Effects of thermal processing by nanofluids on vitamin C, total phenolics and total soluble solids of tomato juice

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Abstract In this research, our main idea was to apply thermal processing by nanofluids instead of conventional pasteurization processes, to shorten duration of thermal procedure and improve nutritional contents of fruit juices. Three different variables of temperature (70, 80 and 90 °C), nanofluid concentration (0, 2 and 4%) and time (30, 60 and 90 s) were selected for thermal processing of tomato juices by a shell and tube heat exchanger. The results demonstrated that 4% nanofluid concentration, at 30 °C for 30 s could result in 66% vitamin C retention of fresh juice while it was about 56% for the minimum nanofluid concentration and maximum temperature and time. Higher nanoparticle concentrations made tomato juices that require lowered thermal durations, because of better heat transfer to the product, and total phenolic compounds dwindle less severely; In fact, after 30 s thermal processing at 70 °C with 0 and 4% nanoparticles, total phenolic compounds were maintained by 71.9 and 73.6%, respectively. The range of total soluble solids for processed tomato juices was 5.4–5.6, meaning that nanofluid thermal processing could preserve the natural condition of tomato juices successfully. Based on the indices considered, a

nanofluid thermal processing with 4% nanoparticle concentration at the temperature of 70 °C for 30 s will result in the best nutritional contents of final tomato juices.

Keywords Tomato juice · Nano-fluids · Heat exchanger · Total phenolics · Vitamin C

Introduction

In food factories, heating and cooling food products, especially for pasteurization and sterilization, are carried out by heat exchangers. Plate heat exchangers and shell and tube heat exchangers are most conventional types deployed for thermal processing of food products, especially in milk and beverage industries. Shell and tube heat exchangers are comprised of tubes with circular cross sections installed in large cylindrical shells. In these exchangers, baffles (blades and plates for transferring flows) are used to increase heat transfer coefficients on shell side and to hold tubes. Selecting a specific arrangement inside the shell and tube is dependent on several factors e.g. heat capacity, pressure drop, pressure area, fouling formation, constructing and costing methods, erosion control, and cleaning issues.

In fruit juice factories, hot water or water steam are usually used as thermal media in heat exchangers for HTST/LTLT pasteurization (72 °C for 15 s and 63 °C for 30 min, respectively). Restricted thermal capacity and properties of conventional heating fluids and their low heat transfer efficiencies results in color changes, particles separation and changes in aroma, flavor or odor of food products due to extended thermal processing time or temperature (Shin and Bhowmik 1995). Furthermore, low efficiency of heat transfer from heating fluids to food liquids affects energy consumption, operational costs and

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final price as well as food quality. So, intensifying efficiency of heat transfer is one of important methods for improvement of product quality and optimization of energy consumption in pasteurization unit operations and food factories. On the other hand, most solids, especially metals, have higher thermal conductivities compared with conventional fluids. When solid particles, particularly at nano-sizes (lower than 100 nm), are added to base fluids, a stable suspension called nanofluid is formed, which has higher thermal conductivities than base fluids. Accordingly, nanofluids have high potentials in improving heat transfer and are appropriate substitutions for conventional fluids. So, researches have recently started to inspect cutting-edge characteristics of nanofluids and their application in diverse fields. Palm et al. (2006) studied heat transfer enhancement with the use of nanofluids in radial flow cooling systems considering temperature-dependent properties; the results clearly indicated that considerable heat transfer enhancement could be obtained by using these fluid/solid particle mixtures. Nanoparticles volume fraction of only 4% in water/ Al_2O_3 nanofluid could result in 25% increase in the average wall heat transfer coefficient of the base fluid (water). Farajollahi et al. (2010) studied heat transfer characteristics of $\gamma\text{-Al}_2\text{O}_3$ /water and TiO_2 /water nanofluids in a shell and tube heat exchanger under turbulent flow condition, and investigated the effects of Peclet number, volume concentration of suspended nanoparticles, and particle type on the heat characteristics. Based on their results, addition of nanoparticles to the base fluid caused a significant enhancement of heat transfer characteristics. In detail, at a certain Peclet number, heat transfer characteristics of TiO_2 /water nanofluid were greater than those of Al_2O_3 /water nanofluid at their optimum nanoparticle concentrations while the latter nanofluid led to the better heat transfer features at higher nanoparticle concentrations. Zeinali Heris et al. (2011) investigated laminar flow-forced convective heat transfer of Al_2O_3 /water nanofluid in a triangular duct under constant wall temperature condition. The results indicate that the addition of nanoparticles to base fluid, besides the thermal conductivity increment, affects the structure of the flow field and leads to heat transfer enhancement. Their results showed that increasing the nanoparticles concentration increased Nusselt number at a constant particle size. They concluded that in the case of using triangular cross-sectional ducts in thermal engineering systems, because of their resulting low pressure drops, the decrement of heating exchange rate could be compensated by the use of nanofluids in these systems. Mare et al. (2011) compared thermal performances of two types of commercial nanofluids alumina (Al_2O_3) and carbon nanotubes (CNTs) dispersed in water under a laminar out-flow mode. They reported that the gain could reach 22% for alumina and 150% for carbon nanotubes, and that

both nanoparticles showed a better thermal-hydraulic performance in terms of balance between heat transfer enhancement and pumping power loss in comparison with pure water. Pandey and Nema (2012) surveyed effects of nanofluid (Al_2O_3 in water 2, 3 and 4 vol%) and water as coolants on heat transfer, frictional losses, and energy loss in a counter flow corrugated plate heat exchanger. For a given heat load, the required pumping power increased with increase in nanofluid concentration. Both power consumption and heat transfer rates were lower for water in comparison to the nanofluid for flow rates of 2–5 lpm for hot and cold fluids. These researches demonstrate that heat transfer enhancement is guaranteed by incorporating low concentrations of nanoparticles in heating base fluids.

Tomato, with the scientific name of *Lycopersicon esculentum*, is from *Solanaceae* family. Financially, tomato is the second important produce, after potato, among all fruits and vegetables around the world. In Iran, tomato allocates forth in production rate among all food produces to itself after wheat, milk and sugar, respectively. Tomato is one of favorite vegetables that contains chief nutritional compounds as well as functional compounds and antioxidants such as lycopene, β -carotene, α -carotene, cryptoxanthin, lutein, and other flavonoids, phenolic acids and ascorbic acid (Odriozola-Serrano et al. 2009). Tomato contains 94.5% water and its (approximate amount of) main nutritional substances are: Carbohydrate 3.9 g/100 g, vitamin C 12.7 mg/100 g, vitamin A 833 IU/100 g, and lycopene 2,573 μg /100 g with well-known anticancer properties. Vitamin C, available in tomato products, is necessary for normal metabolic performance of human body; besides, consumption of vitamin C assists us to resist against cancer, improves our cholesterol level, and compensates for collagen shortage in our body (Block 1991).

Considering crucial importance of tomato products and the role of nano-fluids in heat transfer during thermal processing of food products, which were mentioned above in details, the goal of this project was to decrease thermal processing time and, as a consequence, to maintain qualitative and nutritional properties of food products by using nano-fluids as heating fluids for thermal processing.

Materials and methods

Preparing the product

Fresh tomatoes were purchased from a local fruit market (Gorgan, Iran) and stored at 3 ± 1 °C. At the appropriate time, they were crushed using a domestic juice extractor (MJ-W176P, Panasonic, Japan). The juice was filtered on a

sterile double layer cheese cloth to remove seeds from the juice, and processed subsequently.

Nano-fluid preparation

Alumina nano-particles with 99% purity (US research nano-materials, Inc.) were purchased and dispersed with different volume concentrations of 0, 2 and 4% W/V in deionized distilled water. Then, it was stirred completely for an hour with a heater-stirrer at 1500 rpm in order to ensure nano-fluid's stability. No sedimentation was observed in the prepared nanofluid after 24 h.

Intelligent thermal processing system

This system contains a shell and tube heat exchanger, two separate reservoirs, one for liquid food, and the other one equipped with a 1 kw heater for heating the fluid (water or nano-fluid) and flow loop tubes for transferring the fluids from the reservoir to the heat exchanger. All the components were made of 316 L stainless steel, and were insulated to reduce heat loss. Required power to overcome the pressure drop was supplied by two 0.55 kw steel centrifugal pumps (3-phase induction motor, Western Electric, Australia). Control, return and drain valves were installed in proper places. Heating performance and temperatures of nano-fluid and liquid foods were controlled by thermocouples to regulate fluids flowing (Fig. 1) (Jafari et al. 2017a).

Nanofluid and food liquid exchanged their thermal energy by passing through 13 tubes with external diameter, thickness and length of 8, 2 and 800 mm (respectively) and passing from a shell with internal diameter of 100 mm in a

countercurrent pattern. Controlling flowing speed of two fluids and converting three-phase electricity of electropumps to single-phase state are carried out through a computer and two N700E vector inverters (Hyundai heavy industries Co. Ltd., Korea). Temperature changes in fluids were monitored by PT100 sensors. Heating relays, inverters (pumps) and PT100 thermal sensors were attached to the entrance of micro-controller, and were connected to the monitoring system through USB ports. Sending commands to the system and depicting temperature–time diagrams were carried out by Visual Studio 2012 software and Microsoft Excel 2010, respectively. The latter was implemented on-line and automatically every 10 s (Fig. 1).

Vitamin C determination

The samples were transferred into 250 mL volumetric flasks and diluted with 150 mL distilled water. Following by 10 drops of 1% starch solution, it was titrated against iodine solution until blue–black color was observed. The concentration of ascorbic acid in 100 mL of the sample was calculated by the following equation:

$$\text{Ascorbic acid} = 0.88 \times \text{Volume of the solution used}$$

Vitamin C retention was estimated by dividing the rate (g) of ascorbic acid per 100 mL tomato juice before treatment to that of after treatment.

For preparation of iodine solution, 5.00 g potassium iodide (KI) and 0.268 g potassium iodate (KIO₃) were dissolved into 500 mL beaker with 200 mL of distilled water. 30 mL of 3 M sulfuric acid was added into the beaker and then diluted with distilled water until 500 mL solution.

Fig. 1 Components of intelligent thermal/heating system for nanofluids with PLC section: (1) insulated stainless steel shell and tube heat exchanger, (2) food liquid reservoir, (3) nanofluid reservoir, (4) heater, (5) pipes of fluid flow, (6) stainless steel centrifugal pump, (7) stainless steel valves for fluid flow controlling, (8) PT100 sensors

Total phenolic compounds

Total phenolic content of each extract was determined by the Folin–Ciocalteu micro-method (Slinkard and Singleton 1977). Briefly, 20 μL of extract solution, after centrifuging (Combi 514 R, Hanil Science Industrial Co. LTD, South Korea), were mixed with 1.16 mL distilled water and 100 μL of Folin–Ciocalteu reagent, followed by addition of 300 μL of Na_2CO_3 solution (20%) after 5 min. Subsequently, the mixture was incubated in a water bath (ULA-40Y, Brookfield, The US) at 40 °C for 30 min and its absorbance was measured at 760 nm. Gallic acid was used as a standard for calibration curve. The phenolic content was expressed as gallic acid equivalents using the following linear equation based on the calibration curve:

$$A = 0.0017C + 0.1329 \quad R^2 = 0.9778 \quad (1)$$

where A is the absorbance and C is concentration as gallic acid equivalents (mg/g). To obtain the calibration curve, 0, 50, 100, 150, 200, 250, 300, 350 and 400 mg/L of gallic acid were prepared, and their absorbance was measured at 760 nm.

Total soluble solids

Total soluble solids (TSS) of the juice samples were measured by a digital refractometer (ABBE, CETI, Belgium) at 20 °C and expressed in terms of °Brix.

Statistical analysis

Tomato juices were treated at three different temperatures of 70, 80 or 90 °C, for 30, 60 or 90 s at three nanoparticle concentrations of 0, 2 or 4%. In this research, response surface methodology and central composite design were applied to study the influences of variable on responses (probability level of 0.05) (Table 1). Design-Expert® 6.0.2 software was used for regression analysis and to determine the best fitting models. After each thermal processing, 250 mL of tomato juices was located in the ice bath for following experiments.

Results and discussion

Vitamin C retention

Linear effects of all three factors were significant on this index ($p < 0.05$). Vitamin C content of unprocessed tomato juice was 395.524 mg/100 mL. The highest and

Table 1 Design of experiments by central composite design for tomato juices treated by nanofluids at different concentrations, temperatures and time

Number of treatment	Time (min)	Temperature (°C)	Nanoparticle concentration (%)
1	90	90	4
2	90	70	0
3	60	80	4
4	30	70	4
5	90	90	0
6	60	80	2
7	90	80	2
8	90	70	4
9	60	80	2
10	60	70	2
11	60	80	2
12	60	80	0
13	30	80	2
14	30	90	0
15	60	80	2
16	30	70	0
17	60	90	2
18	60	80	0
19	30	90	4
20	60	90	2

the lowest rates of vitamin C were 262.143 and 221.358 mg/100 mL, belonging to the treatments number 4 and 5, resulted in 66.27 and 55.96% retention respectively. The most effective factor was temperature; followed by nanoparticle concentration and time had the highest influences. On the whole, decreasing temperature and time, and increasing nanoparticle concentration could maintain higher vitamin C content. Figure 2a represents response surface diagram for simultaneous effects of temperature and concentration. As shown, increasing temperature from 30 to 60 and 90 °C reduces vitamin C content of the juice considerably, and notable drop in this index occurred at high temperatures regardless of nanoparticle concentrations. Figure 2b shows contour illustration for concurrent effects of temperature and concentration on vitamin C content. Steady color of the Figure on concentration side reveals the trifle impact of this factor on vitamin C destruction. More intense black color of the Figure, representing higher retention of vitamin C (262.143 mg/100 mL) during the process, was relevant to the highest nanoparticle concentration and lowest temperature treatment. Odriozola-Serrano et al. (2009) reported that vitamin C retention of tomato juices treated by heat pasteurization at 90 °C for 30 s was about 80.4%.

Fig. 2 **a** Three dimensional and **b** contour diagrams for the simultaneous effect of temperature–time on vitamin C changes

Extending nanoparticle concentration from 0 to 2 and 4% could culminate in the slight better retention of vitamin C owing to more rapid heat transfer from heating media to the food product and lower processing duration, leading to the higher quality of the final product. As an example, at 70 °C, thermal processing at 0, 2, and 4% concentrations could retain 62.73, 65.59, and 66.34% vitamin C content of the product, respectively. These rates changed to 56.05, 57.17 and 58.11% after increasing the temperature to 90 °C, respectively.

The main cause of vitamin C destruction is ascorbic acid oxidation to dehydroascorbic acid (DHAA), followed by 2,3-diketo-gluconic acid hydrolysis, more polymerization and formation of physiologically inactive substances; since heating accelerates oxidation, thermal treatment can cause vitamin C to destroy (Gregory 1996). Similarly, Burdulu et al. (2006) reported that increasing storage temperature from 28 to 45 °C speed up vitamin C elimination in citrus juice concentrates; also, According to Vikram et al. (2005) and Odriozola-Serrano et al.

(2008), thermal treatments resulted in vitamin C drop in orange juice and tomato juice, respectively. The latter decrease is probably on account of intensive sensitivity of vitamin C to heating procedures; so, thermal treatment of tomato pulp and juices should be carried out at the minimal duration. The latter study maintained that lower temperature during the pulp heating could maintain higher rates of vitamin C in tomato juices.

Total phenolic compounds (TPC)

Linear effects of all three factors on TPC were significant ($p < 0.05$). According to the F values, affecting rate of temperature was substantially higher than time and nanoparticle concentration. TPC amount of unprocessed tomato juices was 398.672 $\mu\text{g/g}$ fw. The highest and lowest rates of TPC were 293.542 and 181.247 $\mu\text{g/g}$ fw for treatments number 4 and 5, with preservation being 73.62 and 62.02%, respectively. Generally, lower temperature and time, and higher nanoparticle concentrations could maintain higher rates of TPC. Regarding Fig. 3a, after increasing the processing temperature from 70 to 80 and 90 °C, a significant dwindle occurred in TPC; furthermore, at low temperatures, even at high processing times, TPC was maintained at high rates and was not diminished crucially, realized by nearly flat level of the figure at low temperatures. Turning to Fig. 3b, color changes happened as temperature augmented while the color of the figure was constant across the time axis.

Likewise, Igual et al. (2010) reported that as a consequence of antioxidants sensitivity to the temperature, TPC of the grapefruit juices decreased during conventional and microwave pasteurization; the TPC preservation rate was 75 and 82% for frozen conventional and frozen microwave pasteurized juices. Aghajanzadeh et al. (2016) pasteurized orange juice and concluded that diverse levels of temperature and time had significant effects on TPC destruction. Each 5 °C increase in processing temperature diminished half-life of TPC destruction by 1.39–2.05 times, emphasizing high sensitivity of antioxidant compounds to the temperature. Vallverdú-Queralt et al. (2012) reported that TPC level of tomato juices bought from the retailers was 9.28–10.55 mg/100 g, remarkably lower than fresh tomato juices having 11.34–12.89 mg/100 g, revealing the effects of thermal processing and storage duration on tomato juices.

The prime reason of phenolic compounds destruction could be the role of peroxidase in oxidative destruction of phenolic compounds (Amiot et al. 1997) since Aguilo-Aguayo et al. (2008) announced that after thermal treatment of tomato juices at 90 °C for 60 and 30 s, 10 and 21% of peroxidase enzyme activity were maintained,

Fig. 3 **a** Three dimensional and **b** contour diagrams for the simultaneous effect of temperature–time on total phenolic compounds changes

respectively. However, Odriozola-Serrano et al. (2009) reported that the temperature during production of tomato juices did not affect the TPC of tomato samples significantly. There are two underlying reasons for this event: first, destroying cellular tissue could release some phenolic compounds; second, although destruction of cellular wall led to the release of oxidative and hydrolytic enzymes, high temperatures beyond 70 °C deactivated those enzymes and prevented a decrease in phenolic compounds (Haard and Chism 1996).

Higher nanoparticle concentrations could preserve TPC of fruit juices better than lower concentrations; for example, after 30 s thermal processing at 70 °C with 0 and 4% nanoparticles, TPC rates were maintained by 71.9 and 73.6%, respectively. This better retention could be attributed to the shorter time needed to complete the required thermal processing.

Fig. 4 **a** Three dimensional and **b** contour diagrams for the simultaneous effect of concentration–time on total soluble solids changes

Total soluble solids (TSS)

This index was 5.6 for unprocessed tomato juices and 5.4–5.6 for processed tomato juices, meaning that nano-fluid thermal processing could preserve the natural condition of tomato juices successfully. Regarding the *F* values, among the significant affecting factors, temperature was the most effective factor with the *F* value of 9.02, followed by time–concentration and time–temperature, respectively. On the whole, lower temperatures and time, and higher nanoparticle concentrations could result in lower TSS rates. Response surface figure for the interaction of nanoparticle concentration and time has been represented (Fig. 4a). As can be seen, this figure is nearly flat, expressing trivial effect of these variables on the index. The highest TSS rate belongs to the maximum nanoparticle concentration and processing time, which has been distinguished by the solid

Table 2 Model coefficients and statistics indices of content changes induced by thermal processing through nanofluids

Variable	Model	R ²	Pred R ²	SD
Vitamin C (mg/100 mL)	$9.18 - 0.002x_1 - 0.038x_2 + 0.067x_3 + 0.038x_2^2$	0.98	0.88	2.07
Total phenolic (μg/g)	$27.11 + 0.066x_1 + 8.03x_2 - 3.46x_3 - 0.003x_2x_3 - 0.061x_2^2$	0.99	0.86	1.01
TSS	$2.82 + 0.035x_2 - 4.07x_1x_2 + 12.77x_1x_3$	0.80	0.80	0.17

black color in the contour figure (Fig. 4b); on the contrary, the lowest TSS rate was observed at high concentrations and short times, differentiated by the grey color. By following TSS changes at different nanoparticle concentrations, it could be recognized that increasing concentration of the nanoparticle can result in lowering TSS rates; as the Fig. 4b shows, raising concentration from 1 to 4% at constant temperature of 30 °C lowered the TSS rate from 5.5 to 5.2 °Brix.

High TSS rates were always displayed at higher temperatures than 85 °C, and concentration or processing time could not affect this trend. These results are in agreement with the previous relevant researches. Sánchez-Moreno et al. (2006) reported that both low pasteurization (70 °C/30 s) and high pasteurization (90 °C/1 min) resulted in the same TSS rate of 7.9 °Brix. Rivas et al. (2006) that both thermal pasteurized (98 °C, 21 s) and pulsed electric fields (25 kV/cm, 68 °C) samples resulted in similar TSS rates of 10.2–10.4 °Brix in orange juices. Analogous parallel studies reported that internal indices of food products, i.e. TSS of watermelon juice and pH of tomato juice, were not affected by nanofluid thermal processing significantly, confirming the results of this research (Jafari et al. 2017b, c).

Processing time

Not only could nanofluids reach the maximum temperature sooner than our base fluid when the heater was turned on, but also tomato juices processed with nanofluids reach the maximal temperature much sooner than its counterparts when the pumps were turned on. In fact, processing time was shortened when nanofluid, rather than water, was applied. Nanofluid thermal processing could shorten duration of process from 54 min for water processing of tomato juice to 42, and 29 min for 2 and 4% nanoalumina processing of tomato juices, respectively. This means 22.23 and 46.29% reduction in processing duration of tomato juices which could be of paramount importance in the industry whether its effects on quality indices, as discussed in advance, are considered or its influence on total working hours are regarded. In fact, not only could 4% nanofluid, compared with water, dwindle the temperature required to reach pasteurization point by half, but also it reduced total processing time to the half.

The reasons of reduction in processing time for nanofluids are intrinsic properties of nanofluids and their effects on heat transfer coefficients. Heat transfer coefficient of nanofluids is dependent upon some factors including thermal conductivity, heat capacity of fluids and nanoparticles, viscosity of nanofluids, volume fraction of suspended particles, figure and dimension of particles. The increase in conductive heat transfer coefficient of nanofluids could be ascribed to a raise in their thermal conductivity, turbulence intensification, stopping boundary layer growth, and distribution of suspended particles and their chaotic movements (Kebblinski et al. 2002).

Modelling and optimization procedure

The effective variables on each response and their coefficients have been represented in the Table 2. Optimization goals were defined as the least destructions occur for vitamin C and TPC, and TSS to be kept at their normal values. Based on these terms, if temperature, concentration and time are fixed at 70 °C, 4% and 30 s, overall desirability of 95% will be obtained, and temperature continuously left the highest effect on qualitative indices, followed by concentration factor as processing time could be shortened at higher concentrations.

Conclusion

Higher nanoparticle concentrations and lower temperature or time resulted in higher vitamin C contents, with vitamin C content of about 262.143 mg/kg attaining at this state. All three variables had significant effects on the TPC response; as far as the *F* value was concerned, temperature had the most considerable influence, with nanoparticle concentration and time coming next, on this response. By following TSS changes at different nanoparticle concentrations, it could be recognized that increasing concentration of the nanoparticle can result in lowering TSS rates although the changes in TSS at different rates of variables are really restricted. Linear and interaction models could predict the behavior of each response at different rates of variables successfully. These findings, for the first time, demonstrated that nanofluid thermal technology could be deployed for improving nutritional contents of food

products significantly and successfully. It seems that other nanoparticle concentrations and types should be tested in the same way for their probable effects on thermal properties and qualitative indices of food products to realize the highest possible improvements in such properties.

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