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#### 15 Abstract

Background: Mayonnaise, a high-oil containing product, is susceptible to oxidation resulting in quality deterioration and the formation of undesirable components such as free radicals and reactive aldehydes. A better understanding of the factors affecting lipid oxidation and ways of retarding oxidation in mayonnaise is essential in order to improve the shelf-life of mayonnaise.

Scope and approach: This review presents up-to-date knowledge on the factors affecting 21 lipid oxidation and strategies to retard lipid oxidation in mayonnaise, with an emphasis on 22 natural antioxidants, and application to other similar emulsions. Eliminating possible factors, 23 which will reduce the induction period and hasten rancidity, can increase the shelf life of 24 mayonnaise but one of the most effective means of retarding lipid oxidation in mayonnaise is 25 to incorporate antioxidants. Due to the negative effects and perceptions of synthetic 26 antioxidants, there has been a growing interest in improving oxidative stability of food 27 products with natural ingredients. Therefore, to provide a better base for food engineers to 28 29 design an effective natural antioxidant system for mayonnaise, in this review the emphasis is given to using natural antioxidants in mayonnaise. 30

*Key findings and conclusion:* Recent studies showed that incorporation of natural antioxidants in mayonnaise could increase its oxidative stability. However, natural antioxidants may exert a negative effect on sensory properties and further studies are needed to identify, quantify and overcome this problem. Manipulating the interfacial layer of the oil droplet also shows promise for retarding oxidation; however, there is a lack of literature addressing this area.

37 Key words: Natural antioxidant; Mayonnaise; Oil auto-oxidation; Polar paradox; Sensory
38 value

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#### 40 General introduction to lipid oxidation in food emulsion systems such as mayonnaise

41 Oxidation of unsaturated fatty acids has been the main focus of research that targets chemical instability of emulsions. Mayonnaise is a low-pH oil in water emulsion consisting of three 42 different components: 70-80% oil (the dispersed phase), vinegar (the continuous phase) and 43 44 egg yolk as an emulsifier at the interface (Li, Kim, Li, Lee, & Rhee, 2014). As in the case of all high oil-containing foods, mayonnaise is susceptible to deterioration due to auto-oxidation 45 of the unsaturated fats in the oil. Auto-oxidation proceeds through three steps: during the 46 initiation step, external energy, such as light, acts on unsaturated lipid molecules or fatty 47 acids, in the presence of catalysts such as transition metal, to generate a free radical by losing 48 a hydrogen atom. During the propagation step, the alkyl of the unsaturated lipid ( $\mathbf{R}$ ) reacts 49 very fast with molecular oxygen to form peroxide radicals. This step is always much faster 50 than the following which involves a hydrogen transfer reaction with unsaturated lipids to 51 form hydroperoxides. At this stage, lipid peroxyl radicals (ROO) and hydroperoxides 52 (ROOH) are the primary oxidation products. Lipid hydroperoxides are tasteless, but they 53 further decompose to aldehydes, ketones, alcohols, hydrocarbons, volatile organic acids and 54 55 epoxy compounds known as secondary oxidation products, which are responsible for the offflavour and off-odour of the oil. Primary oxidation products and secondary oxidation 56 products, together with free radicals, constitute the basis for measuring the oxidative 57 58 deterioration of food lipids (Shahidi & Zhong, 2005). In the termination step, the produced 59 radicals from the propagation step can be terminated by self-interactions to form non-radical species, such as oxidized polar/ non-polar dimers or trimers of lipids. 60

In emulsions formed from oil and water, lipid oxidation reactions are generally initiated at the interface between the oil and water, where pro-oxidants (transition metals) in the continuous phase are able to come into close contact with the hydroperoxides located at the

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64 droplet surface (McClements & Decker, 2000). Lipid oxidation in mayonnaise leads to the development of potentially toxic reaction products (Coupland & McClements, 1996), 65 undesirable off-flavours and consequently decreases the shelf life of mayonnaise (Alemán et 66 al., 2015). In order to tackle the problem of lipid oxidation, different strategies such as 67 eliminating factors promoting lipid oxidation and using antioxidants are necessary. One of 68 the common ways of retarding lipid oxidation is the use of antioxidants. The efficacy of an 69 antioxidant is influenced by different factors such as its interaction with other ingredients and 70 its ability to be located at the interface, where oxidation takes place (Coupland & 71 McClements, 1996). Synthetic antioxidants such as butylated hydroxy toluene (BHT), 72 butylated hydroxy anisole (BHA) and ethylene diamine tetraacetic acid (EDTA) (commercial 73 74 antioxidants) are widely used in mayonnaise to prevent rancidity. However, these products suffer from a negative impression for their toxic and carcinogenic effects in high 75 concentrations (Martínez-Tomé et al., 2001). In addition, there is a growing demand from 76 customers for products such as mayonnaise to replace chemical ingredients with natural 77 78 ingredients. Incorporation of natural antioxidants into food products has great potential for improving oxidative stability of food products and will appeal to a wider group of consumers. 79 In addition, these compounds could also have health-promoting benefits which would enable 80 81 mayonnaise producers to hit two desirable targets: health and natural (Hermund et al., 2015).

Low pH and high fat content of mayonnaise makes it resistant to microbial spoilage (Depree & Savage, 2001). Therefore, the objective of the present paper is to review current understanding of factors affecting lipid oxidation and antioxidative strategies to retard lipid oxidation in mayonnaise with a particular focus on current knowledge on the efficacy of natural antioxidants in retarding lipid oxidation in mayonnaise. The aim is to provide important information based on available literature reports concerning lipid oxidation in

mayonnaise to control lipid oxidation and facilitate the replacement of synthetic antioxidantswith natural ones.

#### 90 Factors affecting lipid oxidation in mayonnaise

Lipid oxidation in a complex food system such as mayonnaise, is not simple, so the 91 mechanism of lipid oxidation in mayonnaise is more complex than in bulk oil systems. 92 93 Although the basic oxidation reactions of lipids in mayonnaise are the same as those of lipids 94 in bulk oil systems, factors affecting lipid oxidation are significantly different in mayonnaise and bulk oil systems (Jacobsen, Let, Nielsen, & Meyer, 2008). In this section, data from 95 96 previous studies of factors influencing lipid oxidation in mayonnaise, from intrinsic to extrinsic, will be presented in order to highlight not only the most important factors affecting 97 lipid oxidation in mayonnaise, but also to provide a general view of ways to lessen these 98 factors and control lipid oxidation in mayonnaise. 99

#### 100 Metals

The presence of even small amounts of transition metals in mayonnaise can accelerate 101 oxidation by decreasing the induction period of the oil and making it more susceptible to 102 oxidation. Iron and copper are known initiators of lipid oxidation. Mayonnaise is an acidic 103 product; during manufacturing and packaging, it contacts utensils and machinery. The acid of 104 mayonnaise dissolves out the iron from a tin-lined tank and may become contaminated with 105 106 metals, which accelerate rancidity and shorten the shelf life of the finished product (Epstein, 1929b; Reynolds, 1927). Epstein (1929a) pointed out that the presence of metals in 107 mayonnaise products not only causes rancidity but also it decreases nutritional value of 108 109 ingredients present in the product. However, with proper care and precautions it is possible to lessen the risk of contamination of products, for instance, using aluminium utensils. 110

111 Temperature

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112 We know from lipid oxidation theory that high temperature increases lipid oxidation (Frankel, 1998). Findings from experiments with mayonnaise have shown the increase in 113 oxidation at higher temperatures which are in agreement with lipid oxidation theory. A study 114 investigating the effect of temperature on the oxidation of fish oil mayonnaise, showed that 115 fish oil mayonnaise is more stable at refrigerator temperatures (2 °C) than at higher 116 temperatures (30 °C) (Hsieh & Regenstein, 1991). In addition, a study on the oxidative 117 stability of cholesterol in commercial mayonnaise demonstrated that temperature and time are 118 important factors in oxidation of cholesterol in mayonnaise. They proposed that total 119 formation of cholesterol oxides during 165 days was 20.3  $\mu$ g/g at 4 °C and 30.2  $\mu$ g/g at 25 120 °C. Hence, decreasing storage temperature could be a good way of supressing the oxidation 121 of cholesterol in mayonnaise (Morales-Aizpurúa & Tenuta-Filho, 2005). Based on another 122 study, light mayonnaise (40% oil), even those without fish oil, cannot be stored at 20 °C for 4 123 months because of significant lipid oxidation (Sørensen, Nielsen, Hyldig, & Jacobsen, 2010). 124 Consistent with previous studies, higher totox values and peroxide values were recorded for 125 mayonnaises stored at 25 °C compared with samples stored at 4 °C (Li et al., 2014). 126

127 Light

Lipid oxidation caused by light exposure can be due to either photolytic auto-oxidation or 128 129 photosensitized oxidation. Photolytic auto-oxidation occurs when lipids are exposed to ultraviolet radiation and consequently, free radicals are produced. On the other hand, in the 130 131 presence of photosensitisers and visible light, unsaturated fatty acids undergo photosensitised oxidation. Natural pigments present in foods, such as riboflavin and chlorophylls, are known 132 to be efficient photosensitisers due to their conjugated double-bond system (Bradley & Min, 133 1992). Light, with a wavelength of 365 nm, promotes the oxidation of unsaturated fats due to 134 photosynthesised oxidation but light of wavelengths above 470 nm has no effect. Hence, it is 135 important to protect mayonnaise from wavelengths shorter than 470 nm (Lennersten & 136

Lingnert, 2000). The visible light in the blue range also increases oxidation in mayonnaise.
Considering lights used in supermarkets (significant source of light at 365 nm and in the 410450 nm range) avoiding intensive lighting can help preserving the fresh taste of mayonnaise
(Lagunes - Galvez, Cuvelier, Odonnaud, & Berset, 2002).

141 Packaging

In addition to processing, mayonnaise quality during storage depends on the chosen 142 143 packaging material. Some substances used in these materials may migrate to the food matrices and cause off-flavours. In some cases, gas may permeate the packaging material and 144 cause oxidation of mayonnaise. Producers choose packaging materials based on several 145 factors (e.g. the costs of material, shelf life of the product and the convenience to the user). 146 Usually manufacturers use glass or polyethylene plastics. Glass is a greater barrier against 147 oxygen than many plastics, so it can provide better protection against oxidation of the 148 mayonnaise (Buquet, 1979). Polyester materials such as PET (polyethylene terephtalate) and 149 PEN (polyethylene naphtalate) are also popular in mayonnaise packaging. They have the 150 benefits of glass like lightness, breakability and transparency. However, producers should 151 consider light transmission properties of packaging material in choosing the right packaging 152 material. The polyester materials (PET, PEN and PET/PEN) filter out the ultraviolet radiation 153 154 to different degrees (PEN and PET/PEN > PET) and thereby protect mayonnaise against lipid oxidation, but not against colour changes (Lennersten & Lingnert, 2000). The incorporation 155 of Amosorb as an oxygen scavenger in PET greatly improves the oxidative stability of 156 mayonnaise (Sensidoni, Leonardi, Possamai, Tamagnone, & Peressini, 2004). Other types of 157 packaging, such as Tetra Brik, reduce mayonnaise oxidation and extend shelf life of 158 mayonnaise by protecting it from air and light (Berasategui, 2001). In addition to packaging 159 materials, other factors like type and size of packaging may affect shelf life of mayonnaise. 160 Studies showed that type of package (jar or pouch) do not have an effect on changes during 161

storage time while package size influence aroma of mayonnaise (Martinez, Mucci, Cruz,
Hough, & Sanchez, 1998). Reducing oxygen concentration (minimizing headspace in the
container or packaging under vacuum or nitrogen) can reduce the oxidation rate in
mayonnaise (Hsieh, 1990; Hsieh & Regenstein, 1991).

166 pH

The pH of mayonnaise ranges from 3.6 to 4.0 (Krishnamurthy & Witte, 1996). The highest 167 viscoelasticity and stability of mayonnaise is achieved when the pH is close to the isoelectric 168 point of the egg yolk because of the minimum charge on the proteins (Depree & Savage, 169 2001). However, decreasing pH from neutral to around four can have a strong pro-oxidant 170 effect on mayonnaise by breaking bridges between the egg yolk proteins (low-density 171 lipoproteins, lipovitellin, and phosvitin) and iron. Subsequently, iron releases from the egg 172 yolk and becomes more accessible as oxidation initiator (Jacobsen, Adler-Nissen, & Meyer, 173 1999; Jacobsen, Timm, & Meyer, 2001). In addition to having pro-oxidant activity, the 174 distribution of the volatile compounds (secondary products of oxidation) is dependent on pH. 175 For example at pH 4, carbonyl compounds (propanal) can easily migrate from the liquid 176 phase to gas phase because of weak interactions between the proteineous emulsifier under 177 acidic conditions, so the stability of the mayonnaise flavour could be totally different from 178 oxidative stability (Takai, Endo, Okuzaki, & Fujimoto, 2003). 179

180 Chemical structure of lipids

Susceptibility of a lipid molecule to oxidation is determined by its chemical structure, in particular, the number and location of the double bonds (McClements & Decker, 2000). Saturated lipids are more stable to lipid oxidation than unsaturated lipids. One of the ways of retarding oxidation could be to use saturated lipids but in practice, it is not possible because different types of lipid in mayonnaise can provide special physical and sensory characteristics that cannot be obtained using saturated lipids. Incorporating lipids that contain

187 polyunsaturated fatty acids into mayonnaise can improve consumers' health, but these are less stable because of their double bonds. Surprisingly, from the studies on fish oil enriched 188 mayonnaise, we can see that this mayonnaise does not oxidise faster or more than 189 190 mayonnaises without fish oil. However, development of unpleasant off-odours and offflavours in fish oil enriched mayonnaise is much faster than for mayonnaise without fish oil 191 (Bragadóttir, Þorkelsdóttir, Klonowski, & Gunnlaugsdóttir, 2006; Jacobsen, Hartvigsen, et 192 al., 1999). Off-flavour in fish oil enriched mayonnaise may be caused by specific volatile 193 compounds with low sensory threshold values that stem from the oxidation of 194 eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) (Sørensen, Nielsen, Hyldig, 195 et al., 2010). 196

Recently the interest in using structured lipids for nutritional applications has increased. 197 Three different lipids based on sunflower oil such as: traditional sun flower oil (SO), specific 198 structured lipid from sunflower oil and caprylic acid (SL), and chemically randomized lipid 199 (RL) were used in mayonnaise and their oxidative stability was studied (Jacobsen, Xu, Skall 200 Nielsen, & Timm-Heinrich, 2003). Mayonnaise produced with SL had the least oxidative 201 stability among the three. The low oxidative stability of mayonnaise based on SL could be 202 due to several factors, but the most influential were the structure of the lipid, the lower 203 tocopherol content and the higher initial levels of lipid hydroperoxides and secondary volatile 204 oxidation compounds in the SL itself compared with the RL and traditional sunflower oil 205 206 employed. Although the oxidative stability of mayonnaise was totally dependent on the lipid type, the rheological properties of the mayonnaise was influenced by the structure of the lipid 207 (Jacobsen et al., 2003). Taken together, obtaining a good quality mayonnaise enriched with 208 209 SL, the process of purifying and producing SL based on sunflower oil should be improved. In addition, structured lipids based on fish oil were incorporated in mayonnaise and their 210 oxidative stability was studied (Timm-Heinrich, Xu, Nielsen, & Jacobsen, 2004). The 211

oxidative stability of mayonnaise was significantly dependant on lipid type. The mayonnaise
based on specific structured lipid showed the least oxidative stability and these results were in
good agreement with a previous study on structured lipid based on sunflower oil (Jacobsen et
al., 2003).

Using two different types of oil in mayonnaise can change the oxidative stability of 216 mayonnaise. For instance, mayonnaise made with mixed saturated medium triglyceride and 217 unsaturated linseed oil was less prone to oxidation than mayonnaise made with unsaturated 218 linseed oil (Raudsepp, Brüggemann, Lenferink, Otto, & Andersen, 2014). It can be assumed 219 that mixed unsaturated medium triglyceride decreased the oxidation of unsaturated linseed oil 220 droplets but the oxidative stabilization of mixed oil mayonnaise was not just due to diluting 221 unsaturated triglycerides with saturated triglycerides. Further studies are needed in order to 222 223 identify these effects.

224 Oil concentration

The food industry is trying to develop healthier products such as low fat formulations in 225 response to advice from consumers. Low fat mayonnaise is expected to have a lower rate of 226 oxidation than whole mayonnaise (Abu-Salem & Abou-Arab, 2008) but there are some 227 contradicting results about the effect of reduced fat content on lipid oxidation of mayonnaise. 228 In a study on mayonnaise with 63% oil and mayonnaise based salad with lower amount of 229 oil (24%), it was found that the mayonnaise with more oil was more oxidatively stable than 230 mayonnaise with less oil (Sørensen, Nielsen, & Jacobsen, 2010). Moreover, fish oil enriched 231 mayonnaise used in this study had a greater oxidative stability compared to other fish oil 232 enriched mayonnaises in other studies. Sørensen et al. (2010) referred this higher oxidative 233 stability to a combination of lower total oil content (63%), lower fish oil content (6.3%) and 234 low storage temperature (2 °C). In another study, light mayonnaise (40%) oxidized at the 235 same rate as a full fat mayonnaise (80%) but faster than (63%) mayonnaise (Sørensen, 236

Nielsen, Hyldig, et al., 2010). Therefore, it is likely that factors other than the oil content
affect the oxidation rate in light mayonnaises, and further investigations should be done to
resolve this issue.

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241 Type of emulsifier

In mayonnaise, oil droplets are surrounded by a membrane of emulsifier molecules that provides physical stability of the emulsion and oxidative stability by acting as a barrier against pro-oxidants such as metals and free radicals. Hence, the effect of emulsifier on the oxidative stability of mayonnaise is important.

Mayonnaise is traditionally made from egg yolk, which plays the role of emulsifying agent. 246 Besides egg yolk, several emulsifiers have been used in mayonnaise and their oxidative 247 stability has been studied. Lecithin from different oil sources (sunflower, corn germ and 248 soybean) as well as their modified forms (lecithin-soybean protein isolate and alcohol soluble 249 fraction) were used in low calorie mayonnaise (Magda, Mostafa, El-Deep, & Kishk, 2003). 250 Mayonnaise made with whole egg was the most sensitive to oxidation while soluble lecithin 251 fractions were the best emulsifiers during storage period. It can be concluded that the use of 252 modified lecithins can improve the oxidative stability of low calorie mayonnaise (Magda et 253 al., 2003). On the other hand, a study on oxidative stability of mayonnaise-like emulsions 254 containing salmon oil emulsified with soy milk and whole egg showed that the emulsion 255 prepared with whole egg was more stable to oxidation than the mayonnaise made with 256 soymilk (Takai et al., 2003). Soymilk and whole egg have a variety of proteins so the protein 257 structures in the emulsion might influence the form and size of lipid droplets. Additionally, 258 electric charge differs between whole egg and soymilk. Moreover, phospholipids might act as 259 antioxidants and increase oxidative stability of emulsions containing whole egg. Some studies 260 on fish oil enriched mayonnaise suggested the pro-oxidant effect of iron from egg yolk used 261

262 as emulsifier at low pH (Jacobsen, Timm, et al., 2001). Based on these findings Sørensen et al. (2010) decided to investigate the influence of substituting egg yolk as an emulsifier with 263 an emulsifier with a lower iron content such as milk protein on oxidative stability of fish oil 264 enriched mayonnaise. Surprisingly replacing egg yolk with a lower iron-containing emulsifier 265 did not enhance the oxidative stability of fish oil enriched mayonnaises. Even though the 266 mayonnaise with milk protein as emulsifier was more viscous and probably had a multilayer 267 or a cationic surface around the oil, the peroxide value of it was 100-fold higher than the 268 peroxide value of mayonnaise with egg yolk. From the results of this study, Sørensen et al. 269 (2010) suggested that the initial quality of emulsifier is even more important than its iron 270 content. 271

272 The influence of ingredients

Mayonnaise is composed of an array of components in the aqueous phase and at the oil-water 273 interface, such as NaCl, sugar, lemon juice and vinegar, that can affect lipid oxidation 274 processes. Some of these ingredients contain low concentrations of Fe and Cu (Jacobsen, 275 Hartvigsen, et al., 2000) but egg yolk, a traditional emulsifier in mayonnaise, is a major 276 source of iron and contains >720  $\mu$ M iron, but only ~17  $\mu$ M copper. The iron in the egg yolk 277 is bound to the protein phosvitin (Causeret, Matringe, & Lorient, 1991). Mayonnaise has a 278 low pH and at this pH, the iron bridges between phosvitin, lipovitelin and low-density 279 lipoprotein (LDL) break and cause the release of iron. Ascorbic acid is able to reduce  $Fe^{3+}$  to 280  $Fe^{2+}$ , which is more active as an oxidation catalyst than  $Fe^{3+}$  (Jacobsen, Adler-Nissen, et al., 281 1999; Jacobsen, Timm, et al., 2001). This phenomenon can happen even when  $Fe^{3+}$  is bound 282 to phosvitin as indicated in Figure 1 (equilibrium 3). 283

Accessible iron either at the oil/water interface (as with low pH) or in the aqueous phase (as with the presence of ascorbic acid) catalyses the breakdown of hydroperoxides (LOOH) to secondary oxidation products (LOOH +  $Fe^{2+} \rightarrow LO^{-} + OH^{-} + Fe^{3+}$ ) that make rancid and fishy

287 off-flavours. Lemon juice in the aqueous phase of mayonnaise promotes radical formation. This could be because of the presence of ascorbic acid in lemon juice. As mentioned above 288 ascorbic acid in mayonnaise acts as a pro-oxidant because it can form a complex between 289 290 iron and ascorbate, which breaks lipid hydroperoxides at the oil-water interface (Hsieh, 1990; Jacobsen, Adler-Nissen, et al., 1999). Vinegar is one of the mayonnaise ingredients that act as 291 pro-oxidant because of its ability to reduce the pH and subsequently increase the release of 292 iron from egg yolk (Thomsen, Jacobsen, & Skibsted, 2000). Salt has an important role to 293 play. It contributes to the flavour of mayonnaise and can promote the stability of emulsion. In 294 addition, salt can influence the rate of auto-oxidation. The effect of three types of salt: NaCl, 295 mineral salt (65% NaCl, 25% KCl and 10% MgSO<sub>4</sub>.6H<sub>2</sub>O) and Morton Lite salt (50% NaCl, 296 50% KCl) showed that NaCl and mineral salt increased the oxidation of mayonnaise in the 297 absence of antioxidant while Morton Lite salt did not (Lahtinen & Ndabikunze, 1990). 298 However, antioxidant neutralized their effect but 85% NaCl still increased the oxidation 299 level. On the other hand Thomsen et al. (2000) indicated that neither NaCl nor sugar induce 300 the formation of radicals in freshly produced mayonnaise. Ostrich eggs are a good source of 301 protein, total lipids, carbohydrates, calcium, phosphorus, potassium, sodium and zinc. 302 Interestingly, a study showed that mayonnaise made from ostrich eggs was more oxidatively 303 stable than that made from chicken eggs (Abu-Salem & Abou-Arab, 2008). Also, in that 304 study they found out that by pasteurization of mayonnaise made either with ostrich eggs or 305 with chicken eggs, we can increase oxidative stability of mayonnaise. Pasteurization is likely 306 to stabilise the egg by inactivating pro-oxidation and oxidation factors and oxidation 307 mediating enzymes. 308

309 Physical structure of mayonnaise

Contact between iron and oil in water emulsion droplet surface promotes metal catalysedoxidation. Therefore, it could be assumed that the size of the total oil droplet surface area

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312 influences the rate of oxidation. For this reason, a study investigated the influence of oil droplet size on oxidative stability of mayonnaise (Jacobsen, Hartvigsen, et al., 2000). They 313 found out that kinetics of the initiation and propagation of oxidation process in mayonnaise is 314 315 greatly affected by oil droplet size. Mayonnaise with larger droplets developed fishy and rancid off-flavour slower and later than mayonnaise with smaller droplets in the initial stage 316 of the storage period. These findings show that the initial oxidation of mayonnaise is 317 dependent on interfacial area and support the hypothesis that lipid oxidation is initiated at the 318 oil/water interface. However, the propagation of oxidation is less dependent on interfacial 319 area. Moreover, the droplet size also influences the rheological properties of mayonnaise. It 320 has been proposed that one way of retarding oxidation in oil in water emulsion could be 321 reducing the diffusion rate of oxidation by increasing the viscosity of the aqueous phase 322 (Sims, Fioriti, & Trumbetas, 1979). Although higher viscosity generally decreases the rate of 323 oxidation by reducing the diffusion rate of pro-oxidants, in the case of mayonnaise, higher 324 viscosity increased the oxidation rate due to a smaller average particle size. Therefore, it 325 seems that particle size plays a greater role than viscosity in oxidation processes in 326 mayonnaise. Mayonnaise with small droplet size is more physically stable. In order to meet 327 the needs of food manufacturers for having both physically and oxidative stable mayonnaise, 328 optimal combination of processing conditions and emulsifiers should be adopted (Jacobsen, 329 Hartvigsen, et al., 2000). 330

#### 331

### Retarding lipid oxidation in mayonnaise

Eliminating possible factors, which will reduce the induction period and hasten rancidity, can increase the shelf life of mayonnaise. Some of the means of retarding lipid oxidation is reducing oxygen concentration in food (by packing under vacuum or nitrogen and using packaging materials with good oxygen barrier properties) (Coupland & McClements, 1996) and lowering the storage temperature. For example, exclusion of oxygen by using nitrogen

can retard lipid oxidation more than addition of Tertbutyl hydroquinone (TBHQ) (0.02%) to
mayonnaise produced with fish oil (70%) (Hsieh & Regenstein, 1991). However, exclusion
of oxygen in a used food product is difficult so one of the most effective means of retarding
lipid oxidation in mayonnaise is to incorporate antioxidants. Several studies have been
carried out on the effect of synthetic and natural antioxidants in mayonnaise. Table 1 presents
collated summary on the use of natural and synthetic antioxidants in mayonnaise.

### 343 Synthetic antioxidants used in mayonnaise

According to the codex standard for mayonnaise, utilizing of some chemical antioxidants at defined concentration is permitted. Synthetic antioxidants such as BHT, BHA, TBHQ and EDTA have been used in food industry to prevent the oxidation of fat in food. Although these products are more economical than natural antioxidants, they get a negative impression for being a synthetic product. Among the synthetic antioxidants, BHA and BHT are widely used in food industry (Sanhueza, Nieto, & Valenzuela, 2000).

350 TBHQ

Tert-butylhydroquinon (TBHQ) is a popular synthetic antioxidant. It is a phenolic compound and a polar antioxidant (Belitz, Grosch, & Schieberle, 2009). The maximum level of allowed TBHQ in the finished product is 120 mg/kg according to Codex Alimentarius. It was a successful antioxidant for deodorized and stabilized fish oil mayonnaise stored at 2 °C for 14 weeks (Hsieh, 1990; Hsieh & Regenstein, 1991). In addition, TBHQ was effective at early storage times in lengthening the period before oxidation was initiated (Hsieh & Regenstein, 1992).

358 EDTA

Ethylene diaminotetraacetic acid (EDTA) is a synthetic antioxidant. It was the first widely used chelating agent. EDTA, together with ascorbic acid, propyl gallate, and citric acid, efficiently inhibited off-odor development in mayonnaise based on 100% fish oil (Jafar,

362 Hultin, Bimbo, Crowther, & Barlow, 1994). EDTA can chelate free iron as well as phosvitinbound iron in egg yolk at the oil-water interface (Thomsen, Jacobsen, et al., 2000). Therefore, 363 iron ions are unable to catalyse lipid hydroperoxide decomposition to products that may 364 develop oxidation or probably decompose further to off-flavour volatiles. EDTA has been 365 found to prevent peroxide formation (Thomsen, Jacobsen, et al., 2000) also it has the ability 366 to inhibit formation of heptadienal better than hexanal (Jacobsen, Hartvigsen, Thomsen, et 367 al., 2001) and it can efficiently prevent off-flavours in mayonnaise. From all studies on the 368 effect of EDTA on oxidation in mayonnaise we can conclude that it is an efficient antioxidant 369 in mayonnaise (Jacobsen, Hartvigsen, Thomsen, et al., 2001; Jacobsen et al., 2003; Nielsen, 370 Petersen, Meyer, Timm-Heinrich, & Jacobsen, 2004; Thomsen, Jacobsen, et al., 2000; 371 Thomsen, Kristensen, & Skibsted, 2000). 372

As mentioned above EDTA prevented formation of heptadienal better than hexanal, this 373 difference is due to the different origin of these volatiles (Jacobsen et al., 2008). Heptadienal 374 is formed from n-3 fatty acids (n-3 peroxides) and are more polar than hexanal that is formed 375 from n-6 fatty acids (n-6 peroxides). Therefore, n-3 peroxides may be present more in the 376 aqueous phase or the oil/water interface than n-6 peroxides where EDTA is present. 377 Therefore, co-localisation of EDTA and n-3 peroxides could be a reason of EDTA's 378 efficiency on prevention of formation of n-3 peroxides. Other reason could be, more 379 sensitivity of n-3 PUFA peroxides (because of having more high number of double bonds) to 380 metal catalysed degradation (Jacobsen et al., 2008). 381

When EDTA was added to fish oil mayonnaise the droplet size, decreased but EDTA strongly inhibited oxidation. This result indicated that only when oxidation is catalysed by iron stemming from egg yolk compounds (phosvitin) at the oil water interface the droplet size is important otherwise a decrease in the oil droplet size did not promote oxidation rate (Jacobsen, Hartvigsen, Thomsen, et al., 2001).

387 Propyl gallate

Propyl gallate is an ester of gallic acid. Due to its alkyl chain, it is less polar than gallic acid. 388 It is widely used as antioxidant in food industry. Previous studies suggested that propyl 389 390 gallate was one of the two best phenolic antioxidants in mackerel muscle system (Kelleher, Silva, Hultin, & Wilhelm, 1992). Jafar et al. (1994) decided to use propyl gallate in fish oil 391 mayonnaise as the free radical scavenger (propagation inhibitor) in the oil phase. They used 392 the mixture of citric acid or sodium citrate and propyl gallate in the oil phase, and EDTA and 393 ascorbic acid in the aqueous phase and suggested that this mixture could increase the shelf 394 life of fish oil mayonnaise without antioxidant, from 1 day to an average of 49 days at room 395 temperature as judged by sensory evaluation. In another study, Jacobsen, Hartvigsen, Lund, 396 Meyer, Adler-Nissen, Holstborg, and Hølmer (1999) employed two different types of 397 398 commercial propyl gallate mixtures (oil soluble (Grindox 370) and water dispersible (Grindox 413)) in fish oil mayonnaise. Propyl gallate not only increased the fishy and rancid 399 off-flavour but also affected rheological characteristics of mayonnaise. Mayonnaise 400 containing propyl gallate was less viscous, had bigger droplets and a lower gel strength. In 401 addition, propyl gallate slightly increased the peroxide value of mayonnaise so it could be 402 concluded that propyl gallate had a pro-oxidant effect in mayonnaise. Pro-oxidative property 403 of propyl gallate (polar antioxidant) could be due to its presence at the interface where it can 404 interact with metal ions ( $Fe^{3+}$ ) in the egg yolk and its ability to alter the structural properties 405 406 of the system (Jacobsen, Schwarz, et al., 1999). In order to confirm that if propyl gallate was a pro-oxidant in mayonnaise and omit the effect of type of oil Jacobsen et al. (2003) decided 407 to use propyl gallate in mayonnaise with specific structured lipid (SL) from sunflower oil and 408 caprylic acid . The results from this study were in good accordance with previous studies on 409 fish oil mayonnaise. Mayonnaises had higher amounts of secondary oxidation products and 410 volatile compounds. Therefore, it can be concluded that propyl gallate was a pro-oxidant. 411

412

#### 413 Natural antioxidants used in mayonnaise

414 Because of synthetic antioxidants' chemical stability, low cost and availability they are 415 universally applicable. However, some studies questioned their safety due to their potential 416 risk. Nowadays consumers are more concerned about the safety of preservatives and 417 additives. Therefore, there is a growing trend in consumer preferences toward clean labelling. 418 All of these motivated food industries to explore natural sources of antioxidants.

419 Gallic acid

Gallic acid is a plant phenolic acid. Antioxidant activity of phenolic acids is generally by 420 trapping free radicals. Mayonnaise is a hetrophasic food system, in such a system 421 422 antioxidants may partition into different phases. In mayonnaise, 80% of gallic acid –a polar antioxidant- is partitioned in the aqueous phase but 20% of it is distributed in the interface 423 (Jacobsen, Schwarz, Stöckmann, Meyer, & Adler-Nissen, 1999). In a study on fish oil 424 mayonnaise, addition of gallic acid, caused the increase in the intensity of fishy, rancid and 425 metallic off-flavour due to a faster decomposition of hydroperoxides (Jacobsen, Hartvigsen, 426 Thomsen, et al., 2001). It is hypothesised that 20% of gallic acid was located at the oil-water 427 interface (Jacobsen, Schwarz, et al., 1999) and gallic acid can reduce metal ions and therefore 428 it may interact with metal ions from phosvitin at the oil-water interface. Nevertheless, gallic 429 acid slightly decreased droplet size that could increase the rate of lipid peroxide 430 decompositions in mayonnaise. In conclusion, gallic acid showed a pro-oxidant activity in 431 mayonnaise because of its ability to reduce metal ions to their more active form e.g.  $Fe^{3+}$  to 432 Fe<sup>2+</sup> (Jacobsen, Hartvigsen, Thomsen, et al., 2001; Jacobsen, Horn, Sørensen, Farvin, & 433 Nielsen, 2014). 434

435 Ascorbic acid

436 Ascorbic acid is a natural water-soluble antioxidant. It mainly exerts its antioxidative effect by terminating chain radical reactions via electron transfer (Gülcin, 2012) but may also act as 437 an O<sub>2</sub> scavenger (Pongracz & Kläui, 1981). Ascorbic acid is considered as a pro-oxidant 438 because it can catalyse the breakdown of already existing lipid hydroperoxides (LOOH) via 439 reduction of  $Fe^{3+}$  to  $Fe^{2+}$  (Frankel, 2005). As already mentioned in previous section on propyl 440 gallate, Jafar et al. (1994) employed a mixture of antioxidants (citric acid or sodium citrate 441 and propyl gallate in the oil phase and EDTA and ascorbic acid in the aqueous phase) which 442 increased the shelf life of mayonnaise without antioxidant, from 1 day to an average of 49 443 days at room temperature. In this mixture of antioxidants, ascorbic acid functioned as the free 444 radical acceptor and perhaps as reducing agent. However, addition of ascorbic acid to fish oil 445 mayonnaise developed the formation of strong metallic, fishy and rancid off-flavours in fish 446 oil mayonnaise and acted as a pro-oxidant. The intensity of off-flavours was dependent on 447 ascorbic acid concentration (Hsieh & Regenstein, 1991; Jacobsen, Adler-Nissen, et al., 1999; 448 Jacobsen, Timm, et al., 2001). When ascorbic acid is added to the aqueous phase of 449 mayonnaise, it can reduce  $Fe^{3+}$  to  $Fe^{2+}$ , which is more active as an oxidation catalyst. This 450 may happen even at high pH (Fe<sup>3+</sup> is bound to phosvitin) or at low pH (iron ions are more 451 accessible,  $Fe^{2+}$ , are liberated from egg volk). Hence, ascorbic acid accelerates and promotes 452 oxidation in fish oil mayonnaise (Jacobsen, Timm, et al., 2001). 453

454 Tocopherol

Tocopherols are the best known and most widely used antioxidants (Pokorny, 1987). They have four isomers ( $\alpha$ ,  $\beta$ ,  $\gamma$  and  $\delta$ ). Tocopherols act as primary antioxidants by donating the hydrogen of the hydroxyl group to the lipid peroxyl radical. Also they are efficient singlet O<sub>2</sub> scavengers (Burton & Ingold, 1981). They can react with hydroperoxyl radicals and alkoxyl free radicals formed by the metal-catalysed decomposition of hydroperoxides (Burton & Ingold, 1981; Huang, Frankel, & German, 1994). In mayonnaise, antioxidative effect of

461 tocopherols was dependent on whether it was water (Grindox 1032) or oil (Toco 70) soluble complex and also on its concentration (Jacobsen, Adler-Nissen, et al., 2000). Water soluble 462 tocopherol resulted in the highest antioxidative effect against both peroxides and volatiles 463 formation, reduced the formation of rancid and fishy off-flavours and increased oil droplet 464 sizes and gel strength compared to oil soluble tocopherol in fish oil mayonnaise. However, oil 465 soluble tocopherol (Toco 70) at high concentrations showed pro-oxidative effects on fishy 466 odour and flavour. The antioxidative properties of water-soluble tocopherol may be due to its 467 ability to decrease the total interfacial area and higher gel strength (Jacobsen, Adler-Nissen, 468 et al., 2000). To get a better insight into the effect of tocopherol on oxidation in mayonnaise 469 Jacobsen, Hartvigsen, Lund, et al. (2001) studied the effect of tocopherol concentration on 470 oxidation. Surprisingly their results were in contrast to their previous study. They found out 471 that Grindox 1032 showed pro-oxidative effect in high concentrations (more than 700 mg/kg, 472 corresponding to 140 mg/kg tocopherol) and in low concentration, showed a week 473 antioxidative effect. They indicated that the different effects of low concentration of Grindox 474 1032 and Toco 70 on the formation of volatiles could not be due to differences in droplet 475 size. They suggested that the droplet size could be influenced by parameters other than 476 antioxidant addition e.g., small differences in processing conditions. A significant pro-477 oxidant effect of Grindox 1032 was seen when more than 700 mg/kg Grindox (corresponding 478 to 140 mg/kg tocopherol) was added to mayonnaise. Therefore, it could be concluded that 479 addition of more than 740 mg/kg total tocopherol had pro-oxidative effect. To sum up 480 addition of tocopherol to fish oil mayonnaise is not a good choice of antioxidant. That may be 481 because of it lacks the ability to prevent the metal catalysed decomposition of peroxides. 482

483 Rosemary

Recently a lot of attention has been employed on using natural antioxidants of plant extracts.
Phenolic compounds that act as natural antioxidants are widely distributed in plant tissues

486 like rosemary. The most effective antioxidant constituents of rosemary are phenolic diterpenes carnosic acid and carnosol. Carnosic acid has several times more antioxidative 487 activity than phenolic synthetic antioxidants like BHT and BHA (Richheimer, Bernart, King, 488 489 Kent, & Beiley, 1996). Carnosic acid and carnosol can chelate iron and scavenge peroxyl radical in lipid-based systems (Aruoma, Halliwell, Aeschbach, & Löligers, 1992). However, 490 few studies have been carried out on utilising rosemary as an antioxidant in mayonnaise. 491 Addition of rosemary extract to sunflower oil mayonnaise decreased the level of volatile 492 compounds formed from photooxidation in the headspace (Lagunes-Galvez et al., 2002). 493 494 Rosemary extracts could have a chelating effect in sunflower oil mayonnaise. Also, the antioxidative effect of dried rosemary at a concentration of 1% was studied in fish oil 495 enriched tuna salad (Sørensen, Nielsen, & Jacobsen, 2010). Although rosemary inhibited 496 formation of peroxide and showed antioxidative effect, the taste introduced to the product 497 might be undesirable in tuna salad (Sørensen, Nielsen, & Jacobsen, 2010). 498

499 Lactoferrin

Lactoferrin is a milk glycoprotein occurring naturally in numerous bodily secretions, 500 including milk, tears, mucus, blood, and saliva. Lactoferrin is also the main iron-bearing 501 protein in cow's milk, and it is able to bind two  $Fe^{3+}$  in cooperation with two  $HCO^{3-}$  ions 502 when fully saturated (Nielsen et al., 2004). Synthetic chelating agents like EDTA could be 503 replaced by lactoferrin, which is a natural compound with a metal chelating property. So 504 studies have been done on the possibility of using lactoferrin in mayonnaise as a chelating 505 agent (Jacobsen et al., 2003; Nielsen et al., 2004). In a study on structured lipid (SL) from 506 sunflower oil mayonnaise, 10 µm of lactoferrin was used as an antioxidant. This study 507 showed that lactoferrin at this concentration did not show any antioxidative effect (Jacobsen 508 et al., 2003). Jacobson et al. (2003) suggested that the protein may become denatured at the 509 low pH (pH of mayonnaise was 4.0) and lose its ability to chelate iron. However, only one 510

511 concentration of lactoferrin was tested in SL mayonnaise and other concentrations should be tested. Therefore, in another study the antioxidative effect of lactoferrin (8-32 µM) in fish oil 512 mayonnaise (16% fish oil, 64% rapeseed oil) was studied (Nielsen et al., 2004). Lactoferrin 513 exhibited a concentration dependent protective effect. It worked optimally as an antioxidant 514 at a concentration of 8 µm while it appeared to have pro-oxidative effect at high 515 concentrations. The pro-oxidant effect of lactoferrin at high concentrations has been 516 speculated to be a result of a change in conformation of lactoferrin when it is at the oil/water 517 interface so metal ions are bound at other sites than its metal chelating sites. This binding 518 brings the metal ions in contact with the lipid and results in pro-oxidant effect of lactoferrin 519 (Nagasako, Saito, Tamura, Shimamura, & Tomita, 1993; Nielsen et al., 2004). In conclusion, 520 lactoferrin appeared to have a slightly antioxidative effect in concentrations of 8-12 µm and 521 was a pro-oxidant at higher concentrations. 522

523 Phytic acid

Natural metal chelating substances are present in foods, especially in plant materials. Phytic 524 acid is in the group of natural chelating agents. Food industry and consumers prefer to use 525 natural compounds instead of synthetic ones so in a study Nielsen et al. (2004) tried to use 526 phytic acid in fish oil mayonnaise (16% fish oil, 64% rapeseed oil). However, they found no 527 antioxidative effect of phytic acid in mayonnaise. Lacking antioxidative effect of phytic acid 528 could be due to several reasons: 1. the low pH of mayonnaise that may affect the ability of 529 the phosphoric groups in phytic acid to bind the positively charged Fe ions and 2. a very low 530 formation of the Fe-phytic acid complex. From the available data on phytic acid as an 531 antioxidant in mayonnaise, the efficacy of phytic acid as an antioxidant cannot be concluded 532 so further investigation is needed. 533

534 Mustard

22

535 Mustard is a nutritious food compound (Fahey, Zalcmann, & Talalay, 2001). The characteristic flavour of mayonnaise is principally based on mustard (Depree & Savage, 536 2001). The flavour of mustard derives from a group of isothiocyanates especially allyl 537 538 isothiocynate that are volatile sulphur compounds (Fenwick, Heaney, Mullin, & VanEtten, 1982). They are soluble in oil and slightly soluble in water. In mayonnaise, flavour molecules 539 based on their solubility, partition between oil and aqueous phases. Mustard can act as an 540 emulsifying agent in mayonnaise and stabilise the emulsion (Harrison & Cunningham, 1985). 541 The mustard seed contains natural antioxidants. The antioxidant activity of mustard seed in 542 oil/water emulsions has been studied (McCarthy, Kerry, Kerry, Lynch, & Buckley, 2001; 543 Shahidi, Wanasundara, & Amarowicz, 1994). As meeting the need of using natural 544 antioxidant is important, researchers decided to study the efficacy of mustard as an 545 antioxidant in mayonnaise (Lagunes-Galvez et al., 2002; Milani, Mizani, Ghavami, & 546 Eshratabadi, 2013). In a study on Dijon mayonnaise made with sunflower oil and mustard 547 paste, mayonnaises could be stored for 10 months in closed jars while in mayonnaises 548 without mustard, the oxidation rate was higher and more conjugated dienes were produced. 549 So the protective role of mustard against oxidation in mayonnaise can be concluded from this 550 study (Lagunes-Galvez et al., 2002). Also in another study the effect of different 551 concentrations of yellow powder and paste mustard on rancidity and sensory properties of 552 mayonnaise has been studied (Milani et al., 2013). To eliminate the adverse properties of 553 mustard on mayonnaise such as changes in colour and flavour and also to improve sensory 554 properties of mayonnaise they compared the use of mustard paste, which is made by heating 555 556 treatments, and inactivation of myrosine enzyme to powder mustard. Yellow mustard paste and powder showed antioxidative effect and their antioxidative activity was concentration 557 dependent. Using mustard paste improved sensory properties of mayonnaise by decreasing 558

the pungent flavour also; it made the use of high concentrations of mustard paste (0.75%1.5%) possible.

561 Fenugreek extract

Fenugreek (Trigonella foenumgraecum), is a food ingredient that is used traditionally in the 562 Far East. It is recognised as a possible source of natural antioxidant. The effectiveness of 563 fenugreek extracts in inhibiting/minimizing lipid oxidation in comparison with synthetic 564 antioxidant (TBHQ and BHT) has been studied in mayonnaise (Mostafa, 2003). This study 565 stated that fenugreek extracts at 500 mg/kg in mayonnaise showed the same antioxidant 566 effect as TBHQ (200 mg/kg) but was more effective than BHT (200 mg/kg). Although high 567 concentration of fenugreek (1500 mg/kg) was more effective than synthetic antioxidants, it 568 had a diverse effect on sensory characteristics of mayonnaise. So based on this study 569 fenugreek at 500 mg/kg can be used as a natural antioxidant in mayonnaise but further studies 570 on fenugreek are essential. 571

572 Black glutinous rice

Black glutinous rice is generally used as an ingredient in snacks and desserts. It contains high 573 amounts of phenolic compounds especially anthocyanins in pericarp that has the antioxidant 574 ability and radical scavenging (Hu, Zawistowski, Ling, & Kitts, 2003; Ichikawa et al., 2001). 575 Optimum condition of solvent extraction of black glutinous rice crude extract and its 576 application in fish oil mayonnaise was studied (Tananuwong & Tewaruth, 2010). The highest 577 antioxidant activity was obtained when extracted twice with 70:30 acetone-water mixture 578 (v/v) at pH 6.8 for 4 hours of total extraction times. 1000 mg/kg crude extract could 579 efficiently increase oxidative stability of mayonnaises. Phenolic compounds in black 580 glutinous rice can retard oxidation with three mechanisms: 1. chain breaking, 2. 581 hydroperoxide destroying and 3. metal chelating. In spite of good antioxidative ability of high 582 concentrations of crude extract, greater colour deterioration could be seen. The reasons for 583

the colour changes in mayonnaise might be a result of: 1. maillard reaction and 2. oxidative
degradation of anthocyanin to undesirable brown-coloured products.

586 Lycopene

Lycopene is a natural food colorant. It is incorporated in dairy beverages, powdered 587 beverages, dairy foods, surimi, confectionery, bakery, breakfast cereals, nutritional bars, 588 soups, meal replacement, sauces, salsas, pastas, chips, snacks, dips and spreads. Lycopene 589 crystals from tomato waste skin (50 mg/kg) were used in mayonnaise and its effect on 590 oxidative stability of mayonnaise was studied during storage for four months (Kaur, Wani, 591 Singh, & Sogi, 2011). Lycopene slowed down the development of off-flavour, off-odours, 592 and colour changes and acted as an antioxidant by interrupting the chain of free radicals 593 involved in auto-oxidation. Also, sensory analysis showed that lycopene-treated mayonnaises 594 595 had good consumer acceptability.

596 Ginger powder

Ginger contains polyphenol compounds (6-gingerol and its derivatives), which have a high 597 antioxidant activity (Chen, Kuo, Wu, & Ho, 1986) like TBHQ, BHA and BHT. Water ginger 598 extract is a strong radical scavenger (Y. F. Kishk & El Sheshetawy, 2010). The role of ginger 599 powder on oxidative stability of mayonnaise has been investigated (Y. Kishk & 600 Elsheshetawy, 2013). This study showed that ginger powder at concentrations of 1.0% and 601 1.25% reduced the production of primary and secondary oxidation products (measured by 602 anisidine value) and subsequently retarded oxidation process during storage for 20 weeks. 603 The rheological properties of mayonnaise were not influenced by ginger powder at 604 concentrations mentioned above. Interestingly, ginger powder improved the taste, flavour, 605 mouth feel, and overall acceptability of mayonnaise at zero time, and after 20 weeks the 606 overall acceptability of mayonnaise samples at concentration of 1.0% and 1.25% was 607 improved. 608

609 Grape seed extract

Grape seed is a by-product from wine production that contains catechin and 610 proanthocyanidins. It has radical scavenging and antioxidative properties. The potential use 611 of grape seed extract on the oxidative stability and sensory properties of mayonnaise during 612 storage for 8 weeks has been studied (Altunkaya et al., 2013). High concentrations of grape 613 seed extract (0.15%) showed a very good antioxidative activity. However, mayonnaises 614 without grape seed extract had a higher sensorial acceptability. On the other hand high 615 concentrations of grape seed, extracts increase toxicological risk. Therefore, 0.50 mg/ml did 616 not establish toxicological health and improved oxidative stability with acceptable sensory 617 properties. 618

619 Essential oils extracted from Zenyan

*Carum copticum* is an annual herbaceous plant that grows in the east of India, Iran and Egypt. 620 Its fruits (generally known as "Zenyan" in Iran) have been used extensively in Iranian folk 621 and traditional medicine (Goudarzi, Saharkhiz, Sattari, & Zomorodian, 2010). The potential 622 antioxidative ability of essential oils from Zenyan in mayonnaise has been studied. Also the 623 differences between the antioxidative activity of two different extraction methods (ohmic 624 assisted hydro distillation and conventional hydro distillation) were compared (Mazaheri 625 Tehrani, 2013). The results from this study showed that the antioxidant activity of Zenyan 626 essential oils was independent on extraction methods. However, they suggested ohmic 627 assisted hydro distillation method for extraction of essential oils was more efficient than 628 traditional hydro distillation method as in this method more time and energy will be saved. 629 All concentration of essential oils (0.015%, 0.03% and 0.045%) showed antioxidant activity, 630 against DPPH free radicals, in mayonnaise. They proposed that BHA and BHT (synthetic 631 antioxidants used in mayonnaise) could be replaced by high concentration of Zenyan 632 essential oils. Antioxidative property of Zenyan essential oils mainly is dependent on its 633

thymol content. Thymol is a main component of essential oils and acts as a primary
antioxidant by delaying or preventing the initiation and propagation step (Hashemi,
Niakousari, & Saharkhiz, 2011). The colour of mayonnaises was not influenced by addition
of Zenyan essential oils. However, the odour of mayonnaises with and without Zenyan was
significantly different. Whereas, in the case of preference test there were no differences
between all represented samples (Mazaheri Tehrani, 2013).

640 Chitosan

The antioxidant activity of chitosan derivatives has been reported in several studies but few studies have been done on real foodstuff. Antioxidant activity of chitosan with different molecular weighs was evaluated in mayonnaise (García, Silva, & Casariego, 2014). This study indicated that addition of chitosan not only retarded lipid oxidation of mayonnaises during 63 days of accelerated storage but also improved organoleptic properties (odour and taste). Mayonnaises treated with chitosan with the bigger molecular weight showed better stability during accelerated storage at all temperatures.

648 Tansy extracts

Tansy (Tanacetum vulgare L., Asteraceae) is an aromatic plant spread mainly in the northern hemisphere in Europe, Asia and North America. Its anti-inflammatory, antibactericidal, antifungicidal, insects' repellent and antioxidative activities is reported. Tansy acetone extract at higher concentrations could inhibit oxidation in rapeseed oil (Pukalskas & Venskutonis, 2000). The oxidative stability of mayonnaise treated with tenacy extract was studied. This study showed that all tansy extracts improved oxidative stability of mayonnaise (Baranauskienė, Kazernavičiūtė, Pukalskienė, Maždžierienė, & Venskutonis, 2014).

656 Clove

657 Cloves (Syzygium aromaticum Linn) are aromatic dried flower buds of the family Myrtaceae.658 Generally, clove extracts are used as biopreservatives in preventing food spoilage by

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659 pathogenic contaminants. The potential application of eugenol-lean fraction of clove buds as a flavouring agent (replacing mustard in the classic formulation) and as a natural antioxidant 660 in mayonnaise was studied (Chatterjee & Bhattacharjee, 2014). Mayonnaise incorporated 661 with eugenol-lean clove extract was found to be oxidative stable beyond 6 months. Also had 662 better physical properties such as higher colour intensity, lower thermal and nonthermal 663 creaming, homogenous and compact microstructure and higher consistency index. 664 Organoleptically, addition of eugenol-lean clove extract did not cause any significant changes 665 in body and consistency of all the mayonnaise samples. 666

667 Anthocyanin extracted from purple corn husk

Purple corn cobs, purple sweet potatoes and blueberries are usually the main source of 668 anthocyanins employed as food colorant in commercial production (Kähkönen, Hopia, & 669 Heinonen, 2001). Anthocyanins possess strong antioxidant capacity because they have free 670 radical scavenging (Espin, Soler-Rivas, Wichers, & García-Viguera, 2000) and metal-671 chelating properties (Nam et al., 2006). Purple corn husk has 10 times more anthocyanins 672 than other plant sources of anthocyanins (Li et al., 2008). The feasibility of the use of 673 anthocyanins extracted from purple corn husk extract (PCHE) as a natural antioxidant in 674 comparison with the synthetic antioxidants (BHT and EDTA) was studied (Li et al., 2014). 675 Addition of PCHE reduced the amount of both primary and secondary oxidation products. 676 The antioxidative activity of PCHE was concentration dependent and the mayonnaise 677 containing 0.4 g/kg PCHE showed the strongest antioxidative performance during storage. 678 This study proposed that PCHE could be used as a natural antioxidant in mayonnaise instead 679 of synthetic antioxidants like BHT and EDTA. This study considered the colour difference 680 (mayonnaises with PCHE had purplish colour) as a positive point and related it to the sign of 681 natural antioxidant in consumers' food (Figure 2). 682

683 Seaweed

684 Plants and marine algae can be used as a source of natural antioxidants. Not only these antioxidants show potential for improving oxidative stability of food but also they have a 685 broad array of health-promoting benefits (Hata, et al., 2001; Kim et al., 2009). Seaweed 686 contains different tocopherols, amino acids, sulphated polysaccharides, mono- and 687 polyphenols, and bioactive compounds (Honold et al., 2015). Seaweed extracts from Fucus 688 vesiculosus as a potential antioxidant was used in mayonnaise (Hermund et al., 2015; Honold 689 et al., 2015). Fucus vesiculosus is rich in polyphenols that can act as free radical scavengers 690 and metal chelators. Hermund et al. (2015) evaluated the antioxidant properties of water 691 extract (WE) and an ethyl acetate fraction (EAF) of Fucus vesiculosus and studied their 692 efficacy to inhibit lipid oxidation in fish oil mayonnaise. 2 g/kg of WE was more efficient in 693 lowering the formation of primary oxidation products and secondary oxidation products 694 however, EAF was more efficient in decreasing the degradation of the n-3 PUFA. The 695 effectiveness of EAF and WE was concentration dependant. It was shown that the antioxidant 696 effectiveness of EAF and WE was related to high-total phenolic content, high-radical 697 scavenging activity, moderate- or high-metal chelating ability and high-carotenoid content in 698 the extract. In another study, the antioxidant properties of four extracts (ethanol (EtE), 699 acetone (AcE), and water extracts (WE)) from *Fucus vesiculosus* in fish oil mayonnaise was 700 701 studied (Honold et al., 2015). Ethanol and acetone extracts had higher antioxidant efficacy compared to water extracts. In this study, the concentration dependency of antioxidant 702 efficacy was observed too. Same as Hermund et al. (2015) study, Honold et al. (2015) 703 indicated the relation between the high-total phenolic content and high-radical scavenging 704 activity with antioxidant efficacy but also they pointed out the location of phenolic 705 compounds at the interface of oil droplets and water in mayonnaise. 706

707 Glucose oxidase (GOX)

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708 Glucose oxidase is an enzyme that acts as a natural antioxidant in food products. Glucose oxidase can be isolated and purified from the mould Aspergillus niger (Whitaker, 1993). It is 709 tightly bound to the mycelium so it is difficult to separate it from enzyme catalase (CAT) 710 (Crueger & Crueger, 1990). Hence, in food grade preparation of glucose oxidase it is usually 711 coupled with catalyse. GOX/CAT has the ability to scavenge molecular oxygen by catalysing 712 the reaction of converting two moles of glucose and one mole of molecular oxygen to two 713 moles of  $\delta$ -gluconolactone which spontaneously hydrolyses to gluconic acid and hydrogen 714 peroxide that can be removed by catalase (Crueger & Crueger, 1990; Frankel, 2005). Glucose 715 oxidase has been utilised as an antioxidant in different mayonnaise systems. In mayonnaise 716 containing 790 g/kg soy oil, 450 U/Kg of GOX could retard off-flavour, off-odour and rancid 717 718 taste in dark at 20 °C (Skrede, RØtbotten, & Baardseth, 1991). In fish oil mayonnaise GOX/CAT system acted as an oxygen scavenger and prolonged the shelf life of the product 719 at refrigerator temperature but at room temperature the mayonnaise with and without glucose 720 oxidase showed no detectable differences (Jafar et al., 1994). To see if the enzyme system 721 722 was active at room and refrigerator temperature and to get a better understanding of GOX/CAT activity in mayonnaise Isaksen and Adler-Nissen (1997) investigated the effect of 723 GOX/CAT system in fish oil mayonnaise and soybean oil mayonnaise. They found that 724 GOX/CAT system retarded lipid oxidation at 5 °C and 25 °C. As they expected GOX/CAT 725 scavenged oxygen in the packages under the consumption of glucose. Although GOX/CAT 726 could retard lipid oxidation, using it as antioxidant in mayonnaise cannot be practical because 727 of formation of off-flavour in both mayonnaises at 5 °C and 25 °C. So in order to solve this 728 problem further investigation is needed. 729

- 730 Factors affecting the activity of antioxidants
- 731 Polar paradox and cut off theory

732 The effectiveness of an antioxidant in oil/water emulsion is highly affected by its partitioning properties and its ability to be located in the environment where lipid oxidation takes place 733 that could be at the interface between the oil and water phases (Coupland & McClements, 734 1996). Mayonnaise is a heterophasic food system, antioxidants may partition into at least 735 three different phases, the aqueous phase, the oil phase and the interface between oil and 736 737 water phase (Jacobsen, Schwarz, et al., 1999). In mayonnaise lipophilic antioxidants like tocopherol are located in the oil phase while the more polar antioxidants such as gallic acid 738 (80% of gallic acid) are concentrated in the aqueous phase but a significant proportion of 739 them distributed into interface (20% of gallic acid) (Jacobsen, Schwarz, et al., 1999). The 740 relationship between antioxidant partitioning and antioxidant efficacy is also called "the polar 741 paradox" (Figure 3) (Frankel, Huang, Kanner, & German, 1994; Laguerre et al., 2013). 742 According to the so-called "polar paradox" theory, polar antioxidants would be more 743 effective than their nonpolar analogues in bulk oil, whereas nonpolar antioxidants would be 744 745 more effective in oil in water emulsion than their polar counterparts would. Gallic acid which is a polar antioxidant was added in mayonnaise (Jacobsen, Hartvigsen, Thomsen, et al., 746 2001). Gallic acid showed a poor antioxidative effect and these results were in accordance 747 with the theory of polar paradox that the efficacy of polar antioxidants in oil in water 748 emulsion is poor (Jacobsen, Hartvigsen, Thomsen, et al., 2001). To get more insights into the 749 efficacy of the polar paradox theory in mayonnaise researchers tried two pairs of homologue 750 antioxidants (ascorbic acid/ascorbyl palmitate and gallic acid/propyl gallate) in fish oil 751 enriched mayonnaise (Jacobsen, Hartvigsen, et al., 1999; Jacobsen, Hartvigsen, Thomsen, et 752 al., 2001; Jacobsen et al., 2008). In these studies, surprisingly all employed compounds acted 753 as pro-oxidants and the antioxidant activity did not improve with increased lipophilicity. 754

From these reports authors concluded that in much more complex systems like mayonnaise, in which iron stemming from the egg yolk catalyses lipid oxidation and many different molecules can affect antioxidant activity polar paradox theory cannot predict the antioxidant efficacy.

Moreover there is another theory called "cut off" that shows the nonlinear influence of 759 hydrophobicity on antioxidant capacity (Laguerre et al., 2013). Lipophilization of 760 antioxidants can improve antioxidant efficacy. However, there are not many studies on their 761 effectiveness in real food systems such as mayonnaise. Alemán et al. (2015) investigated the 762 antioxidant effect of caffeic and its esters (caffeates C1-C18) in fish oil mayonnaise. Both 763 caffeic acid and caffeates had antioxidative effect in fish oil mayonnaise. The caffeates with 764 short to medium alkyl chain (butyl, octyl and dodecyl) were the most effective antioxidants. 765 Whereas, the increase in alkyl chain caused a collapse in the antioxidant capacity of esterified 766 phenolic compounds. This phenomenon can be explained by the cut off theory. The caffeates 767 retarded the formation of both primary and secondary oxidation products in fish oil 768 769 mayonnaise.

#### 770 Conclusion and future perspectives

This review has highlighted the important factors affecting lipid oxidation in mayonnaise. A 771 general conclusion is that we can reduce oxidation in mayonnaise by cutting of the factors 772 that reduce the induction period and accelerate rancidity such as lowering the storage 773 temperature, reducing oxygen concentration in food, avoiding intensive lighting, 774 manipulating chemical structure of the oil and physical properties of mayonnaise. In addition, 775 one of the promising ways of retarding oxidation in mayonnaise could be using antioxidants. 776 As people are more concerned about their health there is a worldwide trend toward using 777 natural antioxidants in food products. After reviewing the literature about using natural 778 antioxidants in mayonnaise, it can be concluded that it could be possible to design a 779

780 mayonnaise with greater oxidative stability, by replacing synthetic antioxidants with natural ones. However, as mayonnaise contains a variety of different components still there is a 781 scarcity of knowledge on the influence of these components on the efficacy of natural 782 antioxidant activity. Further elucidation of the mechanism of oxidation in mayonnaise and a 783 better understanding of antioxidant efficacy would have a great technological importance. 784 The importance of interfacial characteristic of oil droplets has been already demonstrated but 785 using new strategies for retarding lipid oxidation by manipulating the interfacial properties of 786 oil droplet in mayonnaise is missing. The challenge in using natural antioxidants in 787 mayonnaise is to obtain a product with good sensory properties and satisfactory shelf-life 788 hence further studies of the influence of natural antioxidant on the sensory properties of 789 790 mayonnaise are required. After gathering information on using natural antioxidants in mayonnaise we came to a conclusion that in future more studies should be done on issues 791 like: elucidating the mechanism of oxidation in mayonnaise in order to find the best 792 antioxidant, more studies on the factors affecting antioxidant efficacy in mayonnaise like 793 systems, manipulating interfacial area of oil in water emulsion droplets to decrease rate of 794 oxidation, finding more sources of natural antioxidants and finally working on how to obtain 795 the best sensory properties with using natural antioxidants. In our laboratory we currently are 796 working on elucidating process of oxidation in mayonnaise stored at different temperatures 797 and finding new sources of natural antioxidants as a potential alternatives of synthetic 798 antioxidant. 799

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Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Fish oil mayonnaise (70%)	TBHQ (0.02%)	There were few signs of oxidation in the deodorized and stabilized fish oil mayonnaise at 2 °C and 14 weeks of storage.	(YT. L. Hsieh, 1990)
Fish oil mayonnaise (70%)	TBHQ (0.08%) Ascorbic Acid (0.5%)	TBHQ was a successful antioxidant for fish oil mayonnaise while ascorbic acid had pro- oxidant effect in fish oil mayonnaise	(Y. T. Hsieh & Regenstein, 1991)
Soy oil mayonnaise (790 g/kg soy oil)	Glucose oxidase (GOX) (450 units per kg (U/kg), 200 U/kg)	450 U/Kg of GOX could retard rancid taste, off-flavour and off-odour developments in mayonnaise stored in dark at 20 °C for 8 months, whereas 200 U/Kg of GOX could not prevent deterioration of the organoleptic quality.	(Skrede et al., 1991)
Fish oil mayonnaise Soy oil mayonnaise Corn oil mayonnaise	TBHQ (0.08%)	In fish oil mayonnaise TBHQ was effective at early storage times in lengthening the period before oxidation was initiated.	(YT. L. Hsieh & Regenstein, 1992)

# Table 1. Summary of the use of natural and synthetic antioxidants in mayonnaise

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Fish oil mayonnaise (menhaden and capelin oils, 100% fish oil)	Propyl gallate, citric acid, EDTA, BHT, BHA, ascorbic acid, carboxymethylcellulose, sodium tripolyphosphate, lecithin, superoxide dismutase, catalase, and GOX/CAT.	GOX/CAT had a significant effect on reduction of the development of off-odour when mayonnaise was stored at refrigerator temperature, but at room temperature, the enzymes were not effective. Citric acid or sodium citrate and propyl gallate in the oil phase and EDTA and ascorbic acid in the aqueous phase increased the shelf-life to an average of 49 days at room temperature	(Jafar et al., 1994)
Fish oil mayonnaise (16% fish oil)	Antioxidant system: ascorbic acid, lecithin and gamma tocopherol (A/L/T) (4 g/kg mayonnaise)	Addition of A/L/T system resulted in strong fishy and rancid off-flavours of mayonnaise despite low peroxide and anisidine values.	(Meyer & Jacobsen, 1996)

Product	Antioxidant/pro-oxidant	Results	Reference
	(Concentration)		
Fish oil mayonnaise (0, 0.25 and 0.50 of the vegetable oil was substituted by fish oil corresponding to contents of 0, 200 and 400 g/kg) Soybean oil mayonnaise	Glucose oxidase/catalase (GOX/CAT) (0, 400 and 800 units/kg)	The enzyme system of GOX/CAT could reduce lipid oxidation in mayonnaise both sored at 25 °C and 5 °C in soybean oil and fish oil mayonnaise.	(Isaksen & Adler- Nissen, 1997)
Fish oil mayonnaise (16% fish oil)	Propyl gallate, oil soluble (Grindox 370) and Propyl gallate, water dispersible (Grindox 413)	propyl gallate was a weak pro-oxidant.	(Jacobsen, Hartvigsen, et al., 1999)
Fish oil mayonnaise (16% fish oil)	ascorbic acid (0-800 mg/kg)	Ascorbic acid increased the formation of fishy off-flavours in fresh mayonnaise.	(Jacobsen, Adler- Nissen, et al., 1999)
Fish oil mayonnaise (16% fish oil)	$\alpha$ -, $\beta$ -, and $\gamma$ -tocopherol and six polar antioxidants (trolox, ferulic acid, caffeic acid, propyl gallate, gallic acid, and catechin)	Antioxidants partitioned in accordance with their chemical structure and polarity.	(Jacobsen, Schwarz, et al., 1999)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Fish oil mayonnaise (16% fish oil)	A/L/T, and two commercial mixtures of tocopherol, an oil-soluble (Toco 70) preparation and a water- soluble (Grindox 1032)	Addition of A/L/T system caused the immediate formation of distinct fishy and rancid off-flavours in the fresh mayonnaises Addition of Toco 70 did not affect the sensory perception of mayonnaise nor the development of volatile off-flavour compounds, but the peroxide values were slightly increased as compared to the other mayonnaises. Mayonnaise with Grindox 1032 seemed to have fewer fishy and rancid off-flavours than mayonnaises without antioxidant.	(Jacobsen, Adler- Nissen, et al., 2000)
Fish oil mayonnaise (16% fish oil)	EDTA (0, 50, 75, 125, and 200 mg/g)	EDTA (down to 50 mg/g) efficiently delayed oxidation in fish oil enriched mayonnaise.	(Thomsen, Jacobsen, et al., 2000)
Fish oil mayonnaise (16% fish oil)	EDTA (0 and 75 mg/kg)	A significant antioxidative effect of EDTA for lipid samples from mayonnaise was noted.	Thomsen et al., 2000 (Thomsen, Kristensen, et al., 2000)

Product	Antioxidant/pro-oxidant	Results	Reference
	Water-dispersible	R '	
Fish oil mayonnaise (16% fish oil)	Grindox 1032 (20–280 mg/kg) Oil-soluble tocopherol preparation, Toco 70 (20–	Addition of tocopherol to a mayonnaise that already contained tocopherol (600 $\mu$ g/g) either had no effect or increased oxidation.	(Jacobsen, Hartvigsen, Lund, et al., 2001)
Fish oil mayonnaise (16% fish oil)	280 mg/kg) Ascorbic acid (0-4000 mg/kg)	Ascorbic acid promoted formation of volatile oxidation compounds and reduced the peroxide value in mayonnaises.	(Jacobsen, Timm, et al., 2001)
Fish oil mayonnaise (16% fish oil)	Gallic acid (200 mg/kg) EDTA (200 mg/kg)	EDTA was an efficient antioxidant in fish oil enriched mayonnaise but gallic acid was a poor antioxidant.	(Jacobsen, Hartvigsen, Thomsen, et al., 2001)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Dijon mayonnaise	EDTA (0.004%) Rosemary extracts (0.03%) Mustard (5%)	Addition of antioxidant (rosemary extract and EDTA) decreased the level of photooxidative volatiles in the headspace. In the absence of mustard, the oxidative degradation was somewhat faster and the amount of conjugated dienes was increased more quickly and to a higher degree than that in the mustard-containing sample.	(Lagunes-Galvez et al., 2002)
Mayonnaise based on specific structured lipid (SL) from sunflower oil and caprylic acid	lactoferrin, (800 mg/kg ~ 10 μM) Propyl gallate, (200 mg/kg) EDTA, (75 mg/kg)	EDTA was a strong antioxidant, while propyl gallate and lactoferrin did not exert any antioxidative effect in the SL mayonnaise	(Jacobsen et al., 2003)
Mayonnaise	TBHQ (200 mg/kg) BHT (200 mg/kg) Fenugreek extract (FE) (500, 1000, 1500 mg/kg)	FE at 500 mg/kg in mayonnaise was as effective in decreasing lipid oxidation as TBHQ and more effective than BHT at their permitted level (200 mg/kg).	(Mostafa, 2003)
Fish oil mayonnaise (16% fish oil, 64% rapeseed oil, 80% fat)	Lactoferrin (8-32 µM), Phytic acid (16-124 µM) EDTA (16-64 µM)	The antioxidative effect of EDTA was much more pronounced than the effect of lactoferrin and, especially, phytic acid in mayonnaise.	(Nielsen et al., 2004)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Fish oil-enriched mayonnaise- based salads (shrimp and tuna salads)	Oregano (1%) Rosemary (1%) Thyme (1%)	In fish oil-enriched shrimp salad, asparagus had an anti-oxidative effect and shrimp a pro- oxidative effect. The addition of spices increased the oxidative stability of tuna salad (oregano>rosemary>thyme).	(Sørensen, Nielsen, & Jacobsen, 2010)
Fish oil mayonnaise	Black glutinous rice flour (500 mg/kg and 1000 mg/kg (oil weight basis))	The addition of dried black glutinous rice crude extract at 500 and 1000 mg/kg (oil weight basis) could retard an increase in conjugated diene hydroperoxides and thiobarbituric acid reactive substances (TBARs)	(Tananuwong & Tewaruth, 2010)
Mayonnaise (rapeseed oil, 74% fat)	Lycopene crystals (50 mg/kg)	Lycopene slowed the development of off- flavour, off-odour, and colour changes in lycopene-added butter, ice cream, and mayonnaise during storage as it interrupts the chain of free radicals involved in auto- oxidation	(Kaur et al., 2011)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Mayonnaise and salad dressing (olive oil 50% and 25%)	Natural spices and herbs such as parsley, ground black pepper, basil and hot paprika) and their extracts	Natural spices and herbs were replaced with their extracts. Pure olive oil, mayonnaise and salad dressing containing extracts had better microbiology and oxidative quality. Also sensory properties of mayonnaise with extracts showed that it had the highest score of the degree of liking.	(Slavchev, Nikovska, & Nenov, 2012)
Rice bran oil mayonnaise	Oryzanol Squalene Tocopherols Tocotrienols	Incorporation of Rice bran oil provides oryzanol (0.8g/100g oil), tocopherol, tocotrienol, squalene which enhances the stability along with providing balanced Fatty acid composition to the mayonnaise.	(Das, Bhattacharya, Kar, Ghosh, & Bhattacharyya, 2013)
Mayonnaise (corn oil)	Ginger powder (GP) (0% - 1.25%)	The addition of GP at concentrations 1.0% and 1.25% could improve oxidative stability of mayonnaise. After 20 weeks, the values of peroxide, anisidine, acid and totox for mayonnaise prepared using 1.0% and 1.25% GP were significantly lower compared to the control.	(Y. Kishk & Elsheshetawy, 2013)

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Product	(Concentration)	Kesuits	Kelerence
Mayonnaise (rapeseed oil)	Grape seed extract (GSE) (0 mg GSE per ml, 0.5 mg GSE per ml (~0.050%), 0.9 mg GSE per ml (~0.10%) and 1.4 mg GSE per ml (~0.15%))	The oxidative stability of the mayonnaises enriched with GSE was slightly improved through storage.	(Altunkaya et al., 2013)
Mayonnaise (corn oil 70%)	Juice of basil leaves (0.5%, 1.0% and 1.5%) BHT (0.01%)	Addition of (1.0% and 1.5%) juice of basil leaves could reduce the oxidation process of mayonnaise during 12 weeks.	(Abou-Zaid, Abdelahafez, & Amer, 2015)
Mayonnaise (sunflower oil, 68% fat)	Essential       oils       (EOs) $(0.015\%)$ $0.03\%$ and $0.045\%$ extracted       from         Zenyan       by       Ohmic       sisted         hydro       distillation       and         convention       hydro       hydro         distillation       hydro         BHA       HT	All concentrations of Zenyan EOs were suitable antioxidants but synthetic antioxidants like: BHA and BHT could be replaced by higher concentrations of Zenyan Eos.	(Mazaheri Tehrani, 2013)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Mayonnaise (Soybean oil, 63% fat)	Yellow powder mustard (0%, 0.01%, 0.02%, 0.03%, 0.04%, 0.05% and 1%,) Paste mustard (0%, 0.75%, 1%, 1.25% and 1.5%)	Powder mustard increased oxidative stability but caused undesired changes in colour and flavour of mayonnaise for this reason they substituted powder mustard with paste mustard so undesired changes in colour and flavour of the sauce were removed to some extent.	(Milani et al., 2013)
Mayonnaise	Chitosan with bigger molecular weight (MW=310 kDa and DD=77.7%) (100 mg/kg) Chitosan with smaller molecular weight (MW=123 kDa and DD=83.2%) (100 mg/kg) EDTA (75 mg/kg)	Addition of chitosan slowed down the lipid oxidation process of mayonnaises. Chitosan with bigger molecular weight showed better stability during accelerated storage at all temperatures. It has been observed that addition of chitosan slowed down the lipid oxidation process of mayonnaises during 63 days of accelerated storage.	(García et al., 2014)
Mayonnaise (soy oil)	Tocopherol (450 mg/kg) TBHQ (150 mg/kg)	Tocopherol limited hydroperoxide formation effectively.	(Shahin, Nayebzadeh, Alizadeh, & Mohammadi, 2014)

	Antioxidant/pro-oxidant		<b>D</b> 4
Product	(Concentration)	Results	Keterence
Mayonnaisa	Taney extracts (1%)	Tansy extracts increased oxidative stability of	(Baranauskienė et al.,
Mayonnaise	Tailsy extracts (170)	mayonnaise.	2014)
	Anthocyanin-rich purple		
	corn husk extract (PCHE)	The antioxidative effect of the mayonnaise	
	(anthocyanin concentrations	containing PCHE was higher than that of	
Mayonnaise (Soybean oil 85%	of 0.1, 0.2, and 0.4 g/kg	mayonnaise with chemical antioxidants BHT	$(\mathbf{L}; \mathbf{a}; \mathbf{a}) = 2014$
fat)	mayonnaise)	and EDTA as positive control.	(LI et al., 2014)
	BHT (0.2 g/kg mayonnaise)	The strongest antioxidative performance was	
	EDTA (0.075 g/kg	in mayonnaise containing 0.4 g/kg PCHE.	
	mayonnaise)		

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Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Fish oil mayonnaise (16% fish oil)	Caffeic acid and lipophilised derivatives of caffeic acid (caffeates): Methyl caffeate (100µM) Butyl caffeate (100µM) Octyl caffeate (100, 200 µM) Dodecyl caffeate (100µM) Octadecyl caffeate (100µM)	Caffeic acid esterified with fatty alcohols of different chain lengths (C1–C20) were better antioxidants than the original phenolic compound. Fish oil enriched mayonnaise (stored for 4 weeks at 20 °C) with caffeates of medium alkyl chain length (butyl, octyl and dodecyl) added resulted in a better oxidative stability than caffeates with shorter (methyl) or longer (octadecyl) alkyl chains. For peroxide value of mayonnaises the shorter lag phase (3 days) was in samples without antioxidant and octyl caffeate at 200 µM and the longest lag phase (9 days) was seen in samples containing butyl caffeate and octadecyl caffeate	(Alemán et al., 2015)

Product	Antioxidant/pro-oxidant (Concentration)	Results	Reference
Mayonnaise	Sesame sprouts (0.5%, 0.75%, 1.0% and 1.25%) EDTA (0.0075%) BHT (0.02%)	Sesame sprouts powder retarded oxidation during 45 days (at $25\pm5$ °C) in mayonnaise but did not have good sensory perception.	(Shabbir et al., 2015)
Mayonnaise (Soybean oil 74% fat)	Eugenol-lean fraction isolated from clove buds (Syzygium aromaticum Linn) (0.42 %)	Mayonnaise formulated with eugenol-lean clove extract had significantly higher antioxidant activity than mustard mayonnaise. The antioxidant activity and phytochemical properties tend to decrease after 30 days for the reference market sample and after 90 days for the experimental control sample while the mayonnaise formulated with eugenol-lean clove extract was found to be stable beyond 6 months.	(Chatterjee & Bhattacharjee, 2015)
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Figure 1. Proposed mechanism for the release of Iron by Ascorbic Acid. Adopted from (Jacobsen, Adler-Nissen, et al., 1999)

Figure 2. Different colours of mayonnaise containing various PCHE contents: (A) PCHE 0.4 g/kg, (B) PCHE 0.3 g/kg, (C) PCHE 0.2 g/kg, and (D) PCHE 0.1 g/kg. Adopted from (Li et al., 2014).

Figure 3. Interfacial phenomena as a possible mechanism of action of the polar paradox in oil-in-water emulsion (a and b) and in bulk oil (c and d). Adopted from (Edwin N Frankel, Huang, Kanner, & German, 1994; Laguerre et al., 2013).

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		+ ascorbic acid		
Phosvitin-Fe3+	$\rightleftharpoons$ Fe <sup>3+</sup>	⋧	Ascorbic acid-Fe2+	
$\sqrt{\frac{1}{3}}$ + ascorbic acid	(1)	(2)		
Phosvitin-Fe <sup>2+</sup>	$\rightleftharpoons\ Fe^{2*}$	$\stackrel{\text{+ ascorbic}}{\rightleftharpoons}$	Ascorbic acid-Fe <sup>2+</sup>	
	(1)	(2)	Q	
K(1) = [Phosvitin-]	Fe <sup>3+</sup> ]/([Fe <sup>3+</sup> ][Ph	osvitin])		

Figure 1.





Figure 3.

# Highlights

- It is possible to increase oxidative stability of mayonnaise by replacing synthetic antioxidants with natural antioxidants
- The most challenging part of using natural antioxidants in mayonnaise is to obtain a product with a good sensory property.
- Factors affecting efficacy of natural antioxidants in mayonnaise are not very well understood.
- Manipulating interfacial layer of oil droplet in mayonnaise to retard lipid oxidation is missing in the literature.

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