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Cold Plasma Technology for Tomato Processing By-Product Valorization: The Case of Tomato Peeling and Peel Drying

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Abstract: The tomato processing industry is focused on product yield maximization, keeping energy costs and waste effluents to a minimum while maintaining high product quality. In our study, cold atmospheric plasma (CAP) pretreatment enhanced tomato processing to facilitate peelability, a specific peeling process, and enhance peel drying. Peeling force analysis determined that CAP pretreatment of whole tomatoes improved peelability under the conditions used. The specific peeling force after CAP treatment decreased by more than three times. It was observed that cold atmospheric plasma pretreatment reduced the duration of infrared drying of tomato peels by 18.2%. Along with that, a positive effect on the reduction of the specific energy consumption of peel drying was shown for CAP-pretreated tomato peels. The obtained data show that the technology of cold atmospheric plasma pretreatment, in particular, when processing whole tomatoes and tomato peels, has a promising application in industry, as it can significantly reduce the specific energy consumption for peeling and drying procedures.

Keywords: cold plasma; process engineering; drying; by-product; tomato peel; energy efficiency

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Citation: Sosnin, M.; Berestenko, E.; Mounassar, E.H.A.; Shorstkii, I. Cold Plasma Technology for Tomato Processing By-Product Valorization: The Case of Tomato Peeling and Peel Drying. *Eng* **2023**, *4*, 2167–2177. https://doi.org/10.3390/eng4030124

Academic Editor: Antonio Gil Bravo

Received: 20 July 2023
Revised: 11 August 2023
Accepted: 14 August 2023
Published: 15 August 2023

1. Introduction

The use of advanced electrical technologies in food processing engineering is a global trend for sustainable development [1,2]. The possibility of reducing the costs of implementing food processes to obtain safe products allows for facilitating the industrial development of such technologies. At the same time, the effectiveness of the suggested emerging technologies must be considered for each specific task.

The removal of the integumentary tissue (skin) from plant materials is an important preparation stage of industrial tomato processing. This operation is in high demand for tomato paste production as well as jams, fruit and vegetable purees [3,4]. The degree of by-product valorization depends on the efficiency and degree of purification of the integumentary tissue of the tomato fruits [5,6]. The cell membrane is a barrier for the transfer of intracellular components (moisture, juice and other dissolved substances) from the plant structure during industrial processing. According to research data, the energy costs for the peeling procedure of tomato fruits can reach 15% of the total energy costs of processing [5]. Progressive climate change, as well as increased control by regulatory authorities on the level of carbon emissions, contribute to the active search for alternative treatment methods to minimize energy costs. In this regard, the use of modern and effective methods of preparing tomato fruits for the peeling procedure, as well as further processing analysis, is of great theoretical and practical interest.

The valorization of tomato processing by-product is of great interest as an alternative pectin source [3,7]. Grassino [7] noted that fresh tomato pomace contains about 32% protein and 30% carbohydrates, making it a valuable by-product. Several authors have noted that the content of pectin substances in the integumentary tissue of tomatoes can reach 25% [8].

The effective peeling is hindered by the strong connection of the mesocarp with the integument of tomato tissue [6]. To weaken this connection, various methods of tomato preparation are used in the industry: physical (thermal), steam-thermal, mechanical, chemical and a combination of suggested methods [9–12]. The currently available methods have both advantages and disadvantages. For example, when processing via the thermal method, the phenomenon of local overheating of the structure of tomato fruits occurs, which decreases the overall quality characteristics. The mechanical method of tomato preparation in the form of an incision in the integumentary tissue improves the efficiency of skin removal. However, mechanical preparation can be used only in combination with thermal or steam-thermal methods.

In this paper, an emerging pretreatment method of tomato fruit using cold atmospheric plasma (CAP) to enhance peelability and peel drying is considered. Cold plasma treatment for fruit and vegetable processing is a novel and prospective method that has appeared in recent years. The mechanism of CAP technology is based on the use of short electrical high-voltage plasma discharges, which cause the electroporation of cell membranes in the air gap [13–15]. Depending on the specific energy consumption, CAP technology can significantly enhance the mass transfer of intracellular compounds [16] and lead to microbial inactivation [17]. Recently, Bao et al. [18] developed the application of cold plasma pretreatment for improving phenolic extractability from tomato pomace. Extracts from plasma-treated tomato pomace had a higher total phenolic content and antioxidant capacity. An overview of the cold plasma preprocessing for the extraction of bioactive compounds was reported by Bezerra et al. [19] and Du et al. [15].

From a thermodynamic point of view, the resulting intense mass transfer in plant materials subjected to electroporation is caused by the formation of a large number of micropores spontaneously and randomly located on the surface of the material along the lines of electric field strength [20]. CAP pretreatment positively affects the dynamics of mass transfer in plant materials due to changes in volumetric porosity. In comparison with other electroporation-based methods, such as pulsed electric field (PEF), CAP treatment can be applied in the air gap. Andrew et al. [21] reported a significant peeling enhancement using pulsed electric field pretreatment for tomatoes. It was found that PEF treatment (0.5–1.5 kV/cm, 0–8000 pulses, 15 µs pulse width) reduced the work required for peel detachment by up to 72.3%. Currently, CAP technology is actively used in various areas of the food industry to improve technological processes, for example, in the production of juices, drying and extraction of plant materials [22–24]. At the same time, the CAP technology is already carried out in a continuous mode of transportation on a scraper conveyor [25].

The purpose of this work is to study the effect of cold plasma treatment on the efficiency of tomato peeling and tomato peel drying, with an assessment of specific energy consumption.

2. Materials and Methods

2.1. Materials

An industrial tomato processing technology based on a local manufacturer (Krasnodar, Russia) with cold plasma application was used as the engineering object in this study. The experimental scheme is shown in Figure 1. Two major processes were chosen for the application of cold plasma technology: tomato skin removal and tomato skin drying.

To compare the effect of CAP technology, the following processing protocols were considered and analyzed: protocol A, whole tomato + cold plasma treatment + peeling; protocol B, whole tomato + peeling; protocol C, tomato peel + CAP treatment + infrared drying; protocol D, tomato peel + infrared drying. In our experiments, we used tomatoes of the Aurora variety as the object of research. The tomatoes were obtained from a local manufacturer (Krasnodar, Russia).

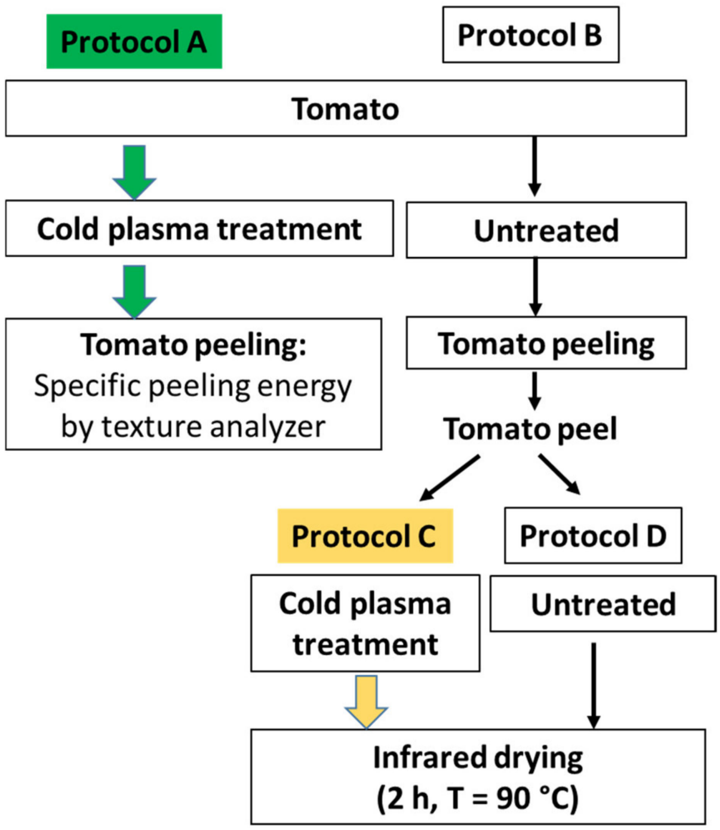

Figure 1. Experimental scheme of tomato processing with cold plasma implementation.

2.2. Cold Plasma Treatment

The principle of operation of the experimental setup in accordance with cold plasma technology [13] is shown in Figure 2a. To generate cold atmospheric plasma in the air gap, a "point-plate" type electrode configuration was used, which included a stainless steel plate as a grounded electrode, and a point steel electrode with a diameter of 1 mm as a high-voltage electrode in a dielectric holder. The gap between the electrodes was set to 70 (for whole tomatoes) and 15 mm (for tomato peels). The processing chamber had a square shape (length 20 cm) made of dielectric material. The grounded electrode in the dielectric holder was mounted on a positional platform with two stepper motors to provide movement along the X-Y axis. The trajectory of the cold plasma treatment nozzle was set using the authors' intelligent object recognition system.

Cold atmospheric plasma (CAP) treatment was performed using a high-voltage power supply system (Matsusada AMPS 20B20, Matsusada Precision, Otsu, Japan) in combination with an Agilent functional generator (Agilent 33220A, Agilent Technologies, Santa Clara, CA, USA) [13]. In all experiments, the pulse duration and the frequency of the plasma discharge were set to 50 microseconds and 100 Hz, respectively. The selected electrical parameters made it possible to precisely control the processing parameters for tomato and tomato peel treatments. Each pulse supplied a voltage of up to 20 kV. In addition, positive pulses with an electric field strength of 2.5 and 8 kV/cm were used for whole tomato and tomato peel, subsequently. The average specific energy consumption of CAP treatment for all experiments was 1.7 kJ/kg at 6000 discharges. The total CAP treatment time was 3 s per cm^2. The temperature difference between the pretreated tomato samples and the control sample was less than 2 °C.

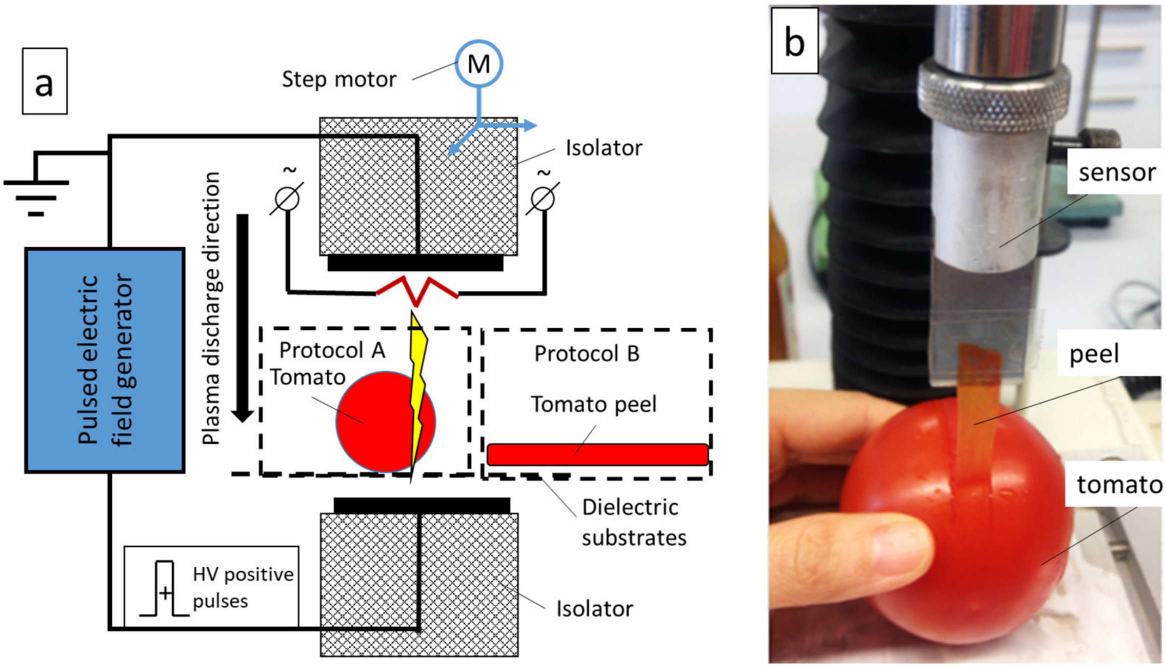

Figure 2. CAP treatment experimental scheme (**a**) and tomato peeling procedure based on texture analyzer system (**b**).

2.3. Skin Removal

In our study, the tomato peeling was performed mechanically using a texture analyzer (CT3-4500 texture analyzer, Brookfield Ametek, Middleborough, MA, USA). The selected precut area on the tomato skin was removed, and the specific peeling force was calculated according to the methodology described in [26]. Figure 2b demonstrates the peeling procedure.

The specific peeling force (F_p) was calculated from the peeling energy and the area of the peeled skin as follows:

$$F_p = \frac{E_p}{S} \tag{1}$$

where E_p—peeling energy from texture analyzer, N; S—area of the peeled skin sample, m^2.

To determine the area of the peeled skin, the sample was photographed on a whiteboard with a tripod positioned at a fixed distance. Afterwards, the pictures were analyzed using ImageJ (v 1.52e), a Java-based image-processing program.

Consequently, the area under the force–displacement curve was interpreted as the peeling energy required for tearing off the tomato peel. The same methodology was described in [27].

2.4. Peel Drying

Tomato peels were dried in a single layer. Drying experiments were carried out in an infrared dryer with IR lamps (Ballu BIH-l-0.3, Hong Kong, China). An average radiative heat flux of 0.11 W/m^2 with a radiation wavelength of 3 microns was applied. The operating parameters were selected based on the data in the literature regarding the depth of IR radiation penetration into apple tissue (about 10 mm at a wavelength of 3 microns) [28]. During the experiment with IR drying, the moisture loss of tomato peel samples was measured at intervals of 10 min. IR drying was conducted until the final moisture content in the samples reached ~0.06 g/100 g (dry matter). The value of the final moisture content was chosen based on the conditions for further storage of tomato peels. The kinetic curves and drying rates were calculated according to a well-known methodology [29].

2.5. Extraction and Quantification of Total Carotenoids and Total Phenolic Compounds

To study the quality effects of CAP treatment, two intracellular compounds were analyzed as follows: carotenoids and polyphenols from tomato peels. Carotenoids were extracted from dried tomato peels using acetone as solvent. Briefly, 6–7 g of CAP-treated (protocol C) and untreated (protocol D) samples were mixed with 2 mL of acetone and 0.01 g of magnesium carbonate. The extracts were centrifuged for 5–10 min at 3000 rpm (AWTech MPW—260 RH centrifuge, Poland) to separate the supernatant. Three mL of petroleum ether was added to the supernatant to separate the upper layer of the organic phase. The organic phase was transferred to a centrifuge tube with 2 g of predried sodium sulfate.

The content of total carotenoids was estimated spectrophotometrically. The absorbance of extracts was measured at 450 nm. The content of total carotenoids was calculated using the following equation:

$$\mathrm{Totalcarotenoids} = \mathrm{A} \cdot 4.00 \cdot \frac{\mathrm{V}_1}{\mathrm{V}_2} \tag{2}$$

where A is the optical density of the solution of extracted substances; 4.00 is an indicator equal to the ratio of the mass concentration (in milligrams per cubic decimeter) of a β-carotene solution in petroleum ether to its optical density at a wavelength of 450 nm and an optical path length of 10 mm; V_1 is the volume of the test sample of the product; V_2 is the volume of the extract in ether.

The concentration of total polyphenols (mg of gallic acid/kg of tomato product) in the tomato peels was determined according to the method described in [21]. The extraction of tomato waste was carried out with a suitable liquid-to-solid ratio (10:1), allowing the maintenance of a homogenous solid–liquid extraction. Ten grams of tomato peels were mixed with 50% ethanol solution and were agitated for 20 min. Total phenolic compounds were quantified using the Folin–Ciocalteu method [30].

2.6. Energy Aspects of CAP Treatment and Drying

The peeling-specific energy consumption (PSEC) was calculated as follows [13]:

$$W_{PSEC} = \frac{W_{peeling} + W_{CAP} + W_{TE}}{M_{tomato}} \tag{3}$$

where $W_{peeling}$—total energy consumption of the peeling procedure, kW/h; W_{CAP}— total energy consumption of cold plasma pretreatment, kW/h; W_{TE}—energy consumption of thermionic emission source, kW/h; M_{tomato}—the weight of tomatoes, kg.

The values of $W_{peeling}$ were calculated as the area under the force–displacement curve, which was interpreted as the peeling energy required for tearing off the tomato peel. The values of W_{TE} were obtained using a wattmeter (IC-M207D wattmeter, Cartool, Ningbo, China) according to the experimental scheme (Figure 2). The energy consumption of the cold plasma treatment W_{CAP} was calculated based on the volt-ampere characteristics as follows:

$$W_{CAP} = n \cdot \int U(t) \cdot I(t) \cdot dt \tag{4}$$

where n—number of electrical discharges; $U(t)$—instantaneous voltage on the electrodes, V; $I(t)$—discharge current passing through the sample, A.

The drying-specific energy consumption (DSEC) was calculated as follows:

$$W_{DSEC} = \frac{W_{IR} + W_{CAP} + W_{TE}}{M_{peel}} \tag{5}$$

where W_{IR}—total energy consumption of the infrared drying procedure, kW/h; M_{peel}—the weight of the tomato peel, kg.

The values of W_{IR} were obtained using a wattmeter.

2.7. Statistical Analysis

All measurements of the above-mentioned characteristics were performed in at least five replicates. Statistical evaluation of the physical properties was performed via ANOVA, using SigmaPlot (Version 14) with the least significant difference (LSD) at $p < 0.05$. Also, a pair-wise Tukey's test was used to find significant differences between treatments using α 0.05.

3. Results and Discussion

3.1. Improvement in Peeling of Whole Tomato

In our experiments, the untreated and CAP-treated samples were compared. When analyzing the cut of tomato fruits for protocol A, the mesocarp color changed. The white streak between the integument and the endocarp was modified, which indicates a change in the internal mass transfer processes. The same effect was observed for the pulsed electric field treatment of the whole kiwifruit [26].

From the authors' point of view, this phenomenon of peelability is described as follows: moisture starts to migrate from the endocarp region to the zone between the mesocarp and the integumentary tissue via the additionally formed pores in the internal structure of the tomato fruit (Figure 3). In this case, the process of internal mass transfer is carried out due to the emerging turgor intracellular pressure. The resulting layer of liquid, due to its hydrodynamic force, contributes to the effective removal of the integumentary tissue. A similar effect was observed by Andreou et al. [21] in a study on the use of a pulsed electric field when the tomato was peeling.

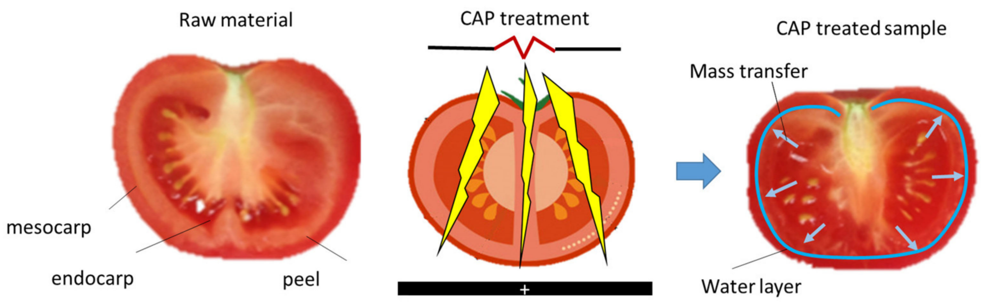

Figure 3. Water migration mechanism in tomatoes caused by cold plasma treatment.

The dynamics of tomato peel removal are presented in Figure 4. For each protocol, the maximum specific peeling force was defined from the graph and is summarized in Table 1. Based on the experimental results, CAP treatment input appeared to significantly influence the tomato peeling procedure. As shown in Figure 4, the peeling procedure of the untreated sample (protocol B) was aborted due to the breakage of the tomato skin. CAP treatment significantly enhanced the peelability of tomato fruits.

Table 1. Comparison of peeling energy between CAP-treated tomatoes (protocol A) and fresh untreated tomatoes (protocol B).

Sample	Specific Peeling Force, N/cm	Peelability, cm^2/g	Energy Consumption of Peeling Procedure $W_{peeling}$, W/kg
Protocol A	18.3 ± 3.35	0.2 ± 0.03	0.065 ± 0.009
Protocol B	52.8 ± 1.12	0.18 ± 0.03	0.170 ± 0.012

The results of our study demonstrated that CAP treatment of whole tomatoes (protocols A and B) produced less specific peeling force as well as energy consumption during the peeling procedure (Table 1). The suggested CAP treatment can be potentially used as an alternative to traditional technologies such as chemical and thermal treatment.

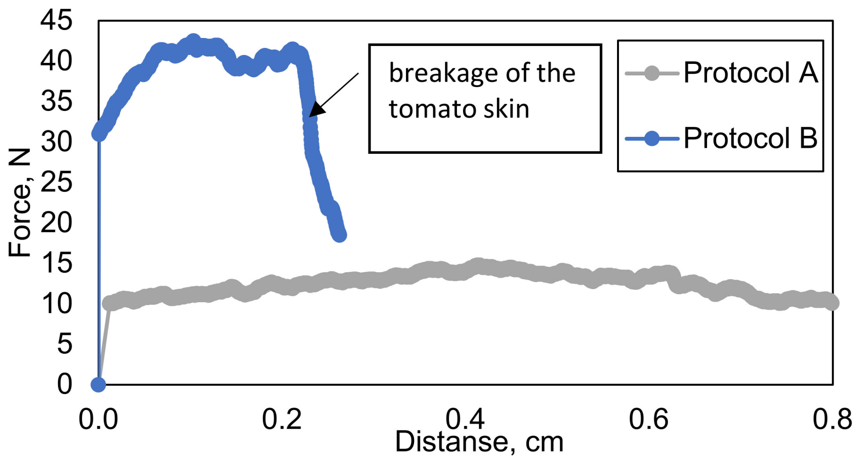

Figure 4. A specific peeling force graph of instrumental tomato peeling analysis for protocols A and B.

Khudyakov et al. [31] compared the efficacy of peeling with pulsed electric field pretreatment (E = 1 kV/cm; specific energy 1, 5, and 10 kJ/kg) for the tomato peeling procedure. It was found that tomato peeling was more effective at 1 kJ/kg. The specific force of mechanical peel removal decreased by 10%. The same PEF technology applied for a tomato peeling procedure resulted in a significant decrease ($p \leq 0.05$) of up to 43% in the force required for mechanical peeling. In comparison with PEF technology, CAP treatment requires less energy. Currently, such methods (PEF and CAP) are still in their developmental stages, requiring optimization and pilot tests before they can be considered for commercialization.

3.2. Improvement in Tomato Peel Drying

Figure 5 shows the kinetics and drying rate of the tomato peels in a thin layer. CAP treatment improved the drying kinetics of tomato peels by forming electrically induced channels with a tree-like structure, which reduced the resistance to moisture transfer during the drying process. A similar result was obtained by the authors Zhang et al., who treated chili peppers with cold plasma and found that the drying time after pretreatment was significantly reduced [32]. A similar explanation for the formation of micro-holes was presented by Zhou et al. when processing wolfberries using CAP treatment [33]. In our experiments, the drying time decreased by 18.2% for tomato peels. The total drying time of the CAP-treated (protocol C) and control (protocol D) samples was 119 and 143 min subsequently. The period of decrease in the diffusion rate, which varied between the control and pretreated samples, was the dominant physical mechanism of the IR drying method [14]. In comparison with control samples (protocol D), where the drying rate reached its peak only after ~10 min, the drying rate of CAP-pretreated tomato peels reached its peak at the very beginning of the drying process. This behavior of the drying rate curve showed that the rate of moisture evaporation from the surface of the sample pretreated by CAP was higher than that which occurred from the inside.

Thus, it can be assumed that with the small-scale production of tomato juice of about 1500 tons of raw materials per day [34], the technology of CAP pretreatment can potentially increase peel-processing capacity by about 10–12%. This can potentially provide additional income, which increases both the rate of return and the return on investment in CAP equipment. Currently, the technologies of CAP pretreatment developed by our research group show great prospects, especially in providing textural and organoleptic characteristics of dried tomato peels. This, in particular, occurs as a result of the use of a stabilized form of electric field distribution.

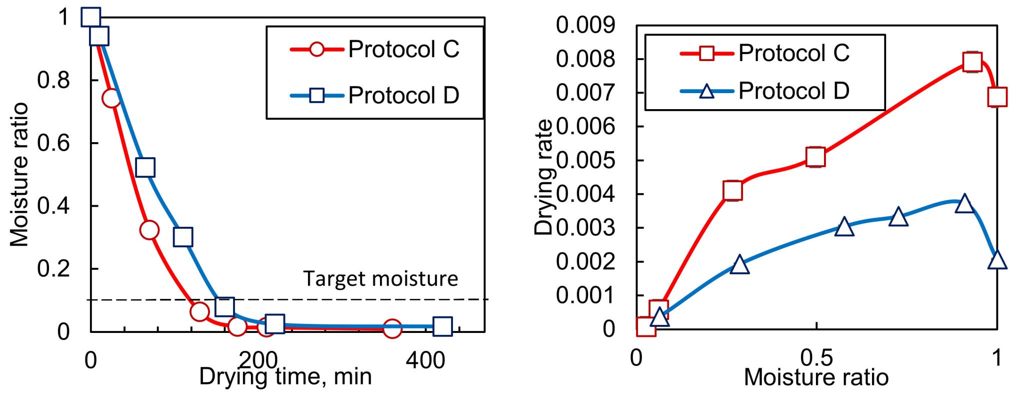

Figure 5. Drying kinetic and drying rate curves of tomato peels for protocols C and D.

3.3. Improvement in Quality Characteristics

Table 2 shows the influence of CAP treatment for protocols C and D on the extraction of high-added-value compounds from tomato peels. Both quality parameters, the total carotenoids and total phenolic compounds, resulted in higher concentrations of all intracellular compounds studied. The results demonstrate the same behavior as discussed by several authors using the pulsed electric field [21] and cold plasma [18]. The electroporation effect might enhance the extraction process due to the better release of these compounds. Andreou et al. [21] reported that PEF treatment doubled the total phenolic compound extraction yield compared to the control. From the authors' point of view, such results might be obtained using electroporation technologies prior to the extraction process. In our experiments, the tomato peels were treated before the drying procedure. The induced pores were used to enhance the drying procedure (Figure 5).

Table 2. Comparison of the drying specific energy consumption of CAP-treated tomato peels (protocol C) and untreated tomatoes (protocol D).

Sample	Total Carotenoids (mg Carotenoids/100 g Tomato Peels)	Total Phenolic Compounds (mg of Gallic Acid/kg Tomato Peels)	The Drying Specific Energy Consumption W_{DSEC}, kW/kg
Protocol C	14.29 ± 3.18	24.43 ± 3.18	3.7 ± 0.21
Protocol D	18.78 ± 2.76	29.51 ± 3.08	4.5 ± 0.33

3.4. Improvement in Energy Consumption

The oscillogram of the current and voltage of the cold plasma discharge is shown in Figure 6. The oscillogram for protocols A and C demonstrates a similar behavior and value due to the high moisture content of the whole tomato as well as tomato peel. The effect of CAP pretreatment on the specific energy consumption of W_{DSEC} for protocols C and D is shown in Table 2. Using Equation (3), an oscillogram of the current and voltage, the value of W_{CAP} was calculated. The total specific energy consumption of W_{PSEC} for CAP-pretreated samples was lower than that of the control samples ($p < 0.05$).

In general, the values of W_{PSEC} for CAP pretreatment depend on the characteristics of the plasma discharge. Since the values of W_{CAP} and W_{TE} are significantly lower than those of W_{IR}, the energy costs for the pretreatment of CAP can be neglected. Potentially, the efficiency of CAP pretreatment can lead to a significant reduction in the energy consumption of the drying procedure of tomato peels. Finally, it was found that the drying-specific energy consumption for CAP-treated samples was 17% lower than that of the control samples.

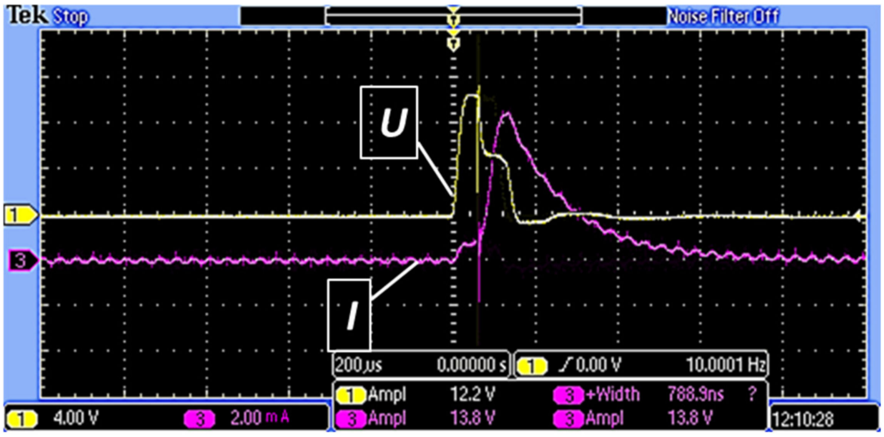

Figure 6. CAP treatment oscillogram of current and discharge.

Santos et al. [35] reported the efficiency of PEF for mango peel drying at E = 4.5 kV cm^{-1} and drying temperature at 70 °C, where the maximum reduction in drying time reached 67%. In comparison with CAP technology, PEF treatment demonstrated a higher effect on peel drying; however, further analysis should be performed.

4. Conclusions

The results obtained in this research showed that cold atmospheric plasma treatment could be applied as a useful tool in the tomato processing industry, leading to decreased energy consumption and increased productivity. Generally, cold atmospheric plasma pre-treatment enhanced the peeling process, including less specific peeling force consumption. In the case of peeling, enhanced peelability can be explained via the migration of water from the mesocarp region under the tomato skin as a result of electroporation. This led to a pressure difference across the tomato skin, reducing the surface resistance and facilitating its removal. Cold atmospheric plasma can easily replace the existing mechanical peeling processes, leading to energy requirements.

Taking into consideration that CAP treatment has low energy requirements (~1.7 kJ/kg of raw material), it could be an economically viable approach for tomato processing industries. CAP treatment could be applied to tomato peel resulting from the tomato processing industry in order to reduce the drying time. The drying-specific energy consumption of the CAP-treated samples was 17% lower than that of the control samples. These could be natural alternatives to high productivity for further tomato peel processing, such as the peel extraction of valuable compounds. In the case of tomato by-product valorization, the efficacy of CAP pretreatment was implemented on tomato waste. CAP treatment could be applied to tomato waste (peel) resulting from the tomato processing industry in order to obtain high extraction yields of carotenoids and phenolic compounds. Future tendencies are directed towards the use of eco-friendly processing technologies to avoid excessive waste generation and promote the circular economy.

Author Contributions: Conceptualization, I.S. and E.H.A.M.; methodology, M.S.; software, E.B.; validation, M.S. and E.B.; resources, I.S.; data curation, E.H.A.M.; writing—original draft preparation, I.S., M.S. and E.H.A.M.; writing—review and editing, I.S.; visualization, M.S.; supervision, I.S.; project administration, I.S.; funding acquisition, I.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was carried out with the financial support of the Kuban Science Foundation and FSBEI HE "KubSTU" under the framework of scientific project No. MFI-P-20.1/40.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data that support the finding of this study are available from the corresponding author upon reasonable request.

Acknowledgments: The authors gratefully acknowledge Claudia Siemer for the useful comments.

Conflicts of Interest: The authors declare no conflict of interest.

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