $0.0001^{NS}$ | 0.9451 | $0.0004^{NS}$ | 0.9373 | 0.2312* | 0.9498 |
| | WM × PS | 1 | 0.2181* | 0.6419 | 0.2156* | 0.3950 | 0.1306* | 0.4944 | 0.1333* | 0.3646 |
| | WM × NS | 1 | $0.0056^{NS}$ | 0.6980 | $0.0056^{NS}$ | 0.6070 | 0.1182* | 0.5969 | $0.0233^{NS}$ | 0.4468 |
| | PS × NS | 1 | $0.0049^{NS}$ | 0.7172 | $0.0121^{NS}$ | 0.4529 | 0.1132* | 0.6519 | $0.0176^{NS}$ | 0.5076 |
| quadratic effects | $IAT^2$ | 1 | $0.0071^{NS}$ | 0.6641 | $0.0004^{NS}$ | 0.8865 | 0.1320* | 0.4849 | $0.0023^{NS}$ | 0.8112 |
| | $WM^2$ | 1 | $0.0136^{NS}$ | 0.5474 | 0.1211* | 0.3255 | 0.1149* | 0.6318 | $0.0263^{NS}$ | 0.4195 |
| | $PS^2$ | 1 | $0.0462^{NS}$ | 0.2748 | $0.0003^{NS}$ | 0.8983 | 0.1165* | 0.7510 | $0.0579^{NS}$ | 0.2367 |
| | $NS^2$ | 1 | $0.0561^{NS}$ | 0.2310 | $0.0152^{NS}$ | 0.4013 | $0.0597^{NS}$ | 0.3435 | $0.0070^{NS}$ | 0.6750 |
| residual | | 15 | 0.1360* | | 2.1204* | | 2.0624* | | 1.0381* | |
| lack of fit | | 10 | 0.1539* | 0.6510 | 3.0305* | 1.7231 | 1.0922* | 0.0006 | 1.0568* | 0.8726 |
| pure error | | 5 | $0.0001^{NS}$ | | $0.0001^{NS}$ | | $0.0029^{NS}$ | | 0.116* | |
| cor total | | 29 | | | | | | | | |

[a]SDMs = spray-dried microcapsules; TPC = total phenolic contents; DPPH = 2,2-diphenyl-1-picryl-hydrazyl; FRAP = ferric-reducing antixodant potential; ABTS = 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid; DF = degree of freedom; MS = mean squares; * = significant; NS = nonsignificant; IAT = inlet air temperature; WM = wall material; PS = pump speed; NS = needle speed.

reveals the interaction impacts of two operating factors on the DPPH assays. Moreover, the *p*-value and *F*-value of linear and quadratic have a significant effect on the DPPH value, as described in Table 3. The *p*-value of the model indicates that the model was significant. On the other hand, the lack of fit was less than 0.05, which shows a significant effect. In the fit statistic, the adjusted $R^2$ was 0.8262 and $R^2$ was 0.9101. The data were analyzed using the CCD of model regression equation of DPPH values in terms of both actual factor $R_1$ and coded factor $R_2$. The regression equation of DPPH value in coded and actual factors was used to make predictions about the dependent variables for given levels of each independent


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**Table 4. Coded and Actual Regression Equations for Dependent Variables after Operating Conditions[a]**

| dependent variables | regression form | regression equations |
|---|---|---|
| TPC ($\mu$g of GAE/mL) | coded | $R_1 = -2.90 - 0.6325A - 0.6325B - 21.73C + 0.4167D + 0.0113AB - 0.1250AC - 0.0062AD - 0.2250BC + 0.0188BD + 0.1750CD - 0.0160A^2 - 0.0223B^2 - 4.10C^2 + 0.0452D^2$ |
| | actual | $R_2 = +27.06 + 0.0041IAT + 0.0093WM + 0.42500PS - 0.2082NS + 0.0001IAT \times WM - 0.0008IAT \times PS - 0.0002IAT \times NS - 0.0045WM \times PS + 0.0018WM \times NS + 0.0087PS \times NS - 0.0007IAT^2 - 0.0008WM^2 - 0.0410PS^2 + 0.0041NS^2$ |
| DPPH (%) | coded | $R_3 = +89.00 - 0.5300A - 0.8071B + 0.9458C + 0.6617D + 0.0188AB - 0.0750AC - 0.0025AD - 0.3125BC + 0.0188BD + 0.2750CD - 0.0040A^2 - 0.0277B^2 + 0.3542C^2 + 0.0235D^2$ |
| | actual | $R_4 = +92.98\text{-}0.0180IAT + 0.0074WM - 0.0429PS - 0.1792NS + 0.0002IAT \times WM - 0.0005IAT \times PS - 0.0008IAT \times NS - 0.0062WM \times PS + 0.0018WM \times NS + 0.0013PS \times NS - 0.0001IAT^2 - 0.0011WM^2 + 0.0035PS^2 + 0.0058NS^2$ |
| FRAP ($\mu$g of AAE/mL) | coded | $R_5 = +28.21 - 0.8017A - 0.6325B - 1.12C + 0.6921D + 0.0150AB - 0.1750AC - 0.0050AD - 0.4375BC + 0.0338BD + 0.2875CD + 0.0342A^2 - 0.0233B^2 + 1.54C^2 + 0.0467D^2$ |
| | actual | $R_6 = +29.16 + 0.0673IAT + 0.0114WM - 0.03187PS - 0.2734NS + 0.0002IAT \times WM - 0.0011IAT \times PS - 0.0001IAT \times NS - 0.0087WM \times PS + 0.0033WM \times NS + 0.0143PS \times NS + 0.0001IAT^2 - 0.0009WM^2 + 0.0154PS^2 + 0.0011NS^2$ |
| ABTS (%) | coded | $R_7 = -16.42 - 0.6852A - 1.17B - 23.49C + 0.7602D + 0.0194AB - 0.1313AC + 0.0031AD - 0.4556BC + 0.0381BD + 0.3312CD + 0.0091A^2 + 0.0309B^2 - 4.59C^2 - 0.0159D^2$ |
| | actual | $R_8 = +17.53 + 0.036521IAT + 0.0106WM + 0.5638PS - 0.1343NS + 0.0002IAT \times WM - 0.0008IAT \times PS + 0.0001IAT \times NS - 0.0091WM \times PS + 0.0038WM \times NS + 0.0165PS \times NS + 0.0004IAT^2 - 0.0012WM^2 - 0.0459PS^2 - 0.0039NS^2$ |

[a]TPC = total phenolic contents; DPPH = 2,2-diphenyl-1-picryl-hydrazyl; FRAP = ferric reducing antioxidant potential; ABTS = 2,2′- azino-bis(3-ethylbenzothiazoline-6-sulfonic acid; $R_1$, $R_2$, $R_3$, $R_4$, $R_5$, $R_6$, $R_7$, $R_8$ = dependent variables; A = inlet air temperature (IAT); B = wall material (WM); C = pump speed (PS); D = needle speed (NS); IAT, WM, PS, NS = independent variables.

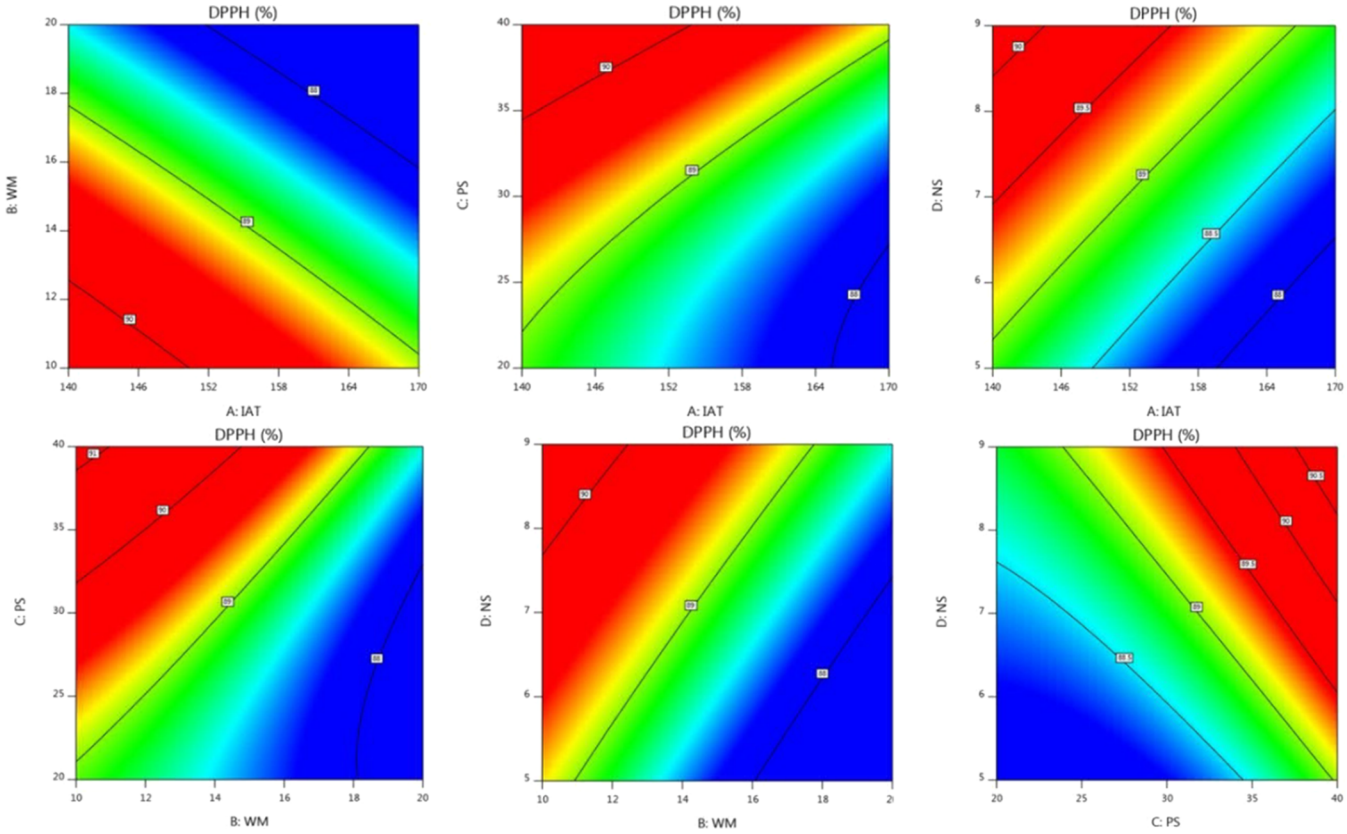


**Figure 3.** Interaction effects of independent factors on DPPH.

variable (Table 4). A similar change in the antioxidant properties of microencapsulated chia oil was found in the research work of Ullah et al.[40] and Copado et al.[41]

*3.2.3. Ferric Reducing Antioxidant Potential (FRAP) Assays.* The ferric reducing antioxidant potential (FRAP) assays is used to measure the combined impact of redox-active antioxidants in oils and their products.[42] The FRAP value of spray-dried microcapsules (SDMs) is shown in Table 2. The results indicated that the amount of FRAP in the SDM obtained from all of the experiments varied from 20.13 ± 0.11 to 21.55 ± 0.18 $\mu$g of AAE/mL on the run order of 26 and 9, respectively. The FRAP in SDMs exhibited a higher value with a low inlet temperature of 140 °C, wall material of 10%, pump speed of 4 mL/min, and needle speed of 5S and the lowest value with an inlet temperature of 170 °C, wall material of 20%, pump speed of 6 mL/min, and needle speed of 9S. The spray-drying operating factors have a significant effect on the FRAP value of SDMs. The analysis of variance (ANOVA) for independent factors impacts the response value of FRAP, as shown in Table 3. The model $F$-value of 4.11 means that the model was significant and adjusted $R^2$ was 0.6006. The obtained data was evaluated using the CCD of the model


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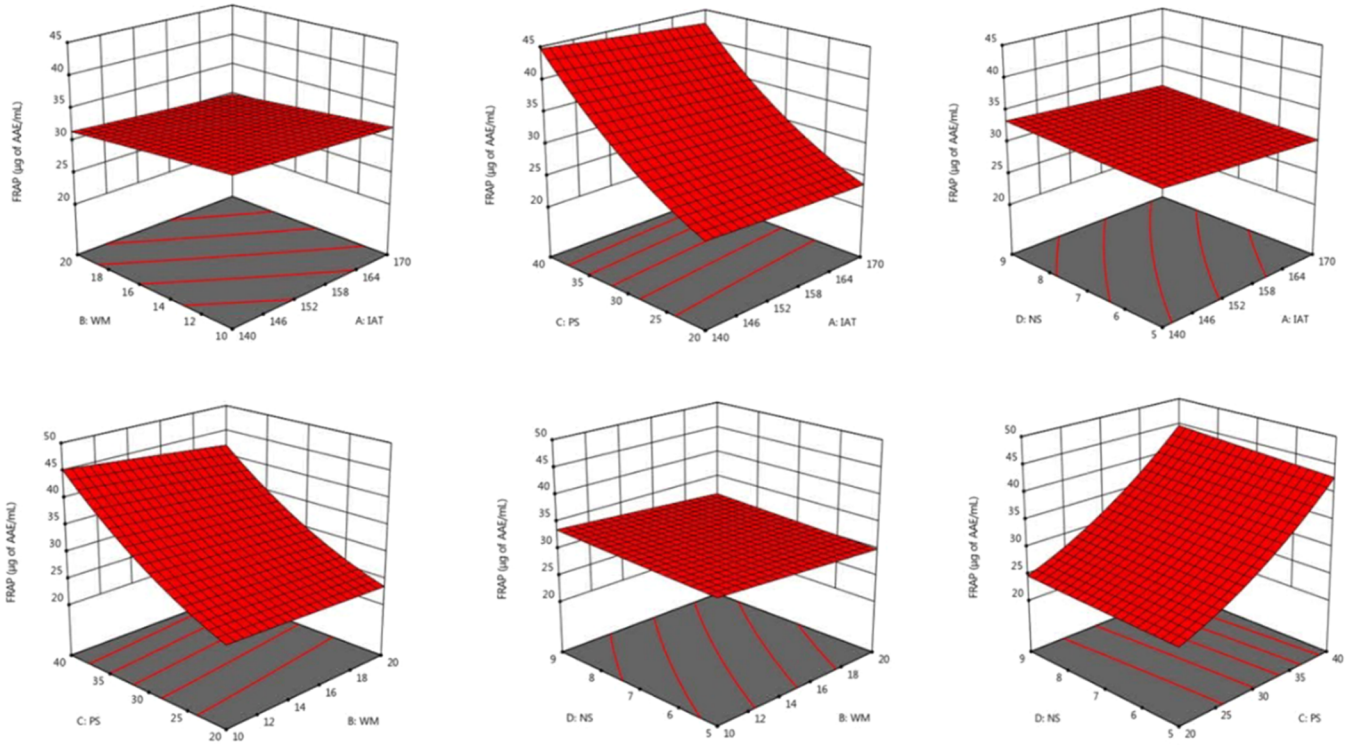


**Figure 4.** Mutual interaction impacts of spray-drying operation on FRAP.

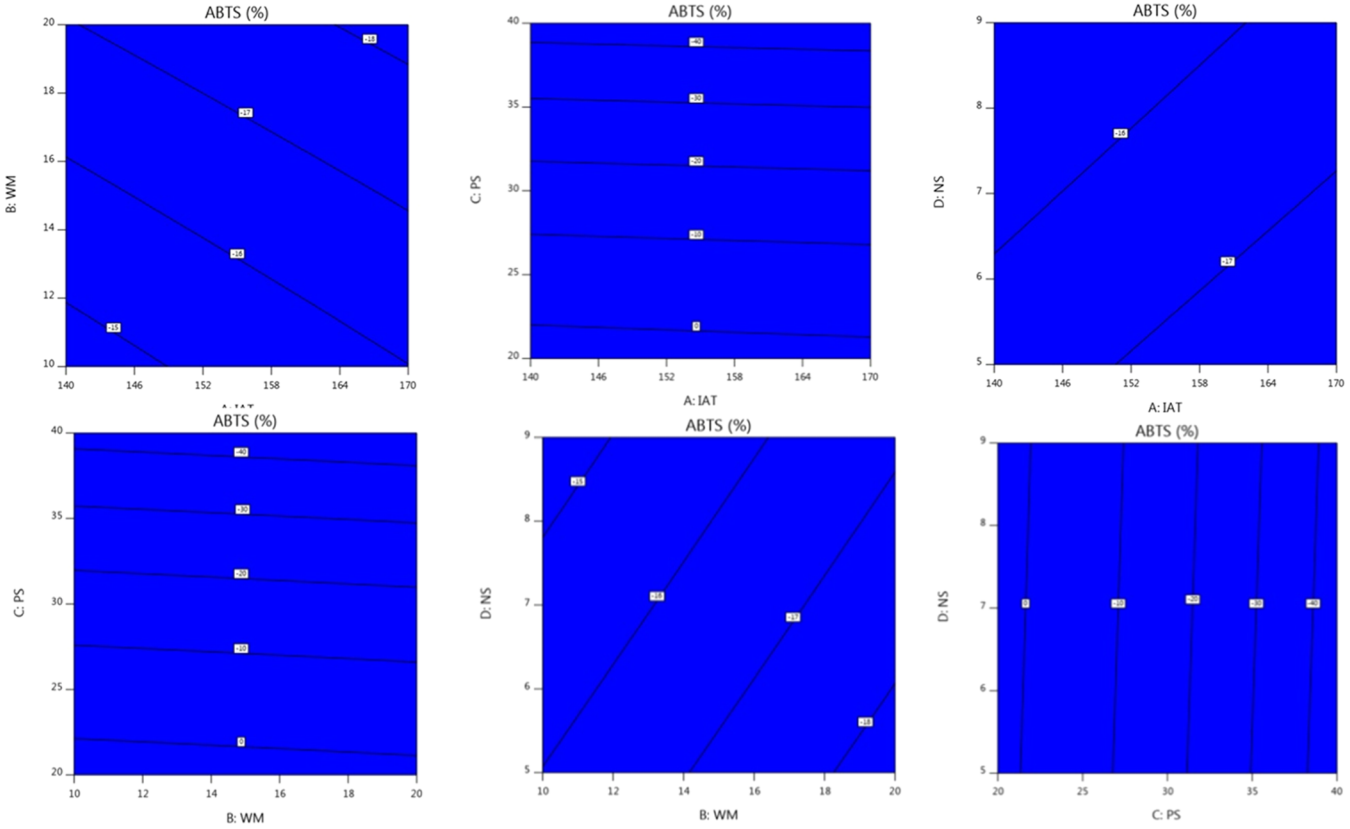


**Figure 5.** Interaction effect of the operating factors on ABTS.

regression equation of FRAP in terms of both actual variables $R_1$ and coded factor $R_2$. The regression equation of FRAP in coded and actual factors was used to make predictions about the dependent variables for given levels of each independent variable (Table 4). Furthermore, the surface plots of FRAP revealed interaction effects between independent variables, as described in Figure 4. Razmkhah et al.[43] reported that the FRAP value in microencapsulated kenaf seed oil was reduced



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by increasing the spray-drying temperature. In another research work conducted by Pashazadeh et al.,[44] the FRAP value of encapsulated phenolic compounds of a maize byproduct was reduced by increasing the time and temperature. Furthermore, the FRAP value of spray-dried soy-rapeseed lecithin/trehalose liposomes was significantly decreased by increasing the storage temperature and relative humidity.[45] FRAP is capable of scavenging free radicals in the human body. Antioxidant compounds in chia seed oil (CSO) reduce the risk of chronic diseases such as heart disease, type 1 diabetes, and cancer. In fact, previous studies have shown a link between the intake of fatty fish and long-chain polyunsaturated FAs containing oils as an antioxidant in plasma and a reduction in the incidence of heart disease.[30,46,47]

*3.2.4. ABTS Assays.* The antioxidant property was determined by the free radical scavenging of 2,2′-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) (ABTS), and the results are indicated in Table 2. The results showed that the maximum ABTS of spray-dried microcapsules (SDMs) was obtained at a run order of 9 (14.16 ± 0.18%) and the minimum amount of ABTS in SDM was 12.84 ± 0.11% at a high temperature of 170 °C. The mutual interaction effects of the operating variables on the ABTS of the SDM are described in Figure 5. Furthermore, the ANOVA quadratic model indicated that the model was significant and the *F*-value of the model was 6.42, as shown in Table 3. In addition, the *p*-value of the linear effects of the independent variables was not significantly affected by the ABTS responses, and a similar phenomenon was observed in the interaction and quadratic effects. The lack of fit of the *F*-value was 92.16, which indicates a significant effect on the ABTS values. The predicted $R^2$ of 0.1794 was not close to the adjusted $R^2$ of 0.7234, and the $R^2$ was 0.8569. The obtained data was evaluated using the CCD of the model regression equation of FRAP in relation to both actual variables $R_1$ and coded variables $R_2$. The regression equation of ABTS in coded and actual factors was used to make predictions about the dependent variables for given levels of each independent variable (Table 4). This observation is based on the results of Morales et al.[48] and Sánchez et al.,[49] who observed similar changes in the antioxidant properties of microencapsulated oil.

## 4. CONCLUSIONS

The main objective of this study is the need for proper blending of oils as no single oil can provide all of the optimal nutrients and improve their heat stability. Here, blending of a multisource oil improves the antioxidant properties of the oils. Antioxidative activity analysis states that blends of FO and CSO can withstand oxidative damage, as all of the results of TPC, DPPH, FRAP, and ABTS are under a satisfactory range. In this research work, it can be concluded that for blends of CSO and FO, the maximum antioxidative property of SDMs was obtained at 140 C, while the minimum amount was estimated at 170 °C. We concluded that there was a correlation between antioxidant analysis and temperature. The blend of FO and CSO can be used for multiple purposes as health-promoting foods. Furthermore, its spray-dried microcapsules (SDMs) can be used in food formulations as well as for therapeutic and pharmaceutical purposes. However, further research work is needed to evaluate the preservation impacts of this type of microcapsules as an additive to control oxidation in several food items.

## ■ ASSOCIATED CONTENT

### Data Availability Statement

Data is contained within the article.

## ■ AUTHOR INFORMATION


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M.A.R. and M.I. performed the methods and investigation. M.N., W.K., and S.A. were responsible for conceptualization, funding acquisition, and writing of the original draft. F.A.K., M.O.A. J.M.R, and I.H. helped in writing the manuscript. M.A.R., W.K., and S.O. helped with software. I.H. and J.M.R. supported in analysis and supervision of research work. M.O.A.F.A. and I.A.M.A. performed the validation, visualization, funding acquisition, and writing, reviewing, and editing of the manuscript. All authors have read and agreed to the published version of the manuscript.



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