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Scope of Edible Packaging for Micronutrient Fortification of Dairy Products

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Abstract

Edible films and coatings have long been used for food protection and shelf life improvement. Essential components for producing edible films and coatings are biopolymers, plasticizers and certain optional ingredients. Edible films and coatings have been used as vehicles for delivery of minerals such as calcium, zinc, iron and magnesium and also for delivery of vitamins such as niacin, riboflavin, vitamin A, vitamin E and vitamin C in food products such as vegetables, rice, etc. However, there are limited studies focused on dairy products. In the present review, the scope of such edible coatings for delivery of micronutrients is presented.

Keywords: *Edible films, plasticizers, Polysaccharides, Milk protein*

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INTRODUCTION

Edible films and coatings have long been used for food protection and shelf life improvement. In twelfth century, wax coating of citrus fruits reduced the rate of dehydration and in sixteenth century, fat coating on meat reduced shrinkage. Also, in fifteenth century, soy protein films were used in Asia to improve appearance and preservation of some foods, while sucrose was used to prevent oxidation of nuts such as almonds since nineteenth century [1].

Considerable interest in edible films and coatings has been renewed due to their environment friendly nature and compatibility to use in food industry. There is no compositional difference between an edible film and a coating however, they differ in their thickness. Films are formed separately by casting process and then are applied on food surface, but coatings are formed directly on the food surface either by spraying, dipping or spreading.

A film can be separated from food surface but the coating generally is considered as an integral part of the finished product. Both can be applied on food surface or in-between the food components. They help in reducing moisture loss, fat migration, flavor loss and also as carrier vehicle for antimicrobials, antibrowning agents, antioxidants, colors, flavors, sweeteners, nutraceuticals, vitamins, minerals, etc. there by enhancing shelf life and nutritional quality. In the present article, application of edible films and coatings as carrier of micronutrients is briefly presented.

COMPONENTS OF EDIBLE FILMS AND COATINGS

Essential components for producing edible films and coatings are polymers and plasticizers. Optional ingredients include several food additives including micronutrients.

Polysaccharides

Polysaccharides used for edible films and coatings include starch, cellulose and its derivatives, pectin, gums and chitosan. Starch consists of two glucose polymers namely amylose and amylopectin. Amylopectin is a branched chain polymer while amylose a linear polymer. Amylose is responsible for the film-forming capacity of starch [2].

Sources of starch include corn (maize), potato, tapioca, wheat and rice. Genetic modification of starch crops has recently led to the development of starches with improved and targeted functionality. Cellulose is composed of repeating units of D-glucose linked through β-1,4 linkages. Due to tight packing and high crystalline structure they resist solublization in aqueous medium. The solubility can be increased by dissolving and swelling cellulose in an alkaline solution followed by reaction with chloroacetic acid, methyl chloride or propyl oxides to yield carboxymethyl cellulose (CMC), methyl cellulose (MC), hydroxyl methyl propyl cellulose (HMPC), hydoxypropyl cellulose (HPC), etc. MC is most resistant to water and lowest hydrophilic in nature. Chitosan, the second abundantly available polysaccharide in the nature after cellulose, is found in the cell walls of fungi of the class *Zygomycetes*, in the green algae *Chlorella* sp., yeast and protozoa as well as in insect cuticles. Commercially, chitosan is prepared from chitin through alkaline hydrolysis of the N-acetyl groups [3].

Chitosan films are transparent, tough, flexible and good barrier to oxygen [4]. Gums in edible film forming preparations are used for their texturizing capabilities. All gums are polysaccharides composed of sugars other than glucose. They are classified into three groups (a) exudate gums, e.g., gum arabic; mesquite gum, (b) extractive gums which come from endosperm of some legume seeds or extracted from the wood, e.g., guar gum and (c) microbial fermentation gums, e.g., xanthan gum. Pectin constitutes a heterogeneous group of acidic structural polysaccharides, found in fruits and vegetables and commercially extracted from citrus peel and apple pomace. It is composed of β-1,4-linked D-galacturonic acid residues, where in the uronic acid carboxyls are either fully (HMP, high methoxy pectin) or partially (LMP, low methoxy pectin) methyl esterified.

Proteins

Milk protein, corn protein, gelatin, soy protein, wheat gluten protein, etc. were investigated for film forming properties and applied in edible food packaging. Protein films are generally prepared by heat denaturation of protein in a suitable solvent followed by evaporation of solvent. Generally, water or ethanol is used as solvent in most of the protein film preparations. Milk contains two major proteins *viz*. casein and whey protein. Commercially, casein is available as calcium caseinate, magnesium caseinate, sodium caseinate and potassium caseinate. The films prepared from caseinates are water-soluble. However, water insoluble films can also be manufactured by treating such films with a buffer at the isoelectric point of casein. Whey proteins are the proteins that remain soluble after casein is precipitated at pH 4.6. They are globular and heat labile in nature and consist of several component proteins, including α-lactalbumin (α-La), β-lactoglobulin (β-Lg), bovine serum albumin, immunoglobulins (Igs), and proteose-peptones. Industrially produced whey protein concentrates (WPC) have a protein content of 25–80%. Whey protein isolates (WPI), which have a protein content of minimum 90%, are prepared from WPC by adding an ion-exchange step. Edible film can be prepared from either total milk protein (TMP) or from different types of milk proteins.

The formation of TMP films is complex due to the presence of lactose, which crystallizes during film formation and leads to nonhomogeneous film and film adheres to surfaces. Lactose can be extracted from nonfat dry matter by ultrafiltration or suspension in ethanol followed by a filtration. Addition of potassium sorbate also inhibits crystallization [5].

Heating TMP solution up to 135°C produces insoluble films, and films are stronger and less brittle due to the dissociation of micelles and intermolecular bonds [6]. Wheat contains two principal proteins *viz*. gliadin and glutenin which forms gluten in presence of water. Gluten film is formed by heat denaturation in ethanol followed by evaporation of ethanol. The gluten film is formed by breaking native disulfide bond and by making new disulfide bonds, hydrogen and hydrophobic bonds [7]. Edible film based on the soy protein can be prepared by either using soy protein isolate or aqueous soy extract (soy milk). Zein, the principle protein in corn, has excellent film forming properties and can be used for fabrication of biodegradable films.

Similar to wheat protein, the zein film is also formed through the development of hydrophobic, hydrogen and limited disulfide bonds between zein chains. Similar to other protein films, there is a need for addition of plasticizer to reduce brittleness of zein film.

Gelatin is prepared by the thermal denaturation of collagen, isolated from animal skin, bones and fish skins. Gelatin is readily soluble in water at temperatures above 40ºC, forming a viscous solution of random-coiled linear polypeptide chains. Gelatin films can be formed by using 20–30% gelatin, 10–30% plasticizer such as glycerin or sorbitol and 40–70% water followed by heating and drying the gel. Surfactants such as lecithin or yucca extract have been successfully used as plasticizer in gelatin-based films [8].

Lipids

Lipid based films and coatings have excellent water vapor barrier property due to relatively low polarity. Wax, acetoglyceride and shellac resins are some commonly used lipid materials in edible films and coatings. There are a variety of naturally occurring waxes such those derived from vegetables, e.g., carnauba, candelilla, and sugar cane waxes; minerals, e.g., paraffin and microcrystalline waxes; and animals including insects, e.g., beeswax, lanolin, and wool grease while some other waxes are synthetically produced such as carbowaxes and polyethylene wax [9]. Shellac resins are secreted by *Laccifer lacca*. The shellac resin coatings were used first to give glossy appearance on fruit surfaces.

Composite Materials

Several researchers attempted to develop composite films by blending different polymers to make a single film or coating to improve the food quality. The different combinations like protein-carbohydrate, protein-lipid, carbohydrate-lipid, synthetic-natural, etc. have been used. Almost all biopolymers discussed above have been used in combination for developing composite films and coatings. Some of the recent examples found in the literature are whey protein isolate and mesquite gum [10], whey protein concentrate and cinnamon essential oil [11], wheat starch and whey protein isolate [12], whey protein, almond and walnut oils [13], fish gelatin and chitosan [14], chitosan-tapioca starch [15], sodium alginate and pectin [16], sodium caseinate and maize germ oil [17], Chitosan and beeswax [18], glutelin and rice starch [19], cassava starch, carnauba wax and stearic acid [20], zein and chitosan [21], etc.

Plasticizers

Plasticizers are the low molecular weight nonvolatile compounds used to improve processing conditions of polymer by lowering glass transition temperature (T_g), improve flexibility, reduce hardness, density, viscosity, reduce tension of deformation and increase resistance to fracture. Plasticizers can be classified as internal or external, primary or secondary and hydrophilic or hydrophobic. Plasticizers used in edible film and coatings are essentially of food grade in nature. Water is the most commonly used solvent in edible films and coatings production. It decreases the T_g and increase flexibility and is considered as the most common “natural” plasticizer. Glycerol is most widely used plasticizer in protein based edible films and coatings. It is hygroscopic, odorless, colorless polyol mostly added to prevent film brittleness. Glycerol content up to 50–60% (w/w) contributes to good film forming property [22].

Sorbitol is another polyol used as a plasticizer. Sorbitol up to 38–40% (w/w) results in less stickiness with improved flexibility. Ethanolamine is a novel plasticizer that can be used in starch films to form continuous phase [23]. Surface-active agents can also be used as plasticizers to reduce surface tension of solution, improve wettability and adhesion of plastic film [24]. Surfactants such as tween 20, tween 80 and soya lecithin were also used as plasticizers in potato starch-based [25] and other films. Oleic and linoleic acids were tried as plasticizers in zein protein film. It was reported that the film exhibited high clarity, high elongation, toughness and low tensile strength. Oleic acid containing film possessed high toughness and water resistance with promising future applications in thermoformed packaging trays [23].

Methods of Formation of Edible Films and Coatings

Film formation generally involves molecular association or cross linking of polymer chain forming a semi rigid 3D network that entraps and immobilize the solvent [26]. During film manufacturing the material must dissolve in a suitable solvent such as; water, alcohol or mixture followed by adjusting the pH, and heating the solution along with plasticizers.

Further, casting and drying is done at desired temperature and relative humidity to form free standing film. The coating on food surface can be obtained by dipping, spraying, brushing and panning followed by drying [27]. Dip method of coating is commonly used for fruits and vegetables. In this method commodity is dipped into coating medium for some time then removed and allowed to dry in air. Spraying is a conventional method in which solution is sprayed at required pressure to form coating layer. This method uses less solution with better coverage. Coating in liquid form can also be applied by a brush or by use of falling-film enrobing techniques, by panning or with rollers [28].

Multilayer structural coating is one of the new methodologies that could be used in edible coatings preparation. It consists of a matrix layer consisting of a biopolymer that allows controlled release of functional ingredient, e.g., vitamin, mineral, antimicrobial agents, etc., an inner layer to control diffusion rate of active substance and a barrier layer that prevent migration of active ingredient and permeability to gases on coated food surface. Multilayer coatings could be produced by using layer-by-layer (LbL) electrodeposition [29]. Multilayer coating is formed by addition of oppositely charged polyelectrolyte to a charged surface, results in charge reversal, which allows successive deposition of oppositely charged polyelectrolytes [30]. Chitosan, poly-L-lysine, pectin and alginate with lipid droplets, micelles solid particles or surfactant were used in LbL coating [31]. Compression molding and extrusion can be used to produce films from some proteins and polysaccharides at low moisture level that shows thermoplastic behavior and this process is called as “dry process” of film formation.

Soy protein and glycerol films were produced at optimum temperature of 150°C, a pressure of 10 MPa and a dwell time of 2 min [32]. Sothornvit *et al*. [33] produced film from whey protein isolate and glycerol and found that dwell time of more than 2 min, pressure between 0.81 and 2.25 MPa and temperature above 140°C results in more degradation, increased cross linking and reduced solubility. Wang and Padua [34] used twin-screw extrusion for formation of zein (100 g) and oleic acid (70 g) film and found that higher pressure and shearing force developed inside the extruder improves tensile strength and smoothness with more uniform structure as compared to single screw extruder. Whey protein isolate sheets plasticized with glycerol were produced at 143–150°C and 200–275 rpm screw speed.

Micronutrient Fortification of Food Products using Edible Films and Coatings

Edible films and coatings have been used as vehicles for delivery of minerals such as; calcium, zinc, iron and magnesium. Baby carrots are good source of vitamin A, dietary fibers and carotene but deficient in calcium and vitamin E. Baby carrots contains 23 mg/100 g of calcium and serving (85 g) carrots only achieve 2.6% dietary reference intake (DRI) value for calcium. Mei *et al*. [35] have reported that DRI value could be increased from 2.6 to 6.6% by using 0.3% xanthan gum coating on peeled baby carrots containing 5% gluconal lactate, a mixture of calcium lactate and gluconate.

Han *et al*. [36] developed chitosan based coating for fresh and frozen strawberries (*Fragaria× ananassa*) and raspberries (*Rubus ideaus*) incorporated with 5% gluconal lactate which gave 34–59 mg of calcium per 100 g, while uncoated samples contained only 19–21 mg of calcium per 100 g. Bastos *et al*. [37] developed a calcium alginate-capsule based ascorbic acid retaining edible film by incorporating 25.6% of ascorbic acid during the preparation of film matrix.

Mridula *et al*. [38] developed calcium fortified rice using different biopolymers by soaking and spraying methods. Hydroxyl propyl methyl cellulose (HPMC), methyl cellulose (MC), combination of MC and HPMC (3:1) and zein in ethanol and water (80:20) were used. It was reported that calcium content in fortified rice premix ranged from 9.93 to 22.39 and 11.84 to 25.39 mg/g for soaking and spraying methods, respectively. Further, it was reported that among all, zein coating showed high retention of calcium followed by combination of HPMC and MC (2%). Mridula and Pooja [39] developed an iron fortified rice premix by coating rice using different iron concentration solutions. The coating material used was hydroxyl propyl methyl cellulose (HPMC) (2%), methyl cellulose (MC) (2%), combination of HPMC and MC (2%), zein, palmitic acid (9%) and stearic acid (7 and 9%). Iron content raged from 1.33 to 7.11 and 1.61 to 4.49 mg/g in steamed and without steamed rice, respectively. It was reported that iron retention in all coated samples ranged from 87.34 to 89.39% as compared to 39.12% in uncoated samples. Moreira *et al*. [40] used pectin as an active delivery matrix of nutraceutical in bioactive packaging application. It was reported that magnesium hydroxide ($Mg\ (OH)_2$) could be used as reinforcing agent in high methoxyl and low methoxyl pectin in concentrations ranging from 0.5 to 5%. De Cruz [41] developed an iron fortified cassava starch film by using starch at a different concentration and iron gluconate as iron source. Genevois *et al*. [42] studied the effect of tapioca starch coating on the pumpkin tissue that was initially fortified with 0.216 g per kg iron by dry infusion process and reported that coating resulted in improved color of pumpkin.

Edible films and coatings have also been used as vehicles for delivery of vitamins such as; niacin, riboflavin, vitamin A, vitamin E and vitamin C. Peil *et al*. [43] studied the micronutrient retention in micronutrient fortified rice by polymer coating. Rice was fortified with different layers of polymer solution containing thiamine (0.64 g) and nicin (5.33 g), riboflavin (0.4 g), iron powder (8.8 g) and vitamin A (12.86 g) by using thiamine monoitrate, riboflavin, nicotinic acid, electrolytically reduced iron and encapsulated vitamin A palmitate, respectively. Polymer solution consisted of 1.2% MC, 3.6% HPMC, 28.5% ethanol (95%) and 66.7% water. After cooking 1 g rice in 100 ml water for 25 min retention of thiamine, niacin, riboflavin, vitamin A and iron were reported to be 18, 18, 21, 70 and 100%, respectively.

Chatiyanont and Wuttijumnong [44] developed vitamin-fortified rice that contained 0.17 mg of thiamine and 27.89 mg of niacin per 100 g of rice. Rice grains (100 g) were coated with mixed vitamins solution prepared by dissolving thiamine hydrochloride (95 mg), riboflavin (52.6 mg) and niacinamide (559 mg) in 8 ml of the distilled water and then coated by using pectin solution. Shrestha *et al*. [45] reported that among different coating materials ethyl cellulose coating found best for folic acid fortification of raw rice. The folic acid content of raw milled rice used for fortification was 41 μg/100 g of rice. Mei *et al*. [35] reported that xanthan gum coating containing α-tocopherol acetate (0.2%) in 0.8% acetylated monoglyceride could be used to increase vitamin-E content of baby carrots up to 67% of DRI value.

Han *et al*. [36] reported that vitamin E content of fresh and frozen strawberries and raspberries could be increased up to 1.1 to 7.7 mg per 100 gm as compared to 0.25 to 1.15 mg per 100 g in natural strawberries and raspberries, respectively by using chitosan coating. Park and Zhao [46] used chitosan-based film as carrier material for fortification of α-tocopherol acetate. Lin and Pascall [47] developed a vitamin E fortified chitosan based edible film using 1–2% chitosan that contained about 250–500 mg of vitamin E with or without premixing with lecithin. Mei and Zhao [48] developed calcium caseinate and whey protein isolate films containing 0.1–0.2% tocopherol acetate (vitamin E) and evaluated the barrier and mechanical properties of the films.

Tapia *et al*. [49] reported that ascorbic acid content of fresh cut papaya could be almost doubled from 363.21 mg per kg to 593.23–611.70 mg per kg by using alginate (2%w/v) or gellan (0.5%w/v) based coating. Genevois *et al*. [42] studied the effect of tapioca starch coating on the pumpkin tissue that was previously fortified with 0.80 g per kg of ascorbic acid by dry infusion process.

Scope of Edible Films and Coatings for Micronutrient Fortification of Dairy Products

Although milk is considered as an ideal food for people of many age groups, it is deficient in certain micronutrients like iron and ascorbic acid. Hence, there exists a scope for finding the right strategy for fortifying milk and milk products with micronutrients. Limited studies that focused on the use of edible films and coatings for micronutrient fortification are outlined here. Chai *et al*. [50] reported that addition of 0.5% free calcium results in serious protein aggregation but pre-encapsulated calcium prepared by spray drying containing 17% Ca^{2+} ions can be added up to 1% into whey protein isolate edible films. This resulted in use of double the calcium content in films without affecting its physical properties. Mei and Zhao [48] developed a calcium caseinate and whey protein isolate film containing 5–10% gluconal calcium. Lima-Lima *et al*. [51] developed a sodium caseinate film containing 5–10% gluconal calcium. The same group of authors reported the development of an ascorbic acid fortified sodium caseinate film containing 0.01% ascorbic acid and studied the physico-chemical properties of the developed films. Traditional Indian dairy products being unique in nature, careful selection of an edible film and coating is essential for maintaining the quality attributes. Recently, successful attempts were made at ICAR-NDRI to use dairy-based edible coatings for micronutrient fortification of a paneer (Figure 1) [52].

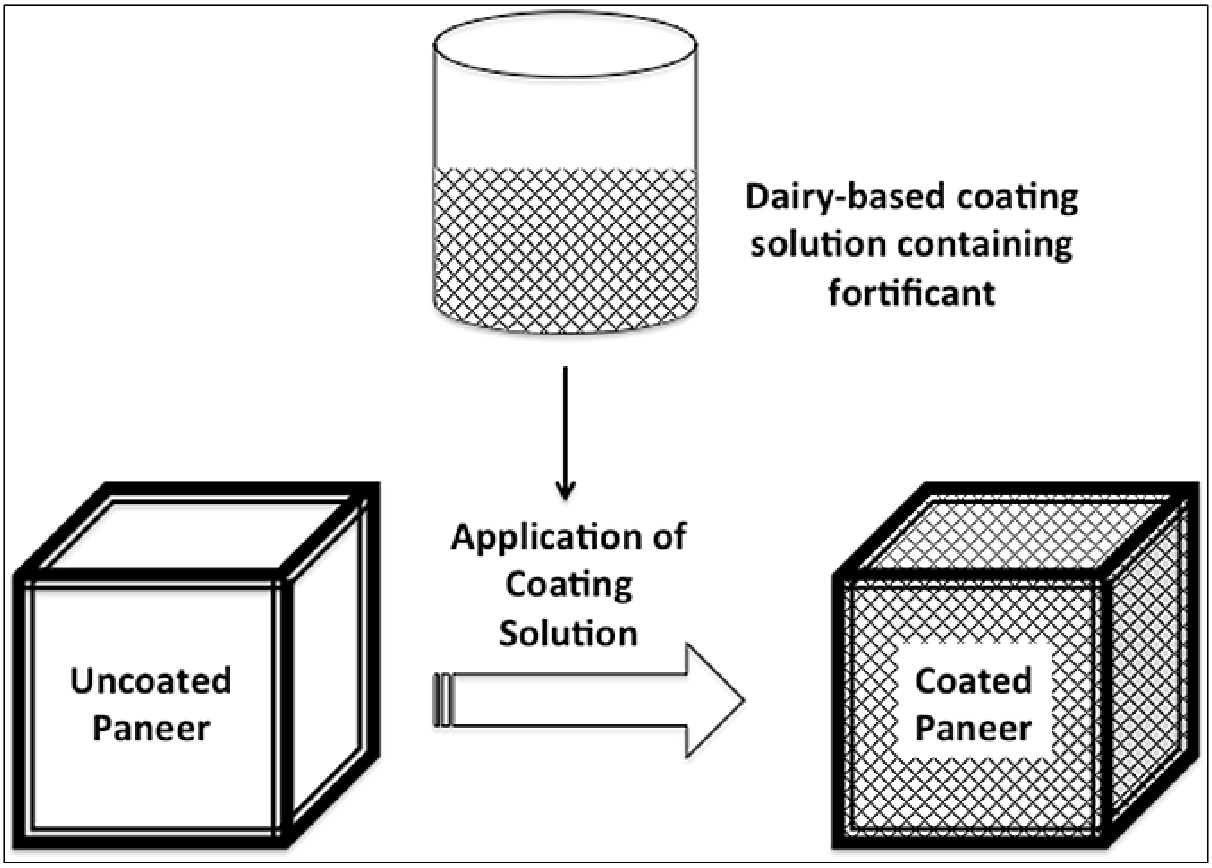

Fig. 1: *An Illustration of Use of Edible Coatings Containing Fortificant for Paneer.*

Bioaccessibility of Micronutrients

It is not important to have higher quantity of nutrients in a diet but higher quantity be available or accessible for human body. Bioavailability is defined as the proportion of the nutrient that is digested, absorbed and metabolized through normal pathways [53]. Edible films produced from different materials by using heat, solvent or other methods may lead to changes in the nutritional qualities of materials from which films were prepared. Also, the bioaccessibility depends on the nature of further treatments given to the fortified food products. For example, it was reported that pectin-coated rice premix resulted in up to 40% and 17% losses of folic acid from fortified grains upon washing and cooking, respectively [45].

Ou *et al*. [54] studied the changes in protein digestibility and available lysine in soy protein isolate films containing ferulic acid, tannin, corn starch and hydrogen peroxide at different pH by *in vitro* assays and reported that *in vitro* digestibility of soy protein decreased by addition of tannin, ferulic acid and hydrogen peroxides because of formation of cross linking with amino acids. With an increase in pH from 7 to 10 there was a decrease in the protein digestibility possibly due to isomerization of protein at high pH, which resulted in an increased resistance to digestion by pepsin. Also, the content of available lysine reduced, by reacting lysine with added ferulic acid and tannin. Matthews *et al*. [55] studied *in vivo* bioavailability of soy protein isolates and corn zein film material before and after film formation by heat and pressure and reported that the protein bioavailability in both corn zein and soy protein did not changed however, it resulted in lower protein efficiency ratio after formation into films.

Hernandez *et al*. [56] studied *in vitro* digestibility of banana, maize, potato and sagu starch edible films produced by boiling in presence or absence of plasticizer (glycerol) and reported that available starch contents lowered in glycerol-containing films that might be due to dilution effect of the plasticizer, while total resistant starch increased in the maize starch-based film but decreased markedly in those prepared from the other starches. Helal *et al*. [57] produced antioxidant-enriched sodium caseinate edible coatings to increase food protection and phenol nutritional intake by using selected phenols as model antioxidants and reported that during digestion, phenols was degraded by alkaline pH of pancreatic fluid. Simultaneously, casein proteolysis led to release of phenols and resulted in phenol index above 80% for all phenols.

CONCLUSION

Considerable interest in edible films and coatings has been renewed in research and development of dairy and food industry. There is no compositional difference between an edible film and a coating. Films are formed separately by casting process and then are applied on food surface while coatings are formed directly on the food surface. They have been successfully used as carrier vehicles for micronutrient fortification in food industry. It can be concluded from the preceding discussion that there exists a large scope for use of edible films and coatings for micronutrient fortification especially the Indian dairy products.

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