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Electrochemical Modification of the Redox Potential of Pasteurized Milk and Its Evolution during Storage

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Several modifications that occur in milk during its processing and storage are driven by different oxidation−reduction reactions. In this study, a smooth electrolytic process was used to modify the redox state of the active species of milk thereby creating a mean to control these reactions. Five electroreduction treatments (2, 4, 6, 8, and 10 V) were applied to pasteurized skim milk. Parameters such as redox potential, dissolved oxygen, pH, conductivity, and current intensity were monitored during the course of each treatment. The proposed technology allows modulation of the redox potential of milk. Significant decreases of redox potential (441−707 mV) and of dissolved oxygen (3.3−8.3 mg L^{-1}) were obtained. However, the results suggest that only a short-term modulation is created and that natural "milk equilibrium" is reestablished after 4−5 days of storage depending on the voltage difference applied during the electrochemical treatment.

KEYWORDS: Milk; electroreduction; redox potential; dissolved oxygen

INTRODUCTION

Oxidation and reduction processes are defined in terms of electron migrations between chemical compounds. Oxidation is a loss of electrons, whereas reduction is a gain of electrons. In a simple reversible system, the oxidation−reduction potential (ORP), also referred to as redox potential, is given by the equation devised originally by Nernst (*1*):

$$E_h = E_o + \frac{RT}{nF}\ln\frac{[ox]}{[red]}$$

in which E_o is the standard ORP (or ORP at equal concentration of oxidant and reductant), R is the molar gas constant, T is the absolute temperature, F is the Faraday constant, n is the number of electrons transferred in the process per molecule, [ox] is the molar concentration of the oxidized form, and [red] is the molar concentration of the reduced form. Therefore, when $|E_h| > |E_o|$, the oxidized form predominates.

In a complex fluid such as milk, several oxidation−reduction systems are active simultaneously and their effect on the ORP depends on several factors such as the reversibility of the system, its E_o value or position on the scale of potential, the ratio of oxidant to reductant, and the concentration of active components of the system (*2*). From a physicochemical point of view, milk is a complex reactive medium in which different modifications occur during its treatment and storage. One of the main chemical transformations caused by oxidation−reduction reactions in milk is lipid oxidation (*3*). Phospholipids are the main fat molecules sensitive to oxidative stress because of their position on the milk fat globule membrane and because they are composed of polyunsaturated fatty acids (*3*). Light, metallic ions, and oxygen catalyze the oxidation of fat molecules. As a consequence, undesirable oxidized/metallic flavors can develop in dairy products. Milk is an important source of vitamins in human nutrition, mainly vitamins A, D, E, K, B_1, and B_2 (*4*). However, the integrity of their structure is important for their functional properties. Factors that promote oxidation of unsaturated lipids enhance the degradation of vitamin A and E either by direct oxidation or by the effect of free radical formation.

Microbial cell metabolism is driven by different oxidation−reduction systems (*5*). It follows that the ORP influences bacterial growth. For example, positive ORP values enhance aerobic microorganisms' growth, while anaerobes are favored by negative ORP values (in relation to the standard hydrogen electrode) (*1*). The majority of microorganisms responsible for milk spoilage are aerobic (*6*). Hence, their growth is facilitated by the positive ORP of milk.

Even though there is an impact of oxidation−reduction reactions on the quality of milk, means to control them are quasi-nonexistent. Heat treatments can decrease the ORP value of milk due to protein denaturation and loss of oxygen (*7*, *8*), but it negatively affects its flavor and nutritive value (*9*). The addition

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of reducing agents, such as cysteine, is effective to decrease ORP (*10*) but would modify the flavor of milk.

Electrolysis is a process that uses electrical energy to force reactions to occur on the surface of an electrode in contact with the liquid to be treated. The electrolysis cell is separated into two compartments by an ion exchange membrane. The fluid to be treated can circulate in contact with an anode where oxidation reactions occur (electrooxidation) or on the other side, in contact with a cathode where reduction reactions take place (electroreduction) (*11*). This type of electrochemical treatment is simple and rapid.

Electrolysis treatments already have some applications in the food industry especially for oxidizing or reducing water (*12*). Electroreduction of tap water (producing alkali-ion water) is becoming increasingly popular in Japan due to the better taste of alkali-ion water and to its ability to improve gastrointestinal health as compared to commercially bottled mineral waters (*13*). Electroreduction of water used for the production of tofu has been shown to increase its protein content and to improve its texture (*14*). Water that has been electro-oxidized and to which 0.1−0.5% NaCl has been added has strong bactericidal effects (*15*). This characteristic served as an impetus for several studies such as the sterilization of fruits and vegetables (*16*−*20*), the disinfection of poultry products (*21*), and the disinfection of various surfaces in contact with food (*22*, *23*). An electrolysis cell was also used for the electroreduction of canola oil to obtain hydrogenated oil with a lower trans-fatty acid content than that of the oil obtained by commercial processes (*24*). Hékal (1983) found that by subjecting a liquid such as a fruit juice to an electrochemical treatment on the cathode side, it was possible to slow its deterioration due to the decrease in dissolved oxygen (DO) content created by the treatment (*25*). Electrolysis treatments have also been applied to milk but, to our knowledge, only to produce milk powder with better flavor quality and without the heated milk odors usually associated with it (*26*). Therefore, the use of a smooth electrolysis treatment to modify the ORP value of milk could provide a means of controlling cell growth as well as the above-mentioned changes.

The objective of this study was to determine the anode/cathode voltage difference required to modulate the ORP of milk by a continuous electrolytic treatment and to follow its effect on other variables such as oxygen concentration, pH, and conductivity of milk. Modulated milk samples were stored at refrigeration temperature in order to assess the stability of the produced changes during storage.

MATERIALS AND METHODS

Materials. The electroreduction treatments were carried out using commercially available pasteurized skim milk (UltraMilk, Natrel, Québec City, Canada).

Methods. *Electroreduction System.* The system used was a Microflow type electrodialysis cell with a membrane electrolysis configuration (ElectroCell AB, Karlskoga, Sweden) (**Figure 1**). The cell was separated into two different compartments by a cationic membrane (CMX-SB, ASTOM Corp.) (Tokuyama Corp., Tokyo, Japan). In each compartment, one polypropylene spacer (2.02 mm thick) was placed to allow the liquid to flow through and to have contact with a 10 cm^2 surface of both the membrane and the corresponding electrode. On one side of the membrane, the milk was in contact with a food-grade stainless steel cathode, and on the other side of the membrane, the electrolyte (0.1 M H_2SO_4) was in contact with a dimensionally stable anode (DSA-O_2). The assembly was made watertight with rubber gaskets (1.23 mm thick) placed next to each of the electrodes, spacers, and membrane. Each cell compartment was connected to its own external tank (300 mL) to allow a continuous circulation during treatments. Both electrolytes were circulated by two centrifugal pumps (Iwaki Magnet Pump, Iwaki Co., Ltd., Tokyo, Japan), and their flows were maintained at 300 mL min^{-1} by two flow meters (Aalborg Instruments and Controls, Inc., Orangeburg, NY). The DC current between the two electrodes was supplied by an electrical power supply (model HPD 30-10, Xantrex, Burnaby, Canada).

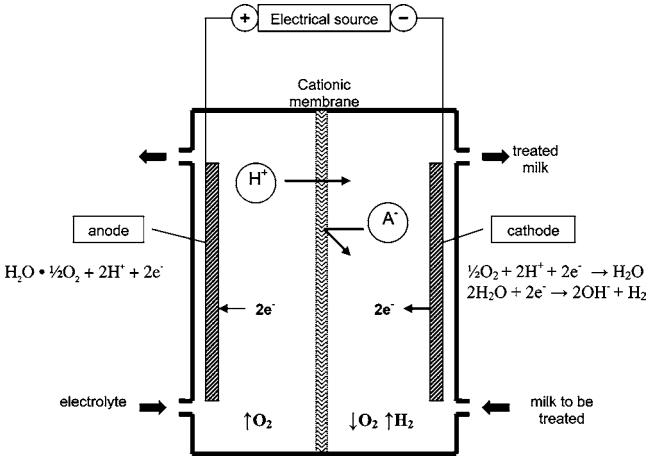

Figure 1. Simplified diagram of the membrane electrolysis cell.

Treatments. Five anode/cathode voltage differences applied between the electrodes were tested (2, 4, 6, 8, and 10 V). Each electroreduction treatment was done in triplicate using 250 mL of skim milk and an equal volume of 0.1 M sulfuric acid. During each treatment, ORP, DO, pH, and conductivity of milk were recorded as well as the current intensity at intervals of 30 s during the first 5 min of each treatment and at intervals of 1 min thereafter.

Storage. After each treatment, a sample of 125 mL of ORP modulated milk was rapidly poured in a polypropylene jar of the same volume and stored at +4 °C. The headspace in each jar was minimized by overfilling the containers. ORP, DO, and pH (except for the control milk) were recorded at 24 h intervals during 8 days in electroreduced and control milk samples.

Analyses. *ORP Measurement.* The ORP was measured using a VWR Symphony platinum electrode (VWR Scientific Products, West Chester, PA) with an internal Ag/AgCl reference electrode and filled with the recommended solution containing KCl and AgCl. This electrode was connected to a VWR Symphony portable SP20 pH/ISE meter. The electrode reading was verified with a solution of potassium ferrocyanide and potassium ferricyanide having an ORP of +234 mV, as described in the electrode user guide.

DO. The DO was measured using a VWR Symphony electrode (VWR Scientific Products) mounted with the specified membrane and filled with the supplied DO electrolyte solution. The electrode was connected to a VWR Symphony SP50D portable DO meter. The electrode was calibrated every 2 h as described in the supplier's manual.

Conductivity. The conductivity was measured with an immersible YSI probe (model 3417, $K = 1$ cm^{-1}, Yellow Springs Instrument, Yellow Springs, OH) connected to an YSI 3232 adaptor to allow readings on the YSI 3100 conductivity meter of the same manufacturer. Because the conductivity varied proportionally with temperature and the values were not automatically compensated by the conductivity meter, all readings were corrected to +25 °C using the method described in ref *27*.

pH. The pH was measured using a VWR Symphony pH electrode (VWR Scientific Products) equipped with an automatic temperature compensation device and connected to a VWR Symphony SR60IC benchtop pH meter.

Current. The current passing through the electrodes was read from a Mastercraft numerical multimeter (Mastercraft, Toronto, Canada).

Statistical Analyses. The evolution of each parameter during treatments and storage was fitted with linear or nonlinear regressions using Sigma Plot (version 8.02 for Windows, SPSS Inc., Chicago, IL). Statistical analyses of the fitted curves were performed by Sigma Plot. Parameters of electroreduction treatments were subjected to an analysis of variance using SAS Software (version 8.02.02 MOP020601, Cary,

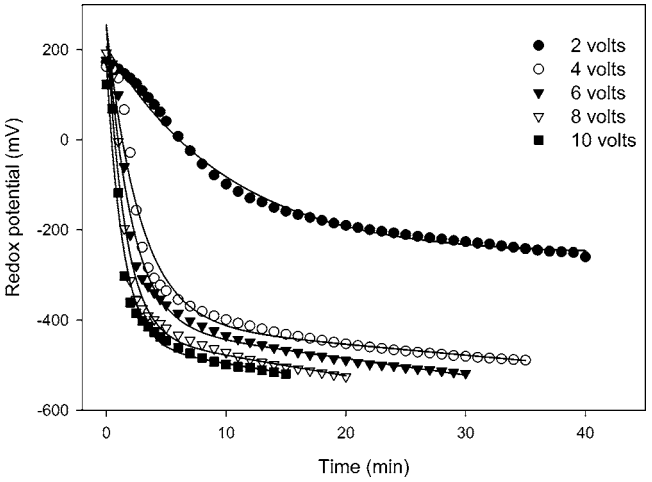

Figure 2. Changes in redox potential (ORP) during electroreduction treatments of milk at different voltages.

NC) followed by simple comparisons in a randomized block design. The evolution of each parameter during storage was subjected to an analysis of variance with repeated measures (PROC MIXED) and contrasts using the SAS Software.

RESULTS AND DISCUSSION

Electroreduction of Skim Milk. *Effect on ORP.* Raw milk has an ORP between +200 and +300 mV under aerobic conditions (*2*, *28*). However, values for pasteurized milk were not found in the literature. The mean initial ORP value for the pasteurized skim milk used during this experiment was +182 mV. All treatments performed on milk samples resulted in a significant exponential decrease of the ORP value as can be seen in **Figure 2** ($P < 0.0001$, r^2 between 0.972 and 0.993). By applying a cell voltage of 2 V, the ORP value decreased to −259 mV in 40 min. Similarly, cell voltages of 4, 6, 8, and 10 V decreased the ORP value to −489 mV in 35 min, −517 mV in 30 min, −525 mV in 20 min, and −520 mV in 15 min, respectively. **Figure 2** also shows a noticeable difference between the reduction kinetics of the ORP values at the beginning of each treatment. The slope of the initial linear part of the decrease is more abrupt as the voltage increases. Statistical analysis demonstrates that there was a significant difference between the final values reached when applying a 2 V difference as compared to the final values obtained by the other voltage differences ($P < 0.0001$).

As the anode/cathode voltage difference increases, electrons are transferred more rapidly from the cathode to the milk reducible species (*11*). As a result, reduction reactions are taking place at a faster rate, which creates a faster decrease of the ORP value. This could explain the amount of foam forming inside the system. Indeed, as the voltage applied increased, the rate of water electrolysis and, hence, the amount of hydrogen gas produced, also increased. Consequently, the treatment was ended sooner for the 6, 8, and 10 V treatments since the quantity of foam produced was becoming more important. No apparent amount of foam was produced during electroreduction at 2 and 4 V.

Effect on DO. **Figure 3** shows the evolution of DO in milk during the electroreduction treatments performed. The mean initial value of DO in the milk used in this experiment was 9.4 ppm. This value was brought down to 6.1, 2.4, 1.4, 1.1, and 1.6 ppm during the course of the electroreduction treatments at 2, 4, 6, 8, and 10 V, respectively. Therefore, each treatment caused a significant decrease in DO in the milk in a quadratic

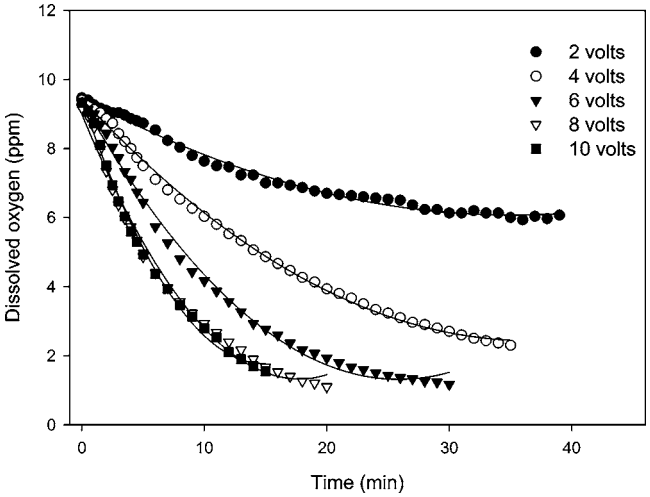

Figure 3. Changes in DO during electroreduction treatments of milk at different voltages.

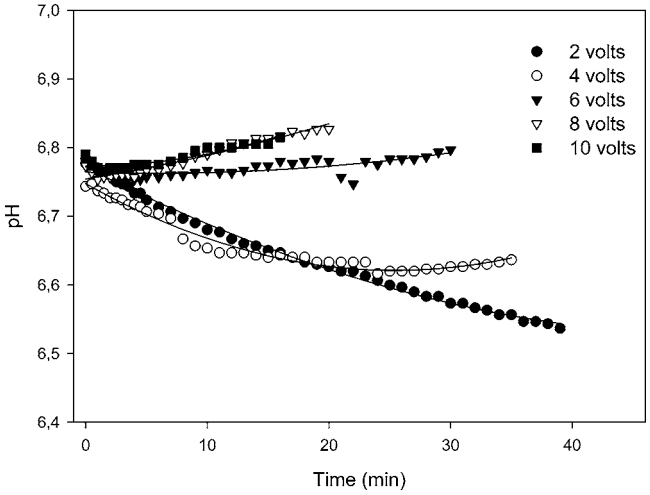

Figure 4. Changes in pH during electroreduction treatments of milk at different voltages.

pattern ($P < 0.0001$, r^2 between 0.992 and 0.998). The decrease in DO was less important when milk was treated at 2 V as compared to the other voltages ($P < 0.0001$).

The decrease in the concentration of oxygen is directly related to its reduction taking place at the cathode. Electrons are transferred from the electric circuit to the cathode and then to electroactive species in milk, one of them being oxygen. As electrons are transferred, the concentration of oxygen decreases as observed in **Figure 3**. The 2 V treatment created a slower decrease in DO concentration, and as a result, the reduction in DO during this treatment was not as extensive as compared to treatments at higher voltages. Furthermore, the initial decrease in the values of DO was faster the higher the voltage was.

Effect on pH. **Figure 4** presents the evolution of pH during the course of each treatment. Different tendencies were obtained for different voltages. The mean initial pH value of the pasteurized milk used in this study was 6.78. When milk was treated at 2 V, the pH decreased 0.2 pH units to reach 6.54. Similarly, during the 4 V treatment, a less important decrease in pH can be observed and a value of 6.63 was reached. At 6 V, the pH seemed to remain constant throughout the course of the process (slope = 0.009). The pH values during both 8 and 10 V treatments rose slightly to 6.83. The changes in pH observed for the 2 and 4 V treatments were significantly different from 6, 8, and 10 V treatments ($P < 0.0001$).

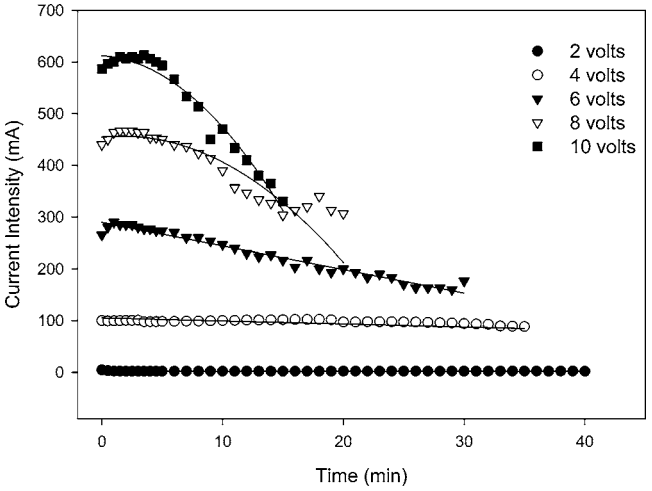

Figure 5. Changes in current intensity during electroreduction treatments of milk at different voltages.

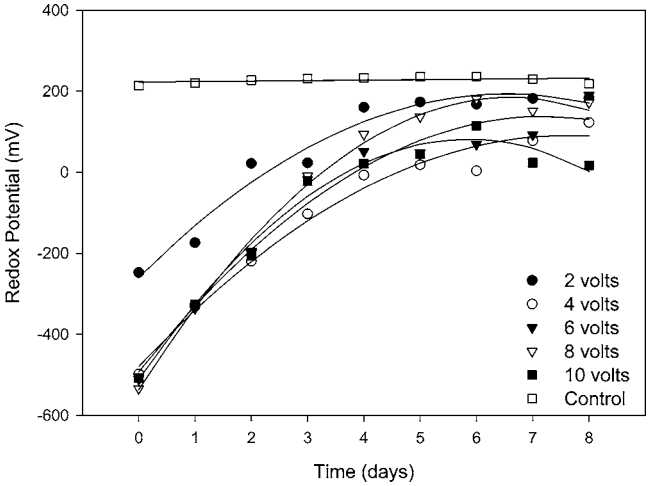

Figure 6. Changes in oxidoreduction potential (ORP) of milk treated at different voltages during its refrigerated storage.

Morris (*28*) has studied the influence of pH on ORP and concluded that ORP changes by approximately 60 mV for every pH unit change. In this experiment, the variations of pH during the electroreduction process were caused by two phenomena. The cationic membrane allowed protons present in the aqueous anolyte H_2SO_4 to migrate through the membrane toward the cathode compartment, and simultaneously, OH^- ions were produced in the cathode compartment. Consequently, H^+ cations migration was responsible for the decrease of pH during 2 and 4 V treatments since there were more H^+ ions crossing the membrane than OH^- ions being produced at the cathode (**Figure 1**). In the other treatments, the more negative electrode potential induced a faster release of OH^- ions, which was not compensated by the proton diffusion and/or migration through the cationic membrane.

Effect on Conductivity. The mean initial value of conductivity of the pasteurized milk used in this study was 4438 μS cm^{-1}. The conductivity of milk did not change over time for any of the treatments. According to the analysis of variance, no significant differences were observed ($P = 0.7953$) (data not shown). Electrical conductivity is a function of the nature and concentration of the different electrolytes present in a solution. The results suggest that the electroreduction treatment did not change the electrolyte composition in milk, except the redox state of electroactive species.

Effect on Current Intensity. The flow of electrons transferred to electroactive species in milk increased as a function of the voltage applied between the electrodes as already mentioned. This phenomenon could be observed in **Figure 5**, which represents the evolution of the electrical current passing through the system during the course of each treatment. When 2 V was applied, there was a very small current passing through the system within the range of 0.47−1.67 mA. This value remained almost constant during the treatment. A similar situation was observed during the 4 V treatment. The current values were almost constant ranging from 88 to 100 mA. On the other hand, the treatments at higher voltages were characterized by a decrease in current intensity, from 280 to 160 mA, from 450 to 307 mA, and from 596 to 330 mA for 6, 8, and 10 V treatments, respectively.

These decreases in current intensity during the course of the 6, 8, and 10 V treatments were mainly caused by the decrease in the concentration of reducible species. However, because the voltage applied between both electrodes was constant, the increase in the global resistance of the system might have also been involved ($U = RI$). This rise in resistance was caused by a slight fouling on the membrane surface as observed during the dismantling of the cell at the end of the treatments. This fouling was probably due to protein coagulation caused by the acidic pH on the surface of the membrane as was also observed by Bazinet et al. (*29*) in a similar system. This phenomenon could be minimized by increasing the flow rate of the milk within the cell.

It appears from the results that electroreduction was an effective process to rapidly modulate the ORP of skim milk. The effectiveness of electroreduction treatment was closely linked to the anode/cathode voltage difference applied. In fact, when the applied voltage was 2 V, the process did not allow complete reduction of the electroactive species in skim milk. On the contrary, when it was higher than 4 V, nondesired foam formation was observed due to H_2 gas production.

From a fundamental point of view, the electrode tension represents the potential difference between the electrode and the electrolyte in its closest vicinity (*11*). This tension is specific to the reaction produced at the electrode and could be maintained constant during electroreduction by using a Luggin−Haber capillary (*30*) connected to a reference electrode and a potentiostat receiving information from the reference electrode (potenstiostatic electrolysis). Luggin−Haber capillaries are too fragile for high flux in hydrodynamic cell such as the one used in this experiment, and furthermore, potentiostats for industrial scale applications would be expensive. Therefore, electrolyses at a constant anode/cathode voltage difference, such as those carried out in this study, would be appropriate in industrial plants (*31*).

Storage of Modulated Skim Milk. Milk samples previously treated by electroreduction were maintained at refrigerated temperature in order to verify if the negative ORP value, the low DO values, and the pH remained stable.

Oxidoreduction Potential. **Figure 6** represents the evolution of ORP in treated milks during 8 days of storage at +4 °C. This figure shows that the ORP value of the refrigerated samples increased in a quadratic fashion to reach after 4−5 days storage a positive value around +180 mV close to the ORP in nontreated control samples ($P < 0.0001$, r^2 between 0.965 and 0.994). Because their initial ORP value was higher, samples treated at 2 V reached a positive ORP value faster than the other samples. ORP in nontreated samples remained stable throughout the storage period.

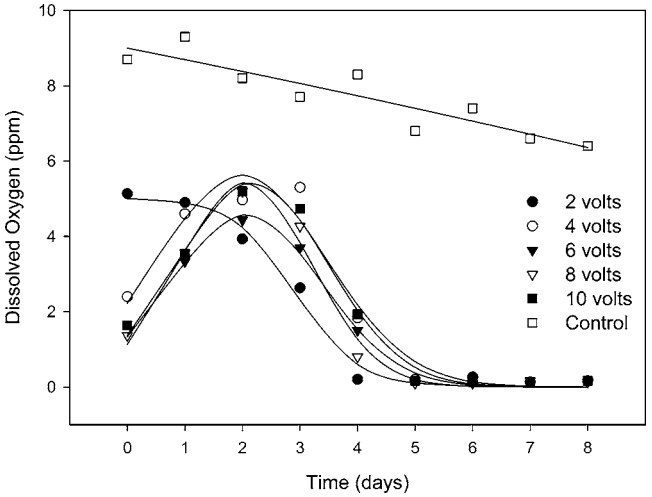

Figure 7. Changes in DO of milk treated at different voltages during its refrigerated storage.

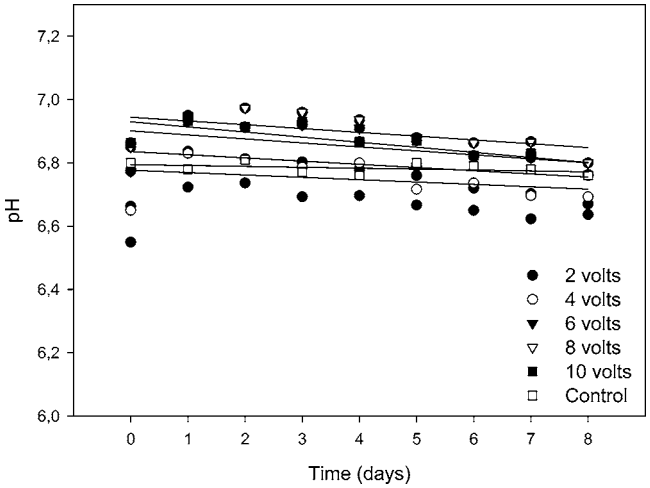

Figure 8. Changes in pH of milk treated at different voltages during its refrigerated storage.

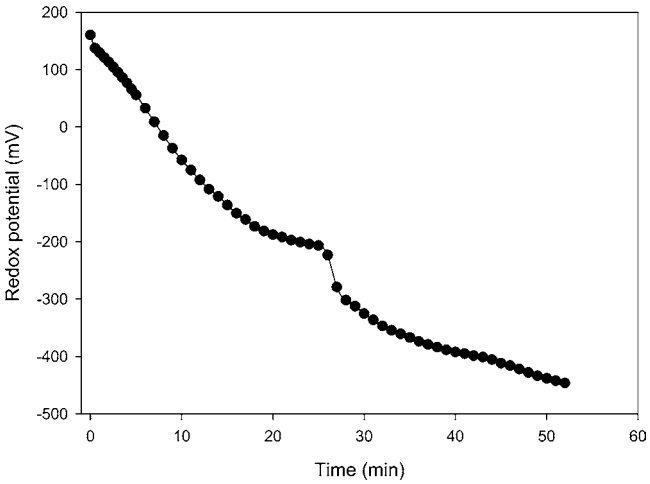

Figure 9. Changes in redox potential (ORP) during electroreduction of milk at 2 V during the first 25 min, at 4 V from 25 to 40 min, and at 6 V thereafter.

This rise in ORP during storage confirms that the changes in redox state of some milk species caused by the electroreduction treatments were neither stable nor irreversible. Even though milk containers were initially overfilled, they were opened daily for measurements to simulate the habits of the majority of consumers. This practice allowed oxygen to be incorporated into the milk, and this could be responsible for the reoxidation of the milk electroreduced species.

DO. **Figure 7** illustrates the evolution of DO in the ORP-modulated milks during storage. Except for the milk sample treated at 2 V, a rise of the oxygen concentration (2−3 ppm) was observed during the first 2 days. Then, it decreased to reach a value of 0.1 ppm for all samples after 6 days of storage. The decrease was slower for the nonelectroreduced control milk.

The initial rise in the oxygen concentration can be mainly attributed to the contact of milk samples with ambient air during measurements as well as to oxygen permeation through the polypropylene containers over time. The concentration of DO was expected to return to a value close to its value in untreated milk. However, after 2 days, a surprising decrease of DO concentration occurred until it reached a value of 0.1 ppm. There are two possible hypotheses or a combination of both to explain this phenomenon. The first one involves the growth of psychrotrophic bacteria. These microorganisms could have consumed the oxygen present in the milk during their growth, decreasing its concentration to 0.1 ppm after 6 days. Although standard plate counts of the pasteurized skim milk showed no colonies (dilution 10^{-1}) on the day of its purchase (data not shown), contamination might have been introduced from contact with the electroreduction unit and/or from ambient air and materials during measurements. The second hypothesis involves the autoxidation of residual lipids. Autoxidation reaction proceeds via typical free radical mechanisms, and its initiation may take place by hydroperoxide decomposition, metal catalysis, or exposure to light (*32*). Even though the concentration of lipids in skim milk was very low (0.3 g in 250 mL), the main fat molecules present in skim milk are phospholipids, which are very sensitive to the presence of oxygen (*4*).

pH. There was no significant change in pH during milk storage (**Figure 8**). This result suggests that the contaminating microorganisms were nonacid-producing microorganisms.

DISCUSSION

From these results, it appears that the electroreduction of milk has an important impact on the ORP and DO values. A voltage difference of 2 V seems to be insufficient for the electroreduction of all milk reducible species. Anode/cathode voltage differences equal to or higher than 4 V have to be applied to reach low ORP (≤400 mV). This situation was also observed when the anode/cathode difference applied to a milk sample was changed during its electroreduction treatment. In this case, the process was started by applying a voltage difference of 2 V. As soon as the ORP decrease started to slow (25 min), the anode/cathode difference was increased to 4 V, which allowed the decrease in ORP to continue further (**Figure 9**). Even though the voltage difference was increased to 6 V thereafter, no further decrease occurred.

Although very negative milk ORP values could be reached, the changes induced in milk were not permanent as observed during storage. The ORP values came back to their initial values after about 5 days. This buffering action of milk species called "poising effect" characterizes the capacity of a system to counteract a variation in potential (*33*). This study showed a strong poising capacity of pasteurized milk since it easily counteracts the changes in redox state created by the electroreduction treatments. This finding is opposed to the statement that

sterilized milk is a poorly poised medium (*28*). Furthermore, whey proteins may be reduced (*34*) and this may have an effect on the ORP evolution. The reduction of whey proteins would release free thiol groups from disulfide bonds reduction (*35*), which may affect the ORP. In addition, albumin is considered as the major circulating antioxidant in the blood (*36*, *37*). Recently, Fukuzawa et al. (*38*) demonstrated the antioxidant effect of bovine serum albumin, which is due to a decrease of the availability of iron for the initiation of lipid peroxidation, in addition to the trapping of active oxygen molecules and free radicals. Consequently, proteins are probably involved in the modulation and stability of ORP as well as DO concentration during storage of electroreduced milk.

The relationship between ORP values and DO level is not well-understood. On one hand, it seems that an oxygen level as low as 2 ppm in milk caused an increase in redox potential during storage, but on the other hand, a decrease in oxygen level by either bacterial cell growth or lipid autoxidation, or a combination of both, did not result in a redox potential decrease. It could be possible that a very low DO level of about 0 ppm (**Figure 7**) would be necessary to slow the ORP increase (**Figure 6**).

As suggested by Morris (*28*), it remains difficult to identify which redox couple dictates the measured ORP value in a biological system. Because milk is a complex mixture of different species, more research needs to be done to obtain a better understanding of the milk behavior during and after electroreduction treatments. This work is part of a broader research project aiming at understanding the impact of redox modulation on the milk stability during processing and storage. Further works are currently under way to identify the species responsible for the decrease in ORP, to characterize and monitor the evolution of lipids in electroreduced milk, and to determine the impact of low ORP on bacterial growth.

ACKNOWLEDGMENT

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