KSÜ Tarım ve Doğa Derg 27 (1), 216-227, 2024
KSU J. Agric Nat 27 (1), 216-227, 2024
https://doi.org/10.18016/ksutarimdoga.vi.1201966

Determination of Changes in Physicochemical and Microbiological Properties of Tomato Paste Exposed to Different Gases of Cold Plasma Technique

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ABSTRACT
This study aimed to reveal the effect of cold plasma application using different gases and mixtures on some physicochemical and microbiological properties of tomato paste. For this purpose, applications were performed in different gases and times, and the effect of each application was examined separately. As a result of the study, the pH values of the samples varied between 3.77 and 4.87, and the aw values between 0.718 and 0.819. When the color values were examined, it was determined that the L* value varied between 22.42 – 32.48, the a* value varied between 23.59 – 30.18 and the b* value varied between 12.16 – 19.52 (P<0.05). In addition to this, when the samples were evaluated microbiologically, TMAB counts varied between 3.02 – 5.42 log cfu/g, TPAB values ranged between 1.80 – 3.37 log cfu/g, total mold counts were between 3.08 – 5.67 log cfu/g, total yeast counts were between 3.13-5.42 and osmophilic yeast counts were between 1.74 – 3.49 log cfu/g. Lycopene values of samples in the study were in the range of 15.30 – 23.42 mg/100g DM. When the data obtained from the study are evaluated as a whole, it is thought that cold plasma application has positive effects on the shelf life and quality of tomato paste. In the research where two different gases and mixtures of these gases were used, oxygen gas application showed the most effect on the physicochemical and microbiological properties of the samples (P<0.05). Mixture and argon gas applications followed this effect, and prolonging the application period also increased the effect. When the data obtained from the study are evaluated as a whole, it has been revealed that cold plasma application delays the mold problem, which is one of the most critical problems in industrially produced tomato paste, extends the shelf life, and slows down the negative changes in physicochemical quality values due to storage.

Food Science

Research Article

Article History
Received : 09.11.2022
Accepted : 06.07.2023

Keywords
Tomato Paste
Cold Plasma
Shelf Life
Quality

Farklı Dozlarda Soğuk Plazma Tekniğine Maruz Bırakılan Domates Salçasının Fizikokimyasal ve Mikrobiyolojik Özelliklerindeki Değişikliklerin Belirlenmesi

ÖZET
Bu çalışmanın amacı, farklı gaz ve karışımları kullanılan soğuk plazma uygulamasının domates salçasının bazı fizikokimyasal ve mikrobiyolojik özelliklerine etkisinin ortaya koyulmasıdır. Bu amaçla farklı gaz ve sürelerde uygulamalar yapılmış ve her uygulamanın etkisi ayrı ayrı incelenmiştir. Çalışma sonucunda numunelerin pH değerleri 3,77 ile 4,87 arasında, a_w değerleri ise 0,718 ile 0,819 arasında değişmiştir (P<0.05). Renk değerleri incelendiğinde L* değerinin 22.42 – 32.48 arasında, a* değerinin 23.59 – 30.18 arasında ve b* değerinin 12.16 – 19.52 arasında değiştiği tespit edilmiştir (P<0.05). Ayrıca numuneler mikrobiyolojik olarak değerlendirildiğinde depolama süresince tüm örneklerde sayıların arttığı (P>0.05), ancak CP uygulamasının bu artışı ciddi anlamda yavaşlattığı belirlenmiştir.Örneklerin TMAB sayıları 3.02 – 5.42 log kob/g, TPAB değerleri 1.80 – 3.37 log kob/g, toplam küf sayısı 3.08 – 5.67 log kob/g, toplam maya sayısı 3.13 – 5.42 log kob/g ve ozmofilik maya sayıları 1.74 – 3.49 log kob/g arası arasında değişmektedir. Çalışmadaki

Gıda Bilimi

Araştırma Makalesi

Makale Tarihçesi
Geliş Tarihi : 09.11.2022
Kabul Tarihi : 06.07.2023

Anahtar Kelimeler
Domates Salçası
Soğuk Plazma
Raf Ömrü
Kalite

örneklerin likopen değerleri 15.30 – 23.42 mg/100g DM aralığındaydı. İki farklı gaz ve bu gazların karışımlarının kullanıldığı araştırmada örneklerin fizikokimyasal ve mikrobiyolojik özellikleri üzerinde en fazla etkiyi oksijen gazı uygulaması göstermiştir (P<0.05). Bu etkiyi karışım ve argon gazı uygulamaları takip etmiş olup uygulama süresinin uzaması etkiyi de artırmıştır. Çalışmadan elde edilen veriler bir bütün olarak değerlendirildiğinde, soğuk plazma uygulamasının endüstriyel olarak üretilen salçalarda en önemli problemlerden birisi olan küflenme sorunu geciktirerek raf ömrü uzattığı ve depolamaya bağlı fizikokimyasal kalite değerlerinde meydana gelen olumsuz değişimleri yavaşlattığı ortaya konulmuştur.

To Cite : Atik, A., Atik, İ., Akarca, G., & Denizkara, AJ., (2023). Determination of Changes in Physicochemical and Microbiological Properties of Tomato Paste Exposed to Different Doses of Cold Plasma Technique. *KSU J. Agric Nat 27* (1), 216-227. https://doi.org/10.18016/ksutarimdoga.vi.1201966

Atıf Şekli: Atik, A., Atik, İ., Akarca, G., & Denizkara, AJ., (2023) Farklı Dozlarda Soğuk Plazma Tekniğine Maruz Bırakılan Domates Salçasının Fizikokimyasal ve Mikrobiyolojik Özelliklerindeki Değişikliklerin Belirlenmesi. *KSÜ Tarım ve Doğa Derg 27* (1), 216-227. https://doi.org/10.18016/ksutarimdoga.vi.1201966

INTRODUCTION

Tomato is one of the agricultural products with high biological activity because it contains components such as lycopene, ß-carotene, flavonoid and ascorbic acid (Himashree et al., 2022). The antioxidant (Briones-Labarca et al., 2019) and anti-inflammatory properties of these components and the use of tomatoes in the production of products with commercial value such as tomato paste, ketchup and fruit juice increase their importance (Uribe-Wandurraga et al., 2021; Aygel & Aslan, 2023). Tomato paste; is an indispensable component of the cuisines of many countries, it has its unique aroma and taste, tomatoes are obtained by subjecting them to different production processes after being harvested from the field. Since tomato is a perishable product, chemical preservatives must be used in tomato paste (Jafari et al., 2021).

Especially molds are effective in spoiling tomato paste and shortening its shelf life. But today, consumers prefer minimally processed and additive-free products. For this reason, studies on minimal treatments as alternatives to traditional heat treatment methods and chemical preservatives have increased. Today, new technologies are being developed in food production in order to, save time and energy and increase the shelf life of food (Ablay et al., 2020). Atmospheric cold plasma (CP) is one of the minimal processes applied to foods. CP has been already effective in maintaining microbial inactivation in previous studies (Herceg et al., 2016). The aim of this study should be written in detail since the optimization procedure has not been applied in this study. CP has also proved its high inactivation efficiency resulting in the detoxification of mycotoxins produced by fungi (Waghmare, 2021). CP is an emerging non-thermal decontamination technique for microorganisms in food products (Gao et al., 2021; Mao et al., 2021; Wan et al., 2021).

Plasma is accepted as the fourth state of matter (Mir et al., 2016; Varilla et al., 2020; Saremnezhad et al., 2021) produced by energizing gas in an electromagnetic field and consists of reactive species, positive and negative ions, and UV photons (Heo et al., 2021). Plasma is defined as the use of ionized gas in cold sterilization (Fernández et al., 2013). On the other hand, CP is the process that occurs as a result of applying an electric current or electromagnetic radiation to some gases at room temperature under vacuum (Yüksel & Karagözlü, 2017). CP application is effective in the decontamination of vegetative Gram-negative and Gram-positive bacteria, yeasts, viruses, and endospores (Rod et al., 2012; Yüksel & Karagözlü, 2017; Albayrak & Kılıç, 2020).

The general belief about the effect of CP application on microorganisms is related to reactive oxygen which occurs during the process and affects the cell nucleus resulting in DNA damage (Albayrak and Kılıç, 2020). Based on the gas type used, examples of reactive oxygen species (ROS) (Sruthi et al., 2022) species commonly associated with antimicrobial activity and inactivation steps include alkoxyl (RO•), peroxyl (ROO•), hydroperoxyl (HO_2•), superoxide anion (O_2•-), singlet oxygen ($1O_2$), hydroxyl radical (•OH), carbonate anion radical (CO_3•-), hydrogen peroxide (H_2O_2), and ozone (O_3). Similarly, examples of reactive nitrogen species (RNS) include nitrogen dioxide radical (•NO_2), nitric oxide (NO•), alkylperoxynitrite (ROONO), peroxynitrous acid (OONOH), and peroxynitrite (ONOO-) (Misra & Jo, 2017).

Although similar studies were carried out on sun-dried tomatoes before (Molina Hernandez et al., 2022), it was important for the planning of this study that no research was conducted to prevent mold growth, which is the most significant critical problem of industrially produced tomato pastes (especially those with low brix degrees). This study aimed to investigate the effect of CP application, which is described as a minimal process, on the microbiological quality and some physicochemical properties of tomato paste. In this context, two different gases and their mixtures were

applied to tomato pastes at two different effect times, and it was aimed to reveal the changes in the quality values of the samples by determining the changes that occurred during storage.

MATERYAL ve METOD

Tomato Paste

The tomato paste (Brix: 28,14°, Total acidity: 1.71g/100g, Ash insoluble in 10% HCl: 0.19%) used in the research was obtained from the local market in Afyonkarahisar – Turkey. The tomato paste sample used in the research was produced from tomatoes collected in the summer of 2022. When provided, tomato paste is in the original tin can package used by the manufacturer. Before the application, approximately 15 g of tomato paste was spread on sterile Petri dishes to form a thin film in a biological safety cabinet.

The reason for using industrial products in the research was that the quality of industrial pastes (especially low brix degree) deteriorates quickly after opening, especially molding, in terms of microbiology.

Cold Plasma Application

The cold plasma application was modified from the method of Yong et al. (2017) (Fig 1). The gases (Habaş, Turkiye) used in CP application were Argon and Oxygen. The gases were purchased from Afyonkarahisar province (Kocasaban Gazları Corp., Afyonkarahisar, Turkey). The gases used in the system were mixed in certain proportions after that given to the system.

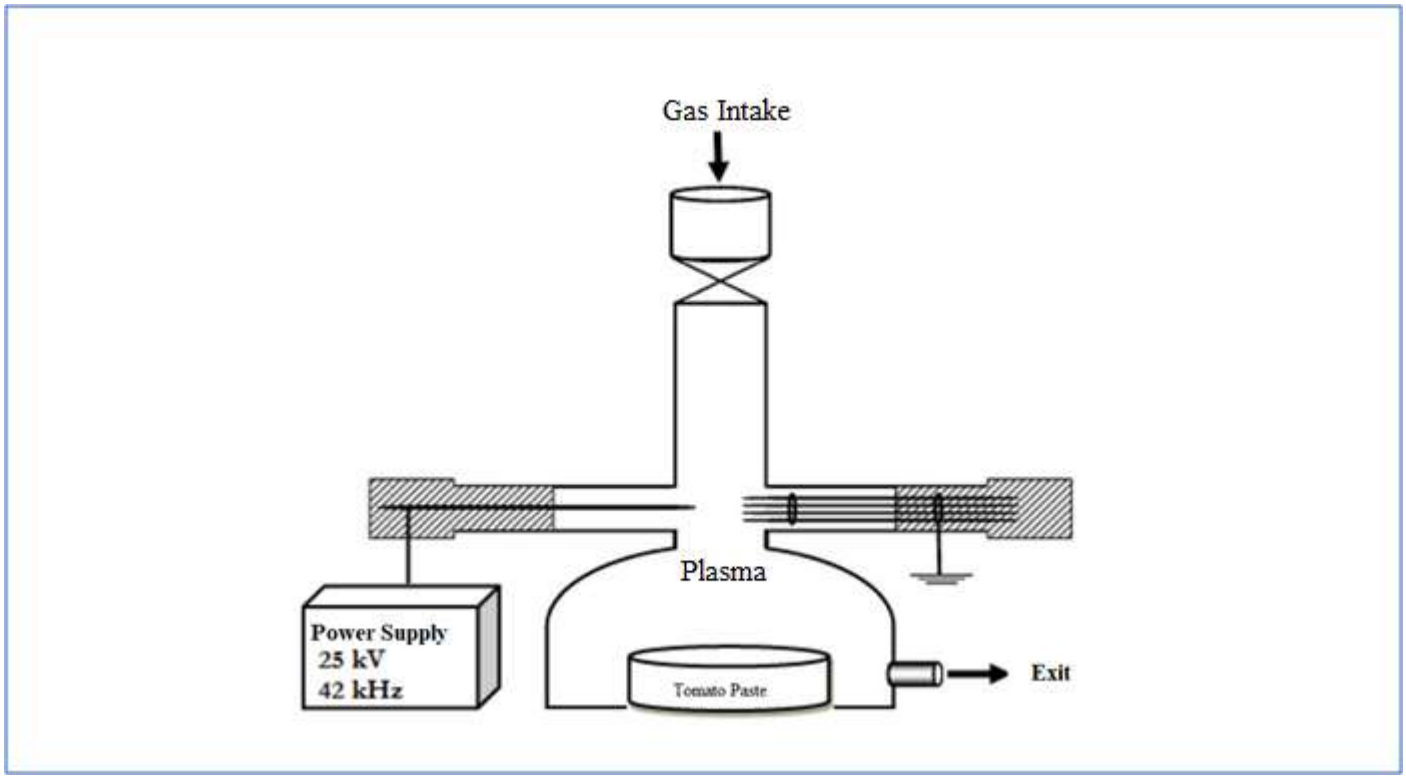

Fig 1. Application of Cold Plasma (Gök et al., 2019)
Şekil 1. Soğuk Plazma Uygulaması (Gök ve ark., 2019)

To maintain the plasma a power supply was used which has a 25 kV potential and 42 kHz frequency. The power supply was designed by a company (Diker Electronics) operating in Afyonkarahisar. The operation was carried out in continuous mode. The media in which the CP application was performed was a semicircular glass chamber with a 28 mm radius. This part is fixed to a plate made of stainless steel by a plastic ring with a diameter of 46 mm.

In order to produce the plasma form, a total of seven 1 mm thick tungsten steel electrodes (Astor, Türkiye) were used. One of these electrodes was placed horizontally in front of the other six electrodes in order to create plasma between the anode-cathode tips. The temperature of the tomato paste samples during the application was measured consistently using an infrared thermometer (Coleman-Parmer, Vernon Hills, IL).

In the research, only oxygen gas plasma was applied to FO1, and FO2 coded samples, only argon gas plasma was applied to FA1, and FA2 coded samples, and 50% oxygen + 50% argon was applied to MOA1 and MOA2 coded samples. Applied gases, mixing ratios, and flow rates are shown in Table 1. The exposure times of the samples to the gases were determined as 20 and 30 minutes. Each application was performed in 3

parallels, separately and on different days.

The processing time was determined due to the preliminary trials made before. Since the further extension of the application period causes severe quality losses in the product, it was kept at these levels. After the procedure, the samples were stored in a closed, sterile glass petri dish under aerobic conditions in the refrigerator at 4°C.

Table 1. Samples and coding used in the research
Çizelge 1. Araştırmada kullanılan örnekler ve kodlamaları

Samples	Processing/Time	Code
1	Control	C
2	%100 O_2/20 min.	FO1
3	% 100 O_2/30 min.	FO2
4	%100 Ar/20 min.	FA1
5	%100 Ar/30 min.	FA2
6	%50 O_2 and %50 Ar/20 min.	MOA1
7	%50 O_2 and %50 Ar/30 min.	MOA2

Microbiological Analysis

Preparation of samples for microbiological analysis

After the tomato paste samples were exposed to CP, 10 g of each sample was weighed on a precision balance (Laboratory Balances, Radwag PS R2.H, Poland) and put into another sterile stomacher bag. Sterile Ringer's solution with a volume of 90 mL (Merck, 115525, Germany) was added to it and homogenized for 2 minutes in a stomacher (BagMixer® 400 P-080921247). Then, the mixture was diluted with Ringer's solution by preparing serial dilutions at the desired ratios. The same procedures were applied in the control group. The control group consisted of untreated samples (Anonymous 2001).

Total mesophilic aerobic bacteria count (TMAB)

Plate Count Agar (Oxoid, CM0325) was used for total aerobic mesophilic bacteria count. It was incubated at 30 °C for 72 hours under aerobic conditions, and the total number of aerobic mesophilic bacteria was determined by counting the colonies that developed at the end of the incubation (ISO 2013a; ISO 2013b).

Total yeast count

Total yeast count was determined on Yeast Exctract Agar (Oxoid, CM0019) after incubation in aerobic conditions for 5-7 days at 30 °C. At the end of the incubation, the colony-forming units were counted and the results were calculated as viable cfu/mL (ISO 2008).

Total mold count

Total mold count was determined on Malt Exctract Agar (Merck, 1.05398) after incubation in aerobic conditions for 5-7 days at 30 °C. At the end of the incubation, the colony-forming units were counted and the results were calculated as viable cfu/mL (ISO 2008).

Osmophilic yeast count

Total mold count was determined on DG18 medium modified with gliserol (Merck, 1.00465) after incubation in aerobic conditions for 5-7 days at 30 °C. At the end of the incubation, the colony-forming units were counted and the results were calculated as viable cfu/mL (ISO 2008).

Physicochemical Analysis

The color values, pH and water activity (aw) of tomato paste samples were examined.

pH value

A 10 g tomato paste sample was mixed with 10 mL distilled water and homogenized (Daihan Wisestir, HS-30T, South Korea). To measure the pH values of the prepared mixtures a pH meter (HANNA, HI 2215 pH/ORP meter) was used (AOAC 2016).

Activity of water (a_w) value

To determine the aw values of the samples a water activity analyzer (Novasina LabTouch-aw, Lachen, Switzerland) was used (AOAC 2016).

Color analysis

Color values of tomato paste samples were determined using a colorimeter (Konica Minolta Chroma Meter CR-400, Osaka). The brightness (L*), redness (a*) and yellowness (b*) values of the samples were measured according to Akarca (Akarca, 2013). The Brown Index value was calculated according to the formulas below (Kurtuldu & Özcan, 2018).

$$x = [a + \frac{1,75 * L}{5,645 * L} + (a - (3,012 * b)]$$

$$BI = [100 * \frac{x - 0,31}{0,17}]$$

Lycopene Analysis

Lycopene analysis was conducted by using HPLC according to the method of Demiray et al. (2013).

Statistical Analysis

The results obtained in the study were made in two parallels and SPSS software program V 23.0.0 was used for the variance analysis. A significant difference was determined by Duncan's multiple range tests (*P<0.05) (Atik & Gümüş, 2021).

RESULTS and DISCUSSION

TAMB counts of the samples increased during storage (P<0.05). When the TMAB results were examined, it was determined that CP application provided 1 log reduction. An increase in TMAB values was observed in the 14-day shelf life. It was determined that the rate of increase was less in the samples treated with CP. Changes in TMAB count of samples are given Table 2.

Although TPAB numbers increased during 14 days of storage in all samples (P<0.05), CP application was effective in TPAB values of tomato paste samples (P<0.05). As a result of the study, a reduction of 2 logs was achieved. The maximum microbial decrease was achieved in the MOA2 sample. An increase was observed in the TPAB counts of the samples during the storage period. While this increase was not significant for control and FO1, it was significant for other CP applications (P<0.05). The TPAB counts of samples are given in Table 3.

Table 2. Changes in TMAB count of tomato paste samples during storage (log cfu/g)
Çizelge 2. Domates salçası örneklerinin depolama süresince TMAB sayısındaki değişiklikler (log kob/g)

Sample	TMAB Count		
	Storage Days		
	0	7	14
C	4.30 ± 0.13^{Ca}	4.79 ± 0.02^{bA}	5.42 ± 0.03^{aA}
FO1	3.23 ± 0.12^{Bd}	3.15 ± 0.04^{bCD}	3.66 ± 0.02^{aE}
FO2	3.16 ± 0.01^{Bd}	3.02 ± 0.01^{cC}	3.35 ± 0.03^{aF}
FA1	3.36 ± 0.10^{Bcd}	3.11 ± 0.03^{cDE}	4.21 ± 0.05^{aB}
FA2	3.51 ± 0.15^{Cc}	3.60 ± 0.01^{bB}	4.01 ± 0.02^{aC}
MOA1	3.98 ± 0.01^{Ab}	3.22 ± 0.02^{bF}	3.90 ± 0.04^{aD}
MOA2	3.90 ± 0.14^{Ab}	3.04 ± 0.02^{cEF}	3.75 ± 0.04^{bE}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)F: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05) TMAB: Total Mesophilic Aerobic Bacteria

Table 3. Changes in TPAB count of tomato paste samples during storage (log cfu/g)
Çizelge 3. Domates salçası örneklerinin depolama süresince TPAB sayısındaki değişiklikler (log kob/g)

Sample	TPAB Count		
	Storage Days		
	0	7	14
C	2.82 ± 0.38^{aA}	3.37 ± 0.01^{aA}	3.24 ± 0.05^{aA}
FO1	2.31 ± 0.09^{aAB}	2.44 ± 0.04^{aC}	2.65 ± 0.09^{aBC}
FO2	2.00 ± 0.02^{bAB}	2.10 ± 0.02^{bD}	2.40 ± 0.04^{aC}
FA1	2.12 ± 0.03^{bAB}	2.75 ± 0.08^{aB}	3.11 ± 0.19^{aAB}
FA2	1.96 ± 0.10^{bB}	2.72 ± 0.04^{aB}	2.97 ± 0.10^{aAB}
MOA1	1.85 ± 0.19^{bB}	2.76 ± 0.02^{aB}	2.93 ± 0.12^{aAB}
MOA2	1.80 ± 0.05^{bB}	2.51 ± 0.05^{aC}	2.96 ± 0.18^{aAB}

a(→)b: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)D: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05) TPAB: Total Psychrophilic Aerobic Bacteria

Mold, yeast, and osmophilic yeast results are given in Table 4, 5, and 6, respectively. Yeast, mold, and osmophilic yeast counts increased during storage in all samples (P<0.05). A 1 log reduction was achieved in the count of mold, which is an important deterioration factor in tomato paste, depending on the gas composition applied and the processing time. During the storage period, especially on the 14th day, the total mold rate increased significantly (P<0.05).

CP application was effective on total yeast count (P<0.05). No significant difference was detected between applied gas concentrations and treatment times (P>0.05). A 0.82 log reduction was achieved with CP application. During the 14-day storage period, while the total count of yeasts increased by 2 log in the control sample, the increase remained below 1 log in the samples treated with CP. In this sense, CP application was effective in the increase rate of microorganisms.

Table 4. Changes in mold count of tomato paste samples during storage (log cfu/g)
Çizelge 4. Domates salçası örneklerinin depolama süresince küf sayısındaki değişiklikler (log kob/g)

Sample	Total Mold Count		
	Storage Days		
	0	7	14
C	4.19±0.12^{bA}	4.25±0.06^{bA}	5.67±0.03^{aA}
FO1	3.34±0.14^{bCDE}	3.34±0.07^{bD}	4.66±0.03^{aB}
FO2	3.08±0.08^{bE}	3.17±0.11^{bD}	4.53±0.02^{aC}
FA1	3.65±0.03^{cBC}	3.99±0.00^{bB}	4.20±0.04^{aD}
FA2	3.27±0.18^{bDE}	3.79±0.04^{aC}	4.14±0.02^{aDE}
MOA1	3.75±0.02^{bB}	3.74±0.03^{bC}	4.16±0.03^{aDE}
MOA2	3.47±0.02^{cBCD}	3.72±0.01^{bC}	4.10±0.02^{aE}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)E: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

Table 5. Changes in yeast count of tomato paste samples during storage (log cfu/g)
Çizelge 5. Domates salçası örneklerinin depolama süresince maya sayısındaki değişiklikler (log kob/g)

Sample	Total Yeast Count		
	Storage Days		
	0	7	14
C	3.95±0.10^{bA}	5.23±0.11^{aA}	5.42±0.00^{aA}
FO1	3.21±0.13^{aB}	3.32±0.06^{aD}	3.52±0.01^{aDE}
FO2	3.13±0.08^{bB}	3.25±0.05^{bD}	3.48±0.01^{aE}
FA1	3.41±0.06^{bB}	3.50±0.01^{abBC}	3.60±0.05^{aBC}
FA2	3.21±0.10^{bB}	3.35±0.04^{abCD}	3.56±0.10^{aCD}
MOA1	3.41±0.03^{bB}	3.58±0.02^{aB}	3.66±0.05^{aB}
MOA2	3.39±0.07^{aB}	3.50±0.11^{aBC}	3.58±0.04^{aC}

a(→)b: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)E: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

Table 6. Changes in osmophilic yeast count of tomato paste samples during storage (log cfu/g)
Çizelge 6. Domates salçası örneklerinin depolama süresince ozmofilik maya sayısındaki değişiklikler (log kob/g)

Sample	Osmophilic Yeast Count		
	Storage Days		
	0	7	14
C	3.14±0.05^{bA}	3.37±0.02^{aA}	3.49±0.02^{aA}
FO1	2.05±0.04^{aC}	2.16±0.13^{aB}	2.34±0.01^{aB}
FO2	1.74±0.04^{cE}	2.07±0.05^{bB}	2.30±0.03^{aB}
FA1	2.21±0.02^{bB}	2.28±0.01^{aBB}	2.34±0.03^{aB}
FA2	2.05±0.01^{bC}	2.19±0.06^{aBB}	2.31±0.01^{aB}
MOA1	2.00±0.02^{bCD}	2.16±0.04^{aBB}	2.30±0.09^{aB}
MOA2	1.91±0.02^{bD}	2.13±0.03^{aB}	2.23±0.06^{aB}

a(→)b: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)E: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

Similarly, approximately 1.4 log reduction was achieved in the count of osmophilic yeasts, depending on the gas applied and the application time. An increase in the count of osmophilic yeasts was observed during the storage period.

The disruption of the cell structure and the resulting microbial inactivation depend on the exposure time to the cold plasma application and the type of gas used for disinfection (Ganesan et al., 2021). Reactive species such as reactive oxygen species (ROS) and reactive nitrogen species (RNS) are significant in the inactivation mechanism of pathogens (Asl et al., 2022). ROS produced during plasma production cause strong oxidative stress, and cells are damaged by enzyme inactivation, lipid peroxidation, and DNA cleavage. In addition, RNS is highly toxic and can cause cell death by damaging DNA (Heo et al., 2021). During the plasma application, the oxygen in the air plasma causes the formation of peroxide and the formation of lethal species such as O*, O2, and O3. In addition, the

accumulation of charged particles on the outer cell membrane, and electrostatic forces could cause subsequent rupture of the cell membrane and subsequent cell death (Devi et al., 2017). DNA damage caused by UV radiation produced during plasma is also thought to be effective in microbial inactivation (Liao et al., 2017).

Akarca et al. (2023) reported that CP application significantly reduced mold growth in kosher cheese. In addition, Molina-Hernandez et al. (2022) reported that CP application significantly slowed spore germination on the growth of mold spores in sun-dried tomatoes. Similarly, Ulbin-Figlewicz et al. (2015) stated that CP application using helium gas reduced the number of yeast and mold on the meat surface by 2 logs. Devi et al. (2017) revealed that CP application to peanuts largely stopped the growth of two important molds, A. flavus and A. parasiticus, in aflatoxin production.

Although the pH values of all samples decreased during storage ($P<0.05$), in general, CP application decreased the pH value (Table 7). The increase in H ions during CP application is thought to be effective in this decrease in pH value. Reactive species produced by plasma, mainly with acidic properties such as nitric acid (HNO3) and nitrous acid (HNO2), are responsible for the pH decrease (Wang et al., 2022). The pH of tomato paste samples decreased during storage. The highest pH decrease was observed in the MOA2 sample.

Table 7. pH changes of tomato paste samples during storage
Çizelge 7. Domates salçası örneklerinin depolama süresince pH değişiklikleri

Sample	pH		
	Storage Time (Day)		
	0	7	14
C	4.39±0.07^{aB}	4.06±0.01^{bF}	3.95±0.06^{bA}
FO1	4.13±0.08^{bC}	4.34±0.02^{aD}	3.87±0.01^{cAB}
FO2	3.99±0.02^{bCD}	4.17±0.05^{aE}	3.84±0.01^{cAB}
FA1	4.57±0.04^{aA}	4.47±0.02^{aC}	3.91±0.09^{bAB}
FA2	3.90±0.02^{bD}	4.33±0.03^{aD}	3.78±0.04^{bAB}
MOA1	4.52±0.03^{bAB}	4.72±0.03^{aB}	3.77±0.05^{cB}
MOA2	3.89±0.04^{bD}	4.87±0.04^{aA}	3.79±0.07^{bAB}

a(→)b: Values with the same capital letters in the same rows for each analysis differ significantly ($P<0.05$)
A(↓)F: Values with the same capital letters in the same column for each analysis differ significantly ($P<0.05$)

The a_w value of tomato paste samples (Table 8) decreased both with CP application and during storage ($P<0.05$). This decrease in aw is thought to be caused by the drying on the surface due to the flow rate of the gas applied during the process. Lee et al. (2020) applied cold plasma to red pepper flakes and reported a similar decrease in a_w. The decrease in a_w could be attributed to the ability of O_2 and Ar gases used in plasma application to take up free water molecules on the tomato paste surface.

Table 8. a_w changes of tomato paste samples during storage
Çizelge 8. Domates salçası örneklerinin depolama süresince a_w değişiklikleri

Sample	a_w		
	Storage Time (Day)		
	0	7	14
C	0.819±0.03^{aA}	0.797±0.03^{bA}	0.788±0.04^{bA}
FO1	0.808±0.02^{aB}	0.740±0.02^{bC}	0.739±0.04^{bB}
FO2	0.798±0.01^{aC}	0.739±0.02^{bC}	0.731±0.02^{cBC}
FA1	0.801±0.02^{aC}	0.742±0.08^{bBC}	0.734±0.03^{bBC}
FA2	0.733±0.01^{aE}	0.735±0.03^{aC}	0.718±0.03^{bD}
MOA1	0.808±0.03^{aB}	0.748±0.02^{bB}	0.727±0.04^{cCD}
MOA2	0.741±0.01^{aD}	0.739±0.01^{aC}	0.734±0.01^{aBC}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly ($P<0.05$)
A(↓)E: Values with the same capital letters in the same column for each analysis differ significantly ($P<0.05$)

Since the plasma treatment is applied only to the product's surface, the chemical reaction takes place on the product's surface. The presence of a chemical reaction can be traced from the change in color, texture, or aroma (Bermúdez-Aguirre et al., 2013). Both CP application and storage process caused an increase in L* value (Table 9) ($P<0.05$). The highest L^* value was determined in the MOA2 sample. In non-thermal applications such as CP, the free radicals formed due to the low-temperature processing of the product and the liberation of intracellular compounds during the application were effective in increasing the product's brightness (Mehta et al., 2019).

Table 9. Changes in L*values of tomato paste samples during storage
Çizelge 9. Domates salçası örneklerinin depolama süresince L değerlerindeki değişiklikler*

Sample	*L** Value		
	Storage Days		
	0	7	14
C	22.24±0.72^{bC}	25.80±1.29^{bB}	30.99±0.04^{aD}
FO1	24.82±1.12^{bBC}	30.22±1.17^{aA}	30.41±0.02^{aE}
FO2	24.32±0.02^{cBC}	28.92±0.36^{bA}	30.12±0.15^{aF}
FA1	29.67±0.07^{bA}	29.99±0.12^{bA}	30.79±0.09^{aD}
FA2	29.31±0.02^{cA}	30.74±0.09^{bA}	31.67±0.07^{aB}
MOA1	29.64±0.48^{bA}	30.59±0.08^{aA}	31.37±0.05^{aC}
MOA2	28.76±1.97^{bAB}	29.94±0.17^{aBA}	32.48±0.03^{aA}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)F: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

CP application and storage time caused a decrease in *a** value (Table 10), but this decrease was not statistically significant (P>0.05). The lowest *a** value was measured in the MOA1 sample. These results are in accordance with Jayasena et al. (2015). Bermúdez-Aguirre et al. (2013) applied atmospheric cold plasma to fresh tomatoes and stated that the *a** value increased after 7 and 10 minutes of application, but this increase was not statistically significant. Similarly, *b** values (Table 11) of tomato paste samples decreased with CP application. Jiang et al., (2017), and Khani et al., (2017) reported that cold plasma application to tomatoes did not cause a significant change in color. It is thought that there is a decrease in *a** and *b** values due to the loss of phenolic components (Mehta et al., 2019). The changes in these color parameters could be a result of oxidation of both pigments and lipids (Olatunde et al., 2019).

Table 10. Changes in a*values of tomato paste samples during storage
Çizelge 10. Domates salçası örneklerinin depolama süresince a değerlerindeki değişiklikler*

Sample	*a** Value		
	Storage Days		
	0	7	14
C	28.28±0.21^{aAB}	28.18±0.07^{aA}	27.29±0.02^{bA}
FO1	28.45±0.22^{aAB}	27.81±0.74^{aAB}	27.36±0.29^{aA}
FO2	30.18±1.99^{aA}	27.57±1.97^{aAB}	26.61±0.27^{aB}
FA1	26.55±0.81^{aB}	26.18±0.05^{aAB}	25.34±0.04^{aC}
FA2	29.36±0.02^{aAB}	25.99±0.87^{bAB}	23.87±0.05^{bD}
MOA1	26.56±0.04^{aB}	25.12±0.03^{bAB}	23.59±0.04^{cD}
MOA2	26.41±0.54^{aB}	25.05±0.02^{abB}	23.81±0.13^{bD}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)D: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

The lycopene results of the samples are given in Table 12. A decrease in lycopene values was observed after 14 days of storage. However, this change is not statistically significant (P > 0.05). It should be noted that only for FO1 the change between day 7 and day 14 of storage was significant (P < 0.05). Cold plasma application caused a significant change in the lycopene values of the samples (P < 0.05). A decrease in lycopene values was observed. In particular, a decrease occurred in the samples that applied 100% oxygen (FO1 and FO2), and the decrease in lycopene values was more remarkable as the application time increased.

The BI indexes of the samples decreased during storage (P<0.05). At the end of storage, the highest decrease was detected in the FO2-coded sample with a value of -3284.84, while the least decrease was detected in the MOA2-coded samples with a value of 4792.79 (Table 12)

The main reason for the decrease in the amount of lycopene is that it has a highly unsaturated structure and as a result, oxidation occurs by photooxidation or auto-oxidation. The colorless end products formed from these oxidative reactions may cause the red color to bleach or lighten (Jayathunge et al., 2019). The leading causes of lycopene degradation at elevated temperatures are isomerization and oxidation. In addition, environmental factors such as weather, light, and temperature can change the effect of these two processes on the lycopene content of tomato products (Jabbari et al., 2018). In cold plasma application, the mechanism responsible for the change in the amount of lycopene is oxidation. It should be stated that the decrease in the lycopene value, especially in the samples using 100% O_2, is due to the increase in oxidation.

Table 11. Changes in b*values of tomato paste samples during storage
Çizelge 11. Domates salçası örneklerinin depolama süresince b değerlerindeki değişiklikler*

Sample	*b** Value		
	Storage Days		
	0	7	14
C	15.93±0.17^{bA}	16.98±0.22^{aBA}	17.94±0.35^{aAB}
FO1	14.48±0.03^{cB}	15.61±0.56^{bAB}	16.93±0.17^{aAB}
FO2	14.24±0.02^{aB}	15.60±1.29^{aAB}	19.52±2.45^{aA}
FA1	13.34±0.04^{bC}	16.39±0.35^{aBAB}	17.20±1.38^{aAB}
FA2	12.16±0.03^{cE}	14.71±0.70^{bB}	16.26±0.07^{aABC}
MOA1	12.65±0.09^{bD}	12.59±0.35^{bC}	14.47±0.05^{aBC}
MOA2	12.20±0.10^{cE}	12.40±0.06^{bC}	13.11±0.02^{aC}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)E: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

Table 12. Changes in Brown Index(BI) values of tomato paste samples during storage
Çizelge 12. Domates salçası örneklerinin depolama süresince Esmerleşme İndeksindeki (Eİ) değişiklikler

Sample	BI Index		
	Storage Days		
	0	7	14
C	5046.38±775.35^{aC}	3071.35±646.86^{aC}	314.546±852.02^{bAB}
FO1	7815.44±315.92^{aBC}	5054.52±180.26^{bB}	2195.24±60.74^{cAB}
FO2	10275.96±3260.81^{aAB}	4795.77±45.35^{abB}	-3284.84±5697.94^{bB}
FA1	7599.95±1447.88^{aBC}	1763.75±162.56^{abD}	-653.72±3511.83^{bAB}
FA2	13005.38±29.37^{aA}	4519.69±1263.77^{bB}	-717.67±79.67^{cAB}
MOA1	8834.24±192.23^{aB}	7255.28±37.78^{bA}	2118.48±54.52^{cAB}
MOA2	9455.06±923.49^{aB}	7500.71±100.43^{bA}	4792.79±195.35^{cA}

a(→)c: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)C: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

Table 13. Changes in lycopene values of tomato paste samples during storage
Çizelge 13. Domates salçası örneklerinin depolama süresince likopen değerlerindeki değişiklikler

Sample	Lycopene mg/100g DM		
	Storage Time (Day)		
	0	7	14
C	23.42±0.26^{aA}	23.20±0.12^{aA}	22.67±0.09^{aA}
FO1	19.58±0.11^{aE}	19.27±0.06^{aF}	18.91±0.04^{bD}
FO2	15.96±0.09^{aF}	15.82±0.10^{abG}	15.30±0.16^{bE}
FA1	22.03±0.10^{aB}	21.96±0.08^{abB}	21.60±0.07^{bB}
FA2	21.75±0.13^{aB}	21.63±0.09^{abC}	21.22±0.11^{bB}
MOA1	20.61±0.12^{aC}	20.44±0.11^{aD}	19.89±0.23^{aC}
MOA2	20.05±0.07^{aD}	19.83±0.06^{abE}	19.21±0.22^{bD}

a-b: Values with the same capital letters in the same rows for each analysis differ significantly (P<0.05)
A(↓)F: Values with the same capital letters in the same column for each analysis differ significantly (P<0.05)

In the study where two different gases and mixtures of these gases were used, oxygen gas application showed the most effect on the physicochemical and microbiological properties of the samples compared to the control sample. Mixture and argon gas applications followed this effect, and prolonging the application period also increased the effect.

CONCLUSION

Tomato paste, one of the indispensable components of many cuisines in the world, is a product especially sensitive to mold growth. In this study, the effect of CP application, which is a minimal treatment, on the microbiological quality of tomato paste was investigated. CP application caused a decrease in pH and aw values of tomato paste. While *L** value increased, *a** and *b** values decreased in the samples. For microbiological evaluation, TMAB, TPAB, total mold, total yeast and osmophilic yeast values were examined. A decrease of 2 log in TPAB value and 1 log

in other microbiological criteria was achieved. The most effective application was determined as the application coded MOA2.

Different brix ranges are produced by companies in the production of industrial tomato paste. Especially a low brix degree is preferred, especially for lower-cost products. As a result, especially after the product is opened, high water activity leads to rapid molding, significantly shortening its shelf life.

Parallel to the results obtained from similar studies on the subject, it was demonstrated in this study that CP application could be used effectively to prevent or at least delay the mold problem, which is one of the biggest problems in tomato paste production. Further studies on the subject should be planned to reveal the positive/negative biochemical and chemical effects of CP application on the product in more detail.

Credit Contribution of the Authors

Azize Atik: Conceptualization (Equal), Writing – Original Draft Preparation (Lead).

İlker Atik: Conceptualization (Equal), Writing – Review & Editing (Lead).

Gökhan Akarca: Methodology (Lead), Formal analysis (Lead).

Ayşe Janseli Denizkara: Investigation (Lead).

Declaration of competing interest

The authors declare that they do not have any competition and any conflicts of interest.

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