

Physical properties and oxidative stability of mayonnaises fortified with natural deep eutectic solvent, either alone or enriched with pigmented rice bran

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ABSTRACT

This article explores the novel use of natural deep eutectic solvents (NaDES) in real food by incorporating them into mayonnaise, either alone or with pigmented rice bran (RB). Results showed that NaDES-fortified mayonnaises could prevent lipid oxidation. Notably, mayonnaises with NaDES2 (betaine:sucrose:water) significantly reduced the production of lipid hydroperoxides, which was maintained to an average of 2.6 mmol LOOH/kg oil, which is 2.9 times lower than the control (7.5 mmol LOOH/kg oil), or 7.4 times lower than mayonnaise with citric acid (19.1 mmol LOOH/kg oil). NaDES2-fortified mayonnaises maintained high tocopherols levels (0.97 g/Kg oil) and reduced volatile compounds from secondary lipid oxidation. This effect may result from NaDES altering the aqueous phase properties of mayonnaise, notably by reducing water activity by ~0.1. Finally, pre-enrichment of the NaDES phase with bioactive molecules (e.g. from pigmented RB) represents an innovative perspective to promote the health benefits of formulated foods.

1. Introduction

Mayonnaise, which is an oil-in water emulsion, is one of the most common sauces used in foods such as salads, sandwiches, and hamburgers. Lipids are the major component in mayonnaises, which constitute 60–80 % of its composition. Other key ingredients include egg yolk, sugar, salt, vinegar, and gum. Vinegar and organic acids (e.g. malic

acid, tartaric acid, and citric acid) are used in mayonnaise to improve texture by thickening and to increase acidity (Panovska et al., 2012; Raurich et al., 2020), thereby preventing the growth of microorganisms such as molds and yeasts and reducing the risk of pathogenic contamination (Hakimian et al., 2022; Ozdemir et al., 2021; Teneva et al., 2021). The reduction of pH in the mayonnaise affects its composition, particularly the egg yolk, which contains metal ions within its protein matrix

Abbreviations: C, Degree Celsius; a*, Red/Green; a_w, Water activity; b*, Yellow/Blue; B + NaDES1-M, Mayonnaise fortified with NaDES1 enriched with Niaw Dum Mor rice bran; B + NaDES2-M, Mayonnaise fortified with NaDES2 enriched with Niaw Dum Mor rice bran; CA-M, Control mayonnaise with citric acid; CIS, Cold Injection System; cm⁻¹, Wavenumber; cyd-3-glu, cyanidin-3-O-glucoside; DES, Deep eutectic solvent; DHS, Dynamic headspace; DLS, Droplet size; EDTA, Ethylenediaminetetraacetic acid; EDTA-M, Mayonnaise with 100 ppm of EDTA; FLD, Fluorescence detector; g, Gram; GC, Gas chromatography; HBA, Hydrogen bond acceptor; HBD, Hydrogen bond donors; HPLC, High-performance liquid chromatography; Kg, Kilogram; L, Liter; L*, Lightness; LOOH, Lipid hydroperoxides; M, Control mayonnaise; min, minute; mL, Milliliter; mm, Millimeter; mmol, Millimole; MS, Mass spectrometry; NaDES, Natural deep eutectic solvent; NaDES1-M, Mayonnaise fortified with NaDES1; NaDES2-M, Mayonnaise fortified with NaDES2; ng, nanogram; nm, nanometer; PDA, Photodiode array; PDI, Polydispersity index; R + NaDES1-M, Mayonnaise fortified with NaDES1 enriched with Hawm Gra Dang Ngah rice bran; R + NaDES2-M, Mayonnaise fortified with NaDES2 enriched with Hawm Gra Dang Ngah rice bran; RB, Rice bran; rpm, round per minute; TDU, Thermal Desorption Unit; TPP, Triphenylphosphine; TPPO, Triphenylphosphine oxide; VAE, Vanillic acid equivalent; w/w, weight by weight; xg, Times gravity; YI, Yellowness index; µg, Microgram; µL, Microliter; µM, Micromolar; µm, Micrometer.

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that are released into the mayonnaise (Mirzanajafi-Zanjani et al., 2019). The presence of metal ions can induce lipid oxidation, reducing the shelf life and oxidative stability of mayonnaise (Félix et al., 2020; Ghorbani Gorji et al., 2016), especially for healthy mayonnaises made with polyunsaturated fatty acids (PUFAs) (Lunn & Theobald, 2006) which are more prone to oxidation (DeLany et al., 2000). Various methods have been employed to prevent lipid oxidation in fats, including the application of synthetic antioxidant compounds such as butylated hydroxytoluene (BHT), butylated hydroxyanisole (BHA), tert-butylhydroquinone (TBHQ), and ethylenediaminetetraacetic acid (EDTA) (Ghorbani Gorji et al., 2016). These are widely used in the food industry due to their low cost and efficiency (Sanhueza Catalán et al., 2000). However, there is increasing focus and interest on natural antioxidants to reduce the use of synthetic compounds, driven by consumer demand for safer, additive-free, and cleaner products. Therefore, antioxidants such as phenolic acids and tocopherols are being emphasized as alternatives (Flammini et al., 2020; Piva et al., 2018; Romeo et al., 2021).

Rice (*Oryza sativa* L.) bran (RB) is a by-product of the rice milling process, which constitutes approximately 12–23 % of the rice grain (Wongwaiwech et al., 2019). RB contains several bioactive compounds, such as tocopherols and γ -oryzanol (Dunford & King, 2000; Itoh et al., 1973), as well as phenolic acids like ferulic acid and p -coumaric acid (Santos et al., 2021). Pigmented rice varieties contain higher levels of these bioactive compounds compared to non-pigmented rice (Siripattanakulkajorn et al., 2024). These compounds offer numerous health benefits to consumers, including antioxidant, anti-inflammatory, anticancer, and antidiabetic properties (Ahsan et al., 2014; Ghazani & Marangoni, 2016; Minatel et al., 2016; Sugano & Tsuji, 1997). However, extracting phenolic acids from RB is challenging because these compounds are largely bound to the cell wall (Ti et al., 2014). Traditional extraction methods often involve processes such as alkaline hydrolysis and acid hydrolysis (Irakli et al., 2018), which are not ideal for food applications. Therefore, green solvent extraction of phenolic acid compounds in RB has been developed using natural deep eutectic solvents (NaDES). These solvents have shown potential for extracting phenolic acids (Alañón et al., 2018; Sombutsuwan et al., 2024; Wei et al., 2015), flavonoids (Oomen et al., 2020), anthocyanins (Zannou & Koca, 2022), polyphenols (Rebocho et al., 2022), lignin (Kumar et al., 2016; Lyu et al., 2018), triterpene saponins (Petrochenko et al., 2023), phlorotannins (Obluchinskaya et al., 2023), isoflavones (Semenov et al., 2023), curcumins (Huber et al., 2021) and other bioactive compounds (Osamede Airouyuwa et al., 2022; Zullaikah et al., 2019) from various sources such as plant metabolites, bioactive compounds in oil, and lignocellulosic biomass. This process does not involve any toxic chemicals, making it safer and more environmentally friendly (Santos et al., 2021).

NaDES are formed by combining two natural source compounds, such as betaine, choline chloride, acids, sugars, polyols, and amides (Smith et al., 2014; Zhang et al., 2012), through hydrogen bonds between a hydrogen bond acceptor (HBA) and a hydrogen bond donor (HBD) (Koh et al., 2023). Those mixtures could render NaDES biodegradable and bio-renewable, offering benefits such as low cost, low toxicity, and minimal energy requirements for production (Alañón et al., 2018; Tomé et al., 2018). NaDES have shown promise in extracting beneficial compounds from plants (Durand et al., 2021). However, they are less effective at coextracting certain toxic trace metals compared to organic solvents (Shikov, Obluchinskaya, et al., 2022; Xie et al., 2017). This characteristic makes NaDES a safer option for use in food, cosmetic and pharmaceutical (Shikov, 2022; Shikov et al., 2022). Nowadays, NaDES are increasingly being developed and improved for use in the food industry for applications such as food preservation, food stability (antimicrobial properties, stabilizing antioxidant compound, cryoprotective agents, biopolymer production, and green antifreeze agents), and the protection of food components (enzymes, colorants, aromas, and bioactive compound) (Basar et al., 2020; Li et al., 2023; Mišan & Pojić,

2021; Obluchinskaya et al., 2023). Despite their many advantages, NaDES mixtures hold significant challenges in food technology, including high viscosity, formulation difficulties, and potential additional costs when added in food recipes. This study aims to demonstrate a novel approach for incorporating NaDES into real food products.

The benefits of fortifying beverages such as chocolate milk and strawberry yogurt-based smoothies with phenolic compounds extracted from cocoa or sunflower pomace using NaDES extracts have recently been documented (Manuela et al., 2020; Şen et al., 2024). Here, NaDES were integrated into an oil-based food product to preserve the NaDES network, which can be disrupted by high dilution in a water environment. It is hypothesized that NaDES with an intact tridimensional structure can alter the microstructure of food matrices, particularly affecting the aqueous phase activity. In mayonnaise, this could be significant as NaDES might change the activity of aqueous phase molecules (e.g. transition metals) or create barriers that prevent contact between lipids and metal pro-oxidants, thereby reducing oxidation rates. To achieve this while avoiding additional costs, the strategy involved using mayonnaise ingredients to create NaDES before incorporating it into the mayonnaise formulation.

Food-grade betaine was used as the base, combined with sucrose or citric acid - compounds commonly used in mayonnaise recipes and known as potential candidates for NaDES formation. Additionally, to enhance the NaDES's potential for stabilizing mayonnaises against oxidative degradation, a preliminary extraction step was performed with NaDES to enrich the liquid with bioactive and antioxidant molecules. Black (B) and red (R) pigmented RB were used to obtain RB-enriched NaDES with antioxidant properties, specifically from phenolic acids. After finely tuning the recipe to produce mayonnaises with incorporated NaDES, we evaluated the viscosity, pH, color, droplet size (DLS), and water activity (a_w). Subsequently, we assessed the oxidative stability kinetics of mayonnaises by measuring lipid hydroperoxides levels, volatile compounds, and tocopherols loss over 69 days.

2. Material and method

2.1. Chemicals and standards

The chemical for NaDES preparation (betaine (≥ 98 %), citric acid (≥ 99 %), and sucrose (≥ 99.5 %)), phenolic acids for HPLC analysis (vanillic acid (≥ 97 %)), volatile compound standards for secondary lipid oxidation (octanal (≥ 99 %), nonanal (≥ 98 %), E-2-heptenal (≥ 95 %), hexanal (≥ 98 %), pentanal (≥ 97 %), E-2-penten-1-ol (≥ 95 %), E-2-pentenal (≥ 95 %), 1-penten-3-one (≥ 97 %), E-E-2,4-heptadienal (≥ 90 %), 2-ethylfuran (≥ 99 %)), mixed tocopherols (α -tocopherol, β -tocopherol, γ -tocopherol and δ -tocopherol), triphenylphosphine (TPP) (≥ 99 %), triphenylphosphine oxide (TPPO) (≥ 98 %), butan-2-one (≥ 99 %), 3-heptanol (≥ 99 %) and some of mayonnaise ingredients (xanthan gum, potassium sorbate (≥ 99 %), ethylenediaminetetraacetic acid (EDTA) (≥ 98.5 %) and sodium chloride (≥ 99 %)) were purchased from Sigma-Aldrich (St. Louis, MO, USA). Water, methanol (≥ 99.9 %), acetic acid (≥ 99 %), hexane (≥ 95 %) and 1,4-dioxane (≥ 99.5 %) (all HPLC grade) were purchased from Honeywell (Riedel-de-Haën, Seelze, Germany). The main ingredients for mayonnaise such as the rapeseed oil (*Brassica napus*), vinegar (8 %) and egg were purchased from a convenience store (Montpellier, France).

2.2. Rice bran samples and preparation

Two Thai rice (*Oryza sativa*) varieties were generous gifts from the Rice Research Center and registered by the Thailand Rice Department. The two varieties Niaw Dum Mor (black pigmented rice) and Hawm Gra Dang Nghah (red pigmented rice) were cultivated in 2019 during growth season (June–December). The rice were de-husked and de-branned by method described by Siripattanakulkajorn et al., 2024. Afterward, rice bran were screened for particle sizes between 106 and 425 μm , and

stored at 4 °C for further analysis.

2.3. NaDES preparation

The NaDES were prepared using the method described by Santos et al. (2021). Betaine was chosen as hydrogen bond acceptor (HBA) in combination with sucrose or citric as hydrogen bond donor (HBD). Betaine is an important nutrient for human health, naturally present in foods such as cereals. It can be biosourced from by-products of the sugar industry, particularly sugar beet, and is considered a novel food (Arumugam et al., 2021). Citric acid and sucrose, commonly found in mayonnaise recipes, also serve as hydrogen bond donor (HBD) candidates for forming the NaDES liquid phase. The ratios for NaDES, combined with water content, are as follows: NaDES1 (Betaine: Citric acid: water in molar ratios of 1.5:1:5) and NaDES2 (Betaine: Sucrose: water in molar ratios of 2:1:8.5). HBD and hydrogen bond acceptors (HBA) were first premixed in the corresponding molar ratio using a flat-bottom flask and mixed in a vacuum rotary evaporator (55 °C, 200 rpm, for 10 min). Next, water was incrementally and intermittently added, about 1/10 of its targeted content every 5 min. For this study, 20 % (w/w) of water content was used to reduce viscosity. The NaDES were stored in the dark at approximately 25 °C for further use.

2.4. Extraction and determination of phenolic acids and anthocyanins from Rice Bran

Total phenolic acids and anthocyanins content were extracted by using adapted method from Santos et al. (2021). RB (300 mg) was extracted with NaDES (15 mL) in a closed amber glass flask with orbital agitation at 250 rpm, 60 °C for 24 h (Cimarec Thermo Scientific Poly 15, Legallais, Montferrier-sur-Lez, France). Afterward, the samples were cooled to room temperature and stored at -20 °C for further analysis. Phenolic acid was analyzed by using supernatant extract from NaDES. Briefly, supernatant (100 µL) was dissolved in 2 mL methanol/water (1:3, v:v) and filtered through a 0.45 µm cellulose filter (Minisart Legallais, Montferrier-sur-Lez, France). Dissolved phenolic acids were analyzed by HPLC-PDA equipped with a Kinetex 5 µm C-18 column (100 Å, 4.6 mm × 250 m, Phenomenex, Le Pecq, France), connected with Dionex Ultimate 3000 pump and autosampler (USA) with chameleon software (USA) using the photodiode-array detector (PDA) SPD-M20A model (Shimadzu, Japan). The mobile phases were: (A) water (0.1 % acetic acid (v/v)) and (B) methanol (0.1 % acetic acid (v/v)). The elution gradient program was: (time (min): solvent B (%)): (0:0), (8:0), (35:100), (42:100), (45:0), (50:0). Flow rate, injection volume and detection wavelength were set at 1 mL/min, 10 µL and 280 nm, respectively. Results were reported as vanillic acid equivalents (VAE). Total anthocyanins was analyzed using method described by Teng et al. (2020) and Lee et al. (2019). Total anthocyanin was reported as cyanidin-3-O-glucoside equivalent (eq. cyd-3-glu) using molecular weight of cyanidin-3-O-glucoside 449.2 g/mol and molar extinction (ε) at 26,900 L/(mol × cm), after measuring absorbance at 520 nm by using microplate assay Infinite M200 microplate reader or SPARK (Tecan, Gröedig, Austria) equipped with Magellan software.

2.5. Mayonnaises preparation

Rapeseed oil (*Brassica napus*) was used for mayonnaise. The composition of the mayonnaise recipe includes: oil (75–77 % w/w), egg yolk (17–18 % w/w), vinegar (2 % w/w), sucrose (2 % w/w), salt (0.3 % w/w), xanthan gum (0.2 % w/w), potassium sorbate (0.1 % w/w), and citric acid (0.3 % w/w for citric acid-included mayonnaises and NaDES1-fortified mayonnaises). The additional compound presents in mayonnaise fortified with NaDES is betaine, at 0.3 % w/w for NaDES1 or 1.9 % w/w for NaDES2. Table 1 presents the 9 different mayonnaise recipes used for the experimental conditions, including a standard control mayonnaise (M), control mayonnaise with 100 ppm of EDTA

Table 1

List of ingredients and content (%w/w) of the different mayonnaises.

Ingredient (%w/w)	M and EDTA-M	CA-M	Mayonnaise fortified with NaDES1 (NaDES1-M, R + NaDES1-M and B+ NaDES1-M)	Mayonnaise fortified with NaDES2 (NaDES2-M, R + NaDES2-M and B+ NaDES2-M)
Rapeseed oil	77.1	76.8	76.5	74.8
Egg yolk	18.3	18.2	18.1	17.7
Vinegar 8 % acid	1.7	1.7	1.7	1.7
Sucrose	2.3	2.3	2.3	2.2
Salt	0.3	0.3	0.3	0.3
Xanthan gum	0.2	0.2	0.2	0.2
Potassium sorbate	0.1	0.1	0.1	0.1
Citric acid	–	0.3	0.3	–
Betaine	–	–	0.3	1.9
Water from NaDES	–	–	0.1	1.0

(EDTA-M), control mayonnaise with citric acid (CA-M), mayonnaise fortified with NaDES1 (NaDES1-M), mayonnaise fortified with NaDES1 enriched with Hawm Gra Dang Nghah rice bran (R + NaDES1-M), mayonnaise fortified with NaDES1 enriched with Niaw Dum Mor rice bran (B + NaDES1-M), mayonnaise fortified with NaDES2 (NaDES2-M), mayonnaise fortified with NaDES2 enriched with Hawm Gra Dang Nghah rice bran (R + NaDES2-M), and mayonnaise fortified with NaDES2 extracted with Niaw Dum Mor rice bran (B + NaDES2-M). Mayonnaise was prepared by mixing egg yolk, sugar, salt, and potassium sorbate using a rotor stator homogenizer (IKA-Werke GmbH & Co. KG, Staufen, Germany) equipped with a whisk, rotated at 180 rpm. After all ingredients were mixed, rapeseed oil was slowly added continuously into the mixture. Xanthan gum and citric acid were added once the emulsion had begun to form. In the end, NaDES was incorporated after the mayonnaise had been formed. NaDES1 and NaDES2 were added to mayonnaises in the appropriate volumes to match the original concentrations of citric acid and sucrose, respectively, found in the control mayonnaises (CA-M and M). The rice bran-enriched NaDES was added in a similar manner. Then, 4 g. of mayonnaise were weighted into 15 mL centrifuge tubes and stored in the dark at 20 °C to perform the oxidative stability test with measurements taken weekly until day 69.

2.6. Physical properties of mayonnaises

Droplet size (DLS), polydispersity index (PDI), viscosity, color, pH and water activity (a_w) of the mayonnaise were analyzed at day 0, following the method described by Miguel et al. (2019). Droplet size of mayonnaises was analyzed using Zetasizer pro (Malvern Panalytical, France) equipped with ZS Xplorer program (Malvern Panalytical, France). Mayonnaise (10 mg) were diluted in water (10 mL) and analyzed by using semi-micro-Polystyrene (PS) Cuvette (BRAND-TECH®Scientific, Germany). Measurement were carried out in triplicate and results were given in term of Z-average (µm) and polydispersity index (PI).

Viscosity of mayonnaises was measured at 25 °C using a Rheonaut module for HAAKE™ MARS™ rheometer model 60 (Thermo Fisher scientific, Illkirch-Graffenstaden, France) with RheoWin Job Manager software to run samples and RheoWin Data Manager software to edit results. Smooth and regulated base (temperature regulator) and upper 35 mm serrated rotor plate (P35/Ti/SE), with a gap of 2 mm, were used. An increasing gradient of stress was applied from 0.5 Pa to 450 Pa at 25 °C. Measurements were carried out in one replicate. Chroma meter CR-400 (Konica Minolta, sensing inc., Japan) was used for color measurement in triplicate. The data was recorded by using CIEL*a*b* color

system space, where L^* refers to lightness, a^* to red/green, and b^* to yellow/blue. Yellowness index (YI) was calculated from these values according to the equation: $YI = 142.86 b^*/L^*$ (Miguel et al., 2019). Water activity (a_w) of mayonnaise was measured in triplicate using AquaLab Pre Water Activity Measuring (Decagon device Inc., USA).

2.7. Oxidative stability of mayonnaises

2.7.1. Lipid extraction

2 mL of distilled water were added to the mayonnaise (4 g) into the centrifuge tube, vortexed and put in freezer at $-4\text{ }^\circ\text{C}$ for 12 h to undergo freeze-thawing, separating lipid and water phases. Afterwards the sample was centrifuged using an Avanti J-E series centrifuge equipped with a JLA-16.250 rotor (Beckman coulter, Villepinte, France) at 12,000g for 15 min. The lipid phase was collected and used for further analysis.

2.7.2. Measurement of lipid hydroperoxides

The hydroperoxide content was analyzed by using method described by (Wind et al., 2024). Briefly, 40 mg of the lipid phase was weighted in hemolysis tube. Freshly prepared triphenylphosphine (TPP) solution at 30 mg/mL in butan-2-one was made in a 25 mL glass bottle. 200 μL of the TPP solution was added in the hemolysis tube containing lipids and vortexed for 30 s. Afterwards, the measurement was performed by using FTIR model tensor 27 (Bruker, France) equipped with a GladiATR™ Single Reflection Diamond ATR unit, a DLATGS detector, and a MIR source at 26 $^\circ\text{C}$. The triphenylphosphine oxide (TPPO) area peak at 542 cm^{-1} was integrated, taking the frequencies 535 cm^{-1} and 560 cm^{-1} as bounds, and used to calculate the peroxide value. Analysis was performed in duplicate and reported as lipid hydroperoxide content in oil (mmol LOOH/Kg of oil).

2.7.3. Tocopherols loss measurement

The lipid extract (10 mg) was weighted in 2 mL vials and dissolved in hexane (1 mL). The HPLC-FLD system comprised a UHPLC focused Dionex Ultimate 3000 pump and autosampler (Thermo Scientific, USA), along with a FLD detector model FL3000 (Thermo Separation products, USA) set at 290–330 nm. Separation was carried out on a Silica column (Kinetex 5 μm , 250 mm length [L] x 4 mm internal diameter [ID], Phenomenex, Le Pecq, France) with a mobile phase consisting of Hexane/1,4-dioxane (97/3). Injection was set at 100 μL with flow rate 1.3 mL/min. Analysis was performed in duplicate, and quantifications were carried out for the following compounds: α -tocopherol, β -tocopherol, γ -tocopherol, and δ -tocopherol. The results were calculated into total tocopherols and expressed in g/kg of oil.

2.7.4. Measurement of secondary lipid oxidation compounds

Lipids sample (500 μL) were added into 10 mL headspace vials. Secondary lipid oxidation compounds were measured using dynamic headspace (DHS) combined with GC-MS (Gas Chromatography-Mass Spectrometry). The DHS was employed under the following conditions: incubation at 60 $^\circ\text{C}$ for 5 min; trapping for 12 min (using Tenax TA type trap at 40 $^\circ\text{C}$ with 300 mL of nitrogen at a flow rate of 25 mL/min, under agitation at 500 rpm); drying phase of 200 mL of nitrogen at 50 $^\circ\text{C}$ at a flow rate of 100 mL/min. For the Thermal Desorption Unit (TDU), a non-fractionated injection at 30 $^\circ\text{C}$ followed by a ramp to 300 $^\circ\text{C}$ at 120 $^\circ\text{C}/\text{min}$ was performed for 7 min. This step aims to concentrate the molecules. In the Cold Injection System (CIS) at 2 $^\circ\text{C}$, a ramp to 300 $^\circ\text{C}$ at 12 $^\circ\text{C}/\text{s}$ followed by a plateau for 5 min was conducted. The DHS, TDU, and CIS systems are from GERSTEL GmbH & Co. KG (Mülheim an der Ruhr, Germany). The gas chromatograph used is the 7890B/MSD 5977 (Agilent Technologies, Palo Alto, CA, USA) with a Gerstel robot and a polar capillary column DB-WaxUI of 60 m length, 0.25 mm inner diameter, 0.25 μm film thickness (J&W Scientific, Folsom, CA, USA) with hydrogen as the carrier gas at a flow rate of 2.2 mL/min. Elution was performed with the following temperature program: isothermal at

40 $^\circ\text{C}$ for 5 min, then 3 $^\circ\text{C}/\text{min}$ from 40 $^\circ\text{C}$ to 150 $^\circ\text{C}$, followed by 15 $^\circ\text{C}/\text{min}$ to 280 $^\circ\text{C}$. The mass spectrum was recorded in EI+ mode at 70 eV between 40 and 350 Da. The temperatures of the analyzer and the source were 150 $^\circ\text{C}$ and 250 $^\circ\text{C}$, respectively. The quantification was carried out for the following volatile compounds: octanal, nonanal, E-2-heptenal, hexanal, pentanal, E-2-penten-1-ol, E-2-pentenal, 1-penten-3-one, E-E-2,4-heptadienal, 2-ethylfuran in mayonnaise samples, using 3-heptanol as internal standard. The calibration curves were prepared from a stock standard solution containing these volatile compounds. The results obtained were processed with Masshunter software version 11.1 (Agilent Technologies, Palo Alto, CA, USA) using a quantitative method based on quantifier and qualifier ions specific to each standard. Peak identification was carried out by comparing the mass spectra with those in the NIST 2020 database (National Institute of Standard Technology). Analysis was performed in duplicate, and the results were expressed in ng/g of mayonnaise.

2.8. Statistical analysis

The means with standard deviations were reported. The statistic calculation was performed by using IBM SPSS Statistics for Windows, Version 20.0 (BM Corp, Armonk, NY: IBM Corp, NY, USA). Significant difference was determined at $P < 0.05$ using Duncan's multiple range test (DMRT).

3. Results and discussion

3.1. Extraction of phenolic compounds from rice bran (RB) with NaDES

Table 2. presents the result of the total phenolic content and total anthocyanin content, after extraction of two pigmented rice bran (RB) samples with NaDES1 (Betaine: Citric acid: water (1.5:1:5)) and NaDES2 (Betaine: Sucrose: water (2:1:8.5)). Whatever the used NaDES, Niaw Dum Mor (black-pigmented rice) exhibited higher phenolic acid and anthocyanin contents than the ones of Hawm Gra Dang Ngah (red-pigmented rice). Specifically, Niaw Dum Mor showed total phenolic acid content ranging from 21.3 to 22.3 mg VAE/g of RB and total anthocyanin content ranging from 15.1 to 17.6 mg eq. cyd-3-glu/g of RB. In contrast, Hawm Gra Dang Ngah demonstrated lower total phenolic acid content ranging from 4.59 to 7.25 mg VAE/g of RB and lower total anthocyanin content, ranging from 2.11 to 3.67 mg eq. cyd-3-glu/g of RB. Those results indicated that black-pigmented rice contains higher phenolic acids and anthocyanins content compared to red-pigmented rice, which is consistent with previous studies by Ghasemzadeh et al. (2018), Peanparkdee et al. (2020), Laokuldilok et al. (2011) and Zannou and Koca (2022). In addition, NaDES showed no significant difference in their ability to extract phenolic acids, while NaDES2 was more efficient than NaDES1 for the extraction of anthocyanins.

Table 2
Phenolic acids and anthocyanins extracted by NaDES.

Rice bran	NaDES composition	Total phenolic acid (mg VAE/ g of RB) ^{1,3}	Total anthocyanin (mg of cyd-3-glu equivalent/g of RB) ^{2,3}
Niaw Dum	NaDES1	22.3 ± 2.0 ^a	15.1 ± 0.1 ^b
Mor	NaDES2	21.3 ± 2.3 ^a	17.6 ± 0.7 ^a
Hawm Gra	NaDES1	7.25 ± 1.41 ^b	3.67 ± 0.51 ^c
Dang Ngah	NaDES2	4.59 ± 1.19 ^c	2.11 ± 0.11 ^d

¹ Values are means of independent quadruplicate determinations ± standard deviation.

² Values are means of independent triplicate determinations ± standard deviation.

³ Values in the same column followed by different letters indicate significant differences ($P < 0.05$).

3.2. Physical properties of the prepared mayonnaises

The droplet size and polydispersity index (PI) were measured on the first day after preparation (Table 3). Although the PDI values remained high, mayonnaises with or without incorporated NaDES did not show significant differences in droplet sizes. However, the fortification of mayonnaise with NaDES2 (NADES2-M, R + NADES2-M, and B + NADES2-M) led to a decrease in water activity (a_w), possibly due to the greater amount of NaDES2 used. In contrast, fortification with NaDES1 (NADES1-M, R + NADES1-M, and B + NADES1-M) did not affect a_w . One may assume that the hydrogen-bond matrix of NaDES properties could trapped water molecules within the mayonnaise, thereby reducing the a_w . Several studies on the physical and thermodynamic properties of NaDES-water mixtures have revealed an increase in hydrogen bonds and the establishment of stronger interactions compared to the raw NaDES, characterized by a diminution in thermodynamic activities (Durand et al., 2013). As depicted in Fig. 1, the color of the mayonnaises was significantly influenced by the pigmented substances from RB loaded into the NaDES. Notably, B + NADES1-M, R + NADES2-M, and B + NADES2-M demonstrated a significant effect on the color change ($P < 0.05$). The presence of anthocyanins extracted in NaDES contributes to a decrease in the L^* value, indicating reduced lightness in the mayonnaise, while increasing the a^* and decreasing the b^* values, leading to an increase in purple, blue, and red colors (Chen et al., 2023; Ghasemzadeh et al., 2018). However, the yellowness index (YI) remains consistent across all mayonnaise samples, suggesting that the increased presence of purple and red colors from anthocyanin pigments does not affect this index. Concerning the pH values (Table 3), the mayonnaises containing citric acid (CA-M, NaDES1-M, R + NaDES1-M and B-NaDES1-M) were more acidic (average pH at 3.83 ± 0.10) than the ones of mayonnaises without citric acid (M, EDTA-M, NADES2-M, R + NaDES2-M and B + NaDES2-M, average pH at 5.02 ± 0.07). The addition of 0.3 % w/w citric acid reduced the pH of mayonnaises by approximately 1.0. Finally, the viscosity of all nine mayonnaises was assessed and compared (Fig. 2). CA-M, NADES1-M, R + NADES1-M, and B + NADES1-M exhibited higher viscosities, suggesting that the presence of citric acid in the mayonnaises contributes to increase the viscosity. Thus, citric acid can act as a thickening agent similar to xanthan gum within the mayonnaises (Panovska et al., 2012; Raurich et al., 2020), thereby increasing the overall viscosity while decreasing pH values of citric acid-containing mayonnaises (Sikora et al. (2008)).

3.3. Oxidative stability

The oxidative stability of mayonnaises was investigated through the kinetic production of lipid hydroperoxides and volatile compounds from secondary oxidation, along with the loss of tocopherols, which are the naturally occurring antioxidants found in rapeseed oil (Farahmandfar et al., 2015; Karasakal & Şeren, 2011; Mirzaee Ghazani et al., 2014; Soler-Velasquez et al., 1998). Therefore, the decrease in tocopherols indicates the effectiveness in preventing lipid oxidation. The study assessed the effectiveness of NaDES-fortified mayonnaises and NaDES-fortified mayonnaises enriched with phenolic acids and anthocyanins from RB extract, aiming to enhance the shelf life of the mayonnaise. The total content in phenolic acids and anthocyanins equivalents after formulation of the enriched NaDES into the mayonnaises was described in Table 4. R + NaDES1-M and B + NaDES1-M exhibited lower total phenolic acid and anthocyanin content than R + NADES2-M and B + NADES2-M. These differences can be attributed to variations in the original content of citric acid and sucrose in the mayonnaise, which create limitations in the volume of NaDES that can be incorporated into the mayonnaise to align with the content in recipes. Similarly, mayonnaises fortified with NaDES enriched with the black-pigmented RB (Niaw Dum Mor) exhibited higher content in phenolic acids and anthocyanins than those enriched with the red-pigmented RB (Hawm Gra Dang Ngh).

The formation of lipid hydroperoxides from day 0 to day 69 in different mayonnaise groups was measured and represented in Fig. 3. Initially, all mayonnaises exhibited lipid hydroperoxides levels ranging between 1.57 and 3.12 mmol LOOH/Kg of oil, and remained stable from day 0 to day 17. However, from day 24, notable increases in lipid hydroperoxides were observed in mayonnaise M, CA-M, NADES1-M, R + NADES1-M, and B + NADES1-M. Remarkably, mayonnaise formulations containing citric acid demonstrated an accelerated lipid hydroperoxides production, reaching 15.2–19.1 mmol LOOH/Kg of oil by day 69, while the raw mayonnaise (M) exhibited a comparatively stable increase, reaching 7.49 ± 0.84 mmol LOOH/Kg of oil at the final measurement. Mayonnaise EDTA-M maintained a low lipid hydroperoxides within the range of 1.35–2.31 mmol LOOH/Kg of oil, confirming the strong antioxidant effect of EDTA. Interestingly, NaDES sucrose-containing mayonnaises (NADES2-M, R + NADES2-M, and B + NADES2-M) also exhibited similar oxidative stability, with lipid hydroperoxides levels stabilized between 1.92 and 3.73 mmol LOOH/Kg of oil.

Volatile compounds from secondary lipid oxidation were also

Table 3
Droplet size, polydispersity index, color, yellowness index and pH values of mayonnaises.

Mayonnaise	Droplet size (μm) ^{2,3}	Polydispersity Index (PDI) ^{2,3}	Water activity (a_w) ^{1,3}	Color ^{1,3}			Yellowness index (YI) ^{1,3}	pH ^{2,3}
				L^*	a^*	b^*		
M	$2.76 \pm 0.55^{\text{cde}}$	$1.00 \pm 0.00^{\text{a}}$	$0.977 \pm 0.001^{\text{ab}}$	$70.3 \pm 0.0_{\text{abc}}$	$-3.33 \pm 0.28^{\text{e}}$	$18.3 \pm 1.6^{\text{abc}}$	$37.2 \pm 3.2^{\text{bc}}$	$4.96 \pm 0.04^{\text{a}}$
EDTA-M	$5.99 \pm 0.74^{\text{a}}$	$0.627 \pm 0.084^{\text{b}}$	$0.970 \pm 0.004^{\text{b}}$	$70.8 \pm 1.0^{\text{a}}$	$-3.04 \pm 0.01^{\text{de}}$	$16.2 \pm 2.7^{\text{c}}$	$32.8 \pm 5.8^{\text{c}}$	$5.05 \pm 0.07^{\text{a}}$
CA-M	$3.43 \pm 0.25^{\text{bc}}$	$0.752 \pm 0.288^{\text{ab}}$	$0.981 \pm 0.002^{\text{a}}$	$69.8 \pm 0.2_{\text{bc}}$	$-2.64 \pm 0.37^{\text{d}}$	$16.5 \pm 0.0^{\text{bc}}$	$33.7 \pm 0.0^{\text{c}}$	$3.90 \pm 0.02^{\text{b}}$
NaDES1-M	$3.94 \pm 0.65^{\text{b}}$	$0.926 \pm 0.128^{\text{ab}}$	$0.970 \pm 0.000^{\text{b}}$	$69.3 \pm 0.3^{\text{c}}$	$-3.53 \pm 0.23^{\text{ef}}$	$19.1 \pm 0.6^{\text{abc}}$	$39.4 \pm 1.4^{\text{abc}}$	$3.90 \pm 0.02^{\text{b}}$
R + NaDES1-M	$1.91 \pm 0.22^{\text{e}}$	$0.688 \pm 0.248^{\text{b}}$	$0.958 \pm 0.001^{\text{c}}$	$70.2 \pm 0.4_{\text{abc}}$	$-3.28 \pm 0.02^{\text{e}}$	$20.0 \pm 1.0^{\text{bc}}$	$40.7 \pm 2.3^{\text{abc}}$	$3.83 \pm 0.11^{\text{bc}}$
B + NaDES1-M	$2.00 \pm 0.04^{\text{e}}$	$1.00 \pm 0.00^{\text{a}}$	$0.967 \pm 0.002^{\text{bc}}$	$67.9 \pm 0.3^{\text{d}}$	$-1.93 \pm 0.10^{\text{c}}$	$20.2 \pm 1.2^{\text{ab}}$	$42.6 \pm 2.8^{\text{ab}}$	$3.72 \pm 0.12^{\text{c}}$
NaDES2-M	$3.04 \pm 0.73^{\text{bcd}}$	$0.828 \pm 0.097^{\text{ab}}$	$0.914 \pm 0.003^{\text{d}}$	$70.4 \pm 0.4_{\text{ab}}$	$-3.93 \pm 0.08^{\text{f}}$	$21.6 \pm 2.9^{\text{a}}$	$43.7 \pm 5.7^{\text{ab}}$	$5.03 \pm 0.04^{\text{a}}$
R + NaDES2-M	$2.29 \pm 0.70^{\text{de}}$	$0.874 \pm 0.200^{\text{ab}}$	$0.900 \pm 0.006^{\text{e}}$	$66.8 \pm 0.4^{\text{e}}$	$-0.08 \pm 0.48^{\text{b}}$	$21.5 \pm 1.0^{\text{a}}$	$45.9 \pm 2.3^{\text{a}}$	$5.09 \pm 0.01^{\text{a}}$
B + NaDES2-M	$2.14 \pm 0.26^{\text{de}}$	$0.710 \pm 0.172^{\text{ab}}$	$0.915 \pm 0.006^{\text{d}}$	$63.1 \pm 0.2^{\text{f}}$	$3.18 \pm 0.08^{\text{a}}$	$17.2 \pm 0.0^{\text{bc}}$	$38.8 \pm 0.1^{\text{abc}}$	$4.98 \pm 0.11^{\text{a}}$

¹ Values are means of independent duplicate determinations \pm standard deviation.

² Values are means of independent triplicate determinations \pm standard deviation.

³ Values in the same column followed by different letters indicate significant differences ($P < 0.05$).

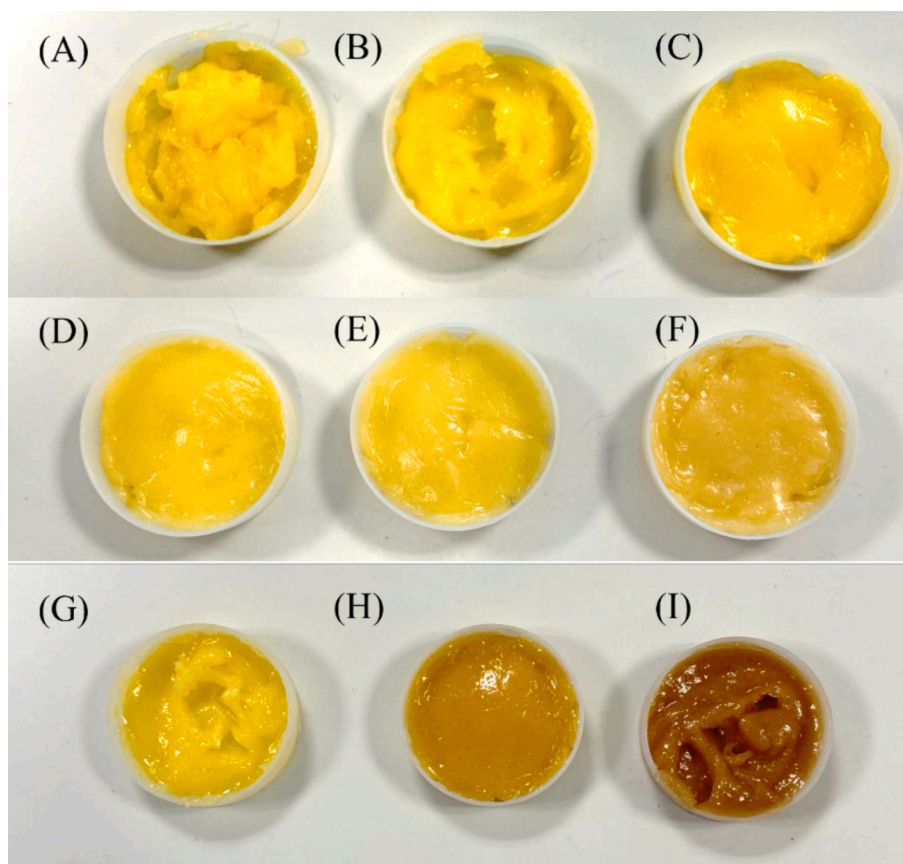


Fig. 1. (A) Control mayonnaise (M); (B) Mayonnaise with 100 ppm of EDTA (EDTA-M); (C) Control mayonnaise with citric acid (CA-M); (D) Mayonnaise fortified with NaDES1 (NADES1-M); (E) Mayonnaise fortified with NaDES1 enriched with Hawm Gra Dang Nghah rice bran (R + NaDES1-M); (F) Mayonnaise fortified with NaDES1 enriched with Niaw Dum Mor rice bran (B + NaDES1-M); (G) Mayonnaise fortified with NaDES2 (NADES2-M); (H) Mayonnaise fortified with NaDES2 enriched with Hawm Gra Dang Nghah rice bran (R + NADES2-M); (I) Mayonnaise fortified with NaDES2 enriched with Niaw Dum Mor rice bran (B + NADES2-M).

kinetically measured and represented in Fig. 4. The analysis revealed no detection (below limits of sensitivity) of E-2-penten-1-ol and 1-penten-3-one compounds throughout the entire duration of the study. Low amounts of volatile compounds, including 2-ethylfuran, pentanal, E-2-pentenal, octanal, 2-heptenal, and nonanal, were detected. These minor compounds have final concentrations ranging from 35.8 to 221.4 ng/g of mayonnaise. The major volatile compounds identified were 2,4-heptadienal and hexanal, which showed the highest concentrations, ranging from 513.1 to 634.8 ng/g of mayonnaise at the end of the storage duration. Mayonnaises CA-M, NADES1-M, R + NaDES1-M and B + NaDES1-M had the highest concentrations of all volatile compounds and the fastest production rate throughout the period of oxidation. Volatile compounds in the raw mayonnaise M showed a similar increase in concentration but at a lower rate. The other mayonnaises (EDTA-M, NADES2-M, R + NADES2-M, and B + NADES2-M) did not show any significant changes in volatile compound concentrations over time. The main volatile compounds identified (e.g. 2,4-heptadienal and hexanal) were present since the beginning of oxidation assay but did not increase during storage. These compounds are primarily derived from polyunsaturated fatty acids, such as linoleic and linolenic acids, which are abundant in the rapeseed oil (Farahmandfar et al., 2015; Mirzaee Ghazani et al., 2014). Indeed, studies on volatile compounds resulting from lipid oxidation identified these two fatty acids as major producers of hexanal, 2,4-heptadienol, 2-ethylfuran, pentanal, and E-2-pentenal (Ghorbani Gorji et al. (2019) and Fereidoon and Won Young (2020)). Additionally, oleic acid contributes to the production of nonanal and octanal. Their studies echo our results where polyunsaturated fatty acids such as linolenic and linoleic acids are key drivers of lipid oxidation in our mayonnaises, with lower levels of nonanal and octanal

(Farahmandfar et al., 2015; Mirzaee Ghazani et al., 2014).

Finally, the loss in tocopherols content from day 0 to day 59 was analyzed (Fig. 5). Initially, all mayonnaise samples had a total tocopherol content of approximately 1.24 ± 0.64 g/kg oil, with γ -tocopherol and α -tocopherol being predominant compounds. The tocopherols content was found to be inversely related to the lipid hydroperoxides formation in the oil, with a starting to decrease after day 24. The rate of tocopherol degradation was similar across the six mayonnaise samples (M, EDTA-M, CA-M, NADES1-M, R + NaDES1-M, and B + NaDES1-M), with final tocopherol levels average at 0.895 ± 0.024 g/kg oil. However, the mayonnaises fortified with NaDES2 exhibited a slower degradation rate, and retained a final concentration of tocopherols average at 1.02 ± 0.01 g/kg oil, which is significantly higher ($P < 0.05$) than other mayonnaise samples.

Overall, oxidative stability results were consistent with each other. Notably, the initial point of lipid oxidation occurred on the same day for all samples, specifically on day 24, when lipid hydroperoxides and volatiles compounds began to form. From day 0 to day 24, tocopherols concentration remained very stable, with no signs of lipid oxidation from lipid hydroperoxides or volatile compounds. This indicates that tocopherols act as primary antioxidants (Butsat & Siriamornpun, 2010; Havaux et al., 2005; Sheppard et al., 1993), preventing lipid oxidation. After this period, tocopherols began to oxidize and an increase in lipid hydroperoxides and volatile compounds was observed. A similar phenomenon was observed by Liu et al. (2020) and Balakrishnan and Schneider (2023), where the accelerated decomposition of tocopherols was also observed to occur faster than lipid oxidation. Nogala-Kalucka and Gogolewski (2000) studied the relationship between fatty acid composition, tocopherol content, and lipid hydroperoxides during

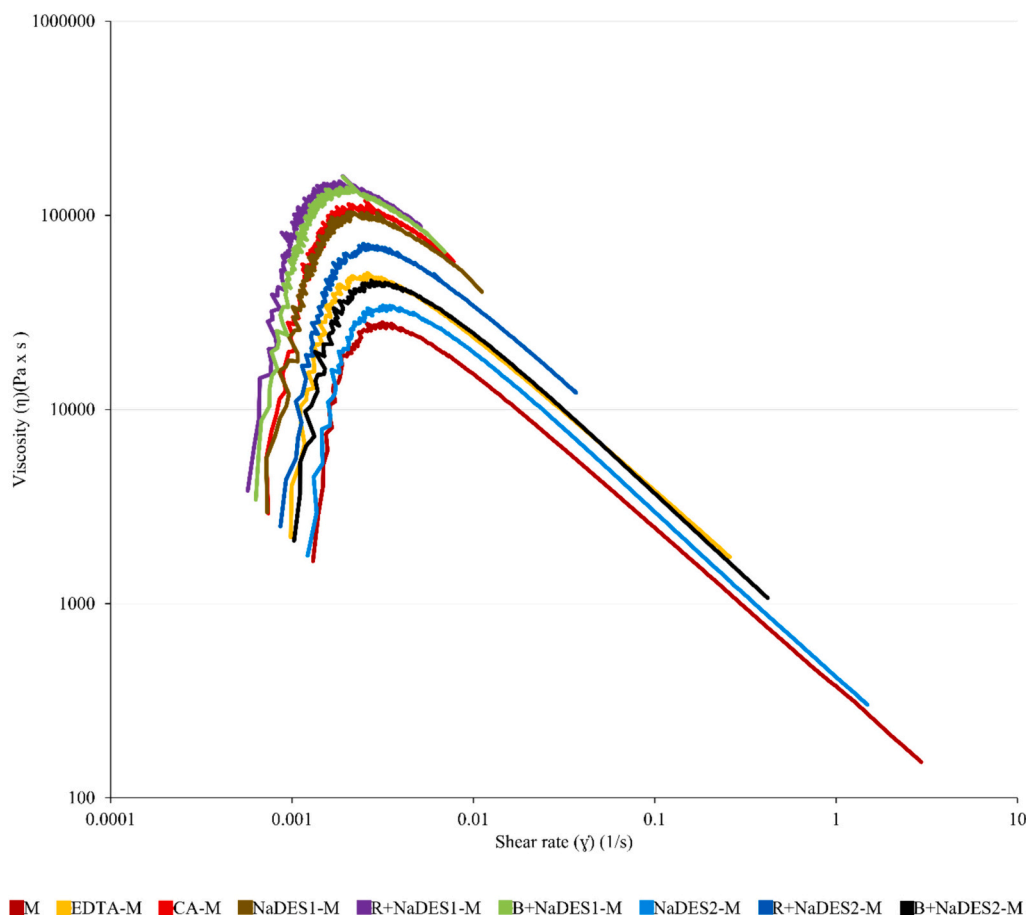


Fig. 2. Viscosity of the mayonnaise samples at day 0; temperature 25 °C. Samples were analyzed in only one replicate due to limited sample availability.

Table 4

Total phenolic acid and anthocyanin content in mayonnaises.

Mayonnaise	Total phenolic acid (mg VAE/g of mayonnaise) ^{1,2}	Total anthocyanin (mg of cyd-3-glu equivalent/g of mayonnaise) ^{1,2}
M	–	–
EDTA-M	–	–
CA-M	–	–
NaDES1-M	–	–
R + NaDES1-M	0.671 ± 0.130 ^d	0.340 ± 0.047 ^d
B + NaDES1-M	2.06 ± 0.18 ^c	1.40 ± 0.01 ^c
NaDES2-M	–	–
R + NaDES2-M	3.91 ± 1.02 ^b	1.80 ± 0.09 ^b
B + NaDES2-M	18.1 ± 2.0 ^a	15.0 ± 0.5 ^a

¹ Values are means of independent quadruplicate determinations ± standard deviation.

² Values in the same column followed by different letters indicate significant differences ($P < 0.05$).

margarine storage. Their findings also support our results, demonstrating that the lipid hydroperoxides in margarine starts to increase after the tocopherols degradation. The onset of secondary lipid oxidation coincided with the accelerated production of lipid hydroperoxides, indicating a consistent oxidation kinetic rate in every mayonnaise samples. Lipid hydroperoxides are known to be the major factor affecting the production rate of secondary lipid oxidation products. Therefore, lipid hydroperoxides are the initial compounds to oxidize, leading to the formation of volatile compounds as secondary lipid

oxidation products, which is particularly observable in mayonnaises containing citric acid (Grebenteuch et al., 2021; Wiczew et al., 2021).

Moreover, our results suggest that the pH contributes in increasing lipid oxidation of mayonnaises. This effect could be due to the disruption of the ion bridge between of iron and phosvitin, leading to increased release of Fe^{2+} or Fe^{3+} (Mirzanajafi-Zanjani et al., 2019), and subsequent acceleration of lipid oxidation kinetics (Félix et al., 2020; Ghorbani Gorji et al., 2016). Yang et al. (2023) reported that lower pH levels may also induce protein oxidation in low-density lipoprotein (LDL), which can release radical species and activate a chain reaction leading to lipid oxidation by interfering with the droplet interface, thereby accelerating the kinetics of lipid oxidation in conjunction with protein oxidation. The different lipid hydroperoxides production kinetics in the presence or in the absence of citric acid in mayonnaises underscores the pH dependency of lipid oxidation rates. Notably, mayonnaise M (pH 4.98 ± 0.04) without antioxidant additives exhibited a lower lipid hydroperoxides increase rate compared to CA-M (pH 3.90 ± 0.02) and all NaDES1-based mayonnaises (NADES1-M, R + NADES1-M, and B + NADES1-M) (pH 3.81 ± 0.11).

The incorporation of NaDES into the mayonnaise matrix showed different lipid oxidation pattern. Regardless of the conditions, mayonnaises fortified with NaDES1 did not provide any effect. One might speculate that the quantity of NaDES incorporated into the mayonnaises was possibly insufficient to impact the structured lipid dispersion or alter the properties of the continuous water phase, as supported by our results. Another explanation could be that the NaDES was diluted too extensively. Indeed, it is well established that the structure of NaDES disintegrates when water becomes dominant, resulting in the loss of its distinctive properties. This occurs because each component reverts to its individual properties, and the overall physicochemical behavior of the

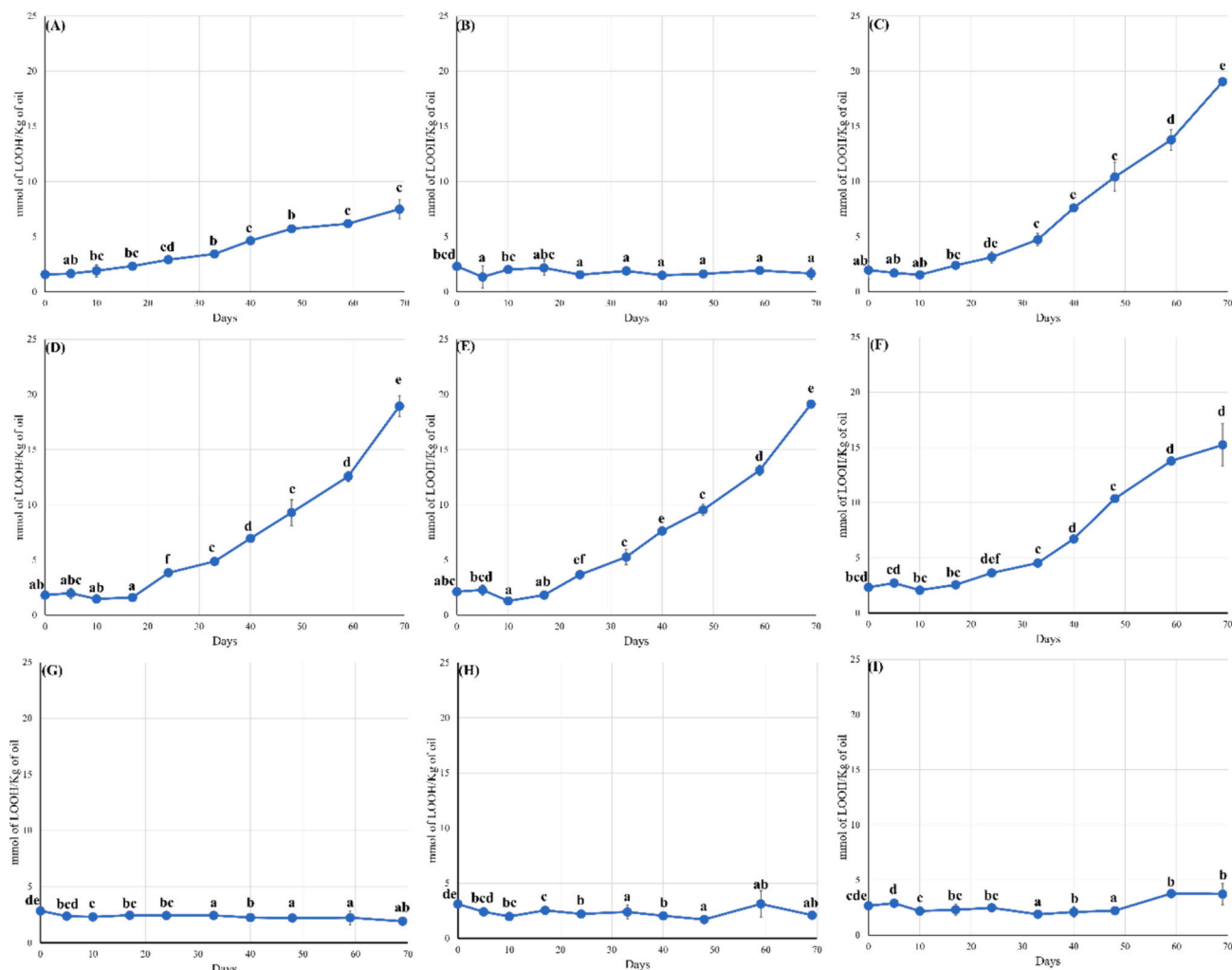


Fig. 3. Lipid hydroperoxides (LOOH) kinetic rate from day 0 to day 69 in each mayonnaise by (A) Control mayonnaise (M); (B) Mayonnaise with 100 ppm of EDTA (EDTA-M); (C) Control mayonnaise with citric acid (CA-M); (D) Mayonnaise fortified with NaDES1 (NADES1-M); (E) Mayonnaise fortified with NaDES1 enriched with Hawm Gra Dang Ngah rice bran (R + NaDES1-M); (F) Mayonnaise fortified with NaDES1 enriched with Niaw Dum Mor rice bran (B + NaDES1-M); (G) Mayonnaise fortified with NaDES2 (NADES2-M); (H) Mayonnaise fortified with NaDES2 enriched with Hawm Gra Dang Ngah rice bran (R + NADES2-M); (I) Mayonnaise fortified with NaDES2 enriched with Niaw Dum Mor rice bran (B + NADES2-M); Analyzed was performed in duplicate and different letter is represented significantly different ($P < 0.05$) by DMRT.

medium is then governed by water, the continuous phase. Studies using 2D NMR, FTIR, and molecular dynamics simulations have shown that the supramolecular complexes characteristic of NaDES breaks down when the dilution reaches approximately 50 % (v/v), which is far below our conditions (Choi et al., 2011; Gutiérrez et al., 2009; Hadj-Kali et al., 2015; Hammond et al., 2017). Therefore, all systems utilizing citric acid (CA-M, NADES1-M, R + NaDES1-M, and B + NaDES1-M) tend to behave similarly, a trend corroborated by all our results. In opposite, mayonnaises fortified with NaDES2, which allowed the highest content applied into the mayonnaise, demonstrated notable and positive influence. Indeed, mayonnaises (NaDES2-M, R + NaDES2-M, and B + NaDES2-M) were the most effective in preventing lipid oxidation, with comparable oxidative stability to mayonnaise containing EDTA (EDTA-M), but much stable than the control mayonnaise (M). This characteristic allowed to preserve tocopherols from oxidation, while preventing the formation of lipid hydroperoxides and the release of volatile compounds. The lower water thermodynamic activity (a_w) in NaDES2-based mayonnaises, may be a first explanation of the reduced oxidation rate, since it may affect the mobility and/or activity of metal ions, therefore preventing lipid oxidation. Vu et al. (2020) investigated the effect of a_w on lipid oxidation in food model (cracker) and highlighted the influence of low a_w to

limit oxidation. Similarly, Barden et al. (2015) showed that lower a_w reduced the mobility of metal ions and prevent lipid oxidation in low moisture foods. In addition, one may expect that the presence of NaDES affects the distribution of antioxidants (tocopherols, phenolics, etc.) and pro-oxidants (metals) molecules, along with the structure of proteins (e.g. LDL, apoproteins, and phosvitin) present in the mayonnaise (Durand et al., 2021). Indeed, as supported by Yang et al. (2023), proteins present at the oil/water droplet interface or as individual particles or aggregates in the continuous aqueous phase are significantly involved in oxidation routes.

Last but not least, although the intrinsic influence of phenolic acids and anthocyanins loaded into the NaDES could not be highlighted due to very low levels of oxidation, even after long-term storage of mayonnaise at 20 °C, the potential to pre-load NaDES with active molecules represents significant interest. Indeed, NaDES could be used to charge bioactive molecules, antioxidants, or other nutraceutical or health-promoting compounds, enhancing the benefits of mayonnaises. Moreover, since mayonnaises are effective foods for delivering omega-3 long-chain polyunsaturated fatty acids of nutritional interest, such as docosahexaenoic acid and eicosapentaenoic acid, it would be interesting to evaluate whether this high oxidative stability with NaDES fortification

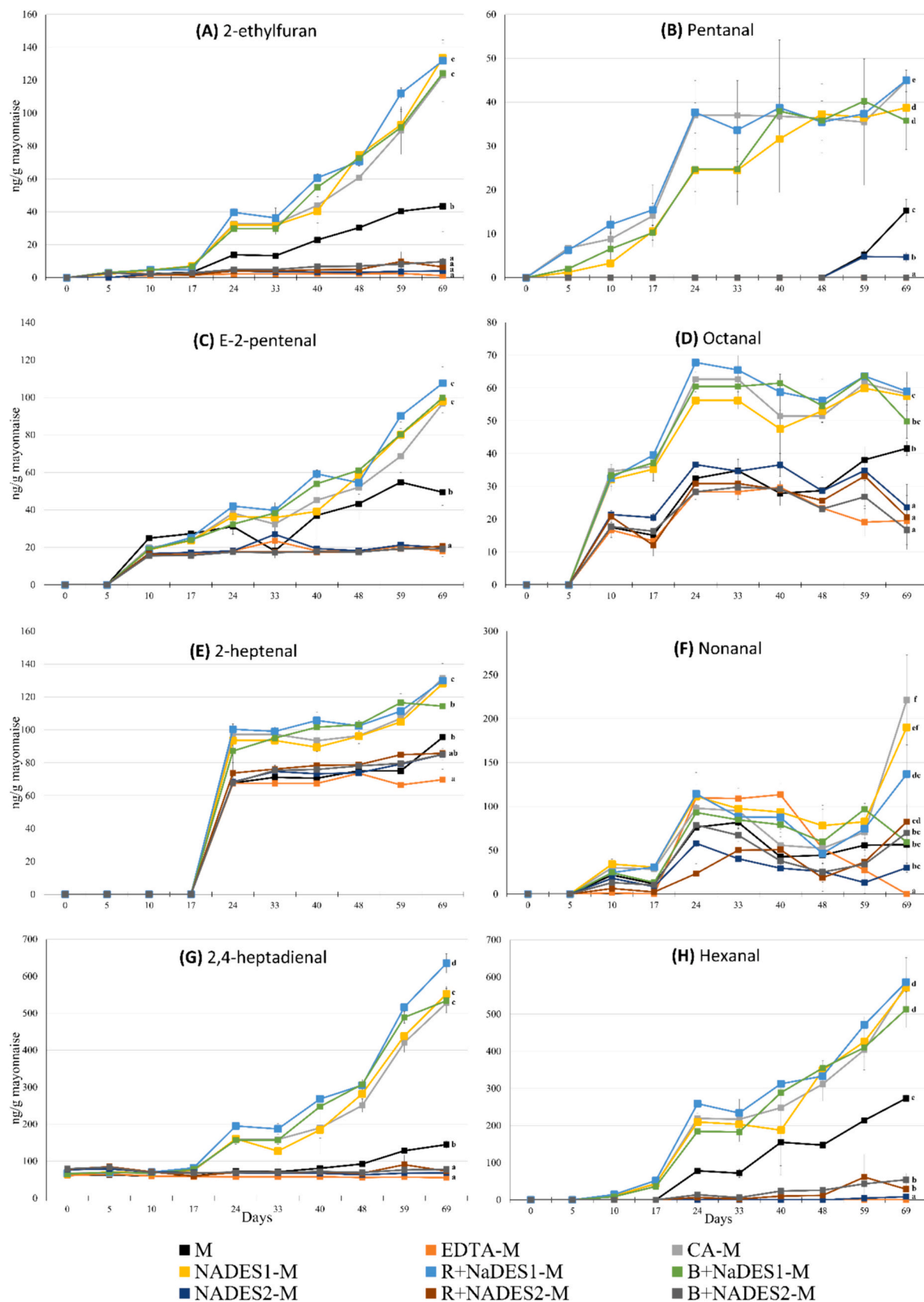


Fig. 4. Volatile compounds content (ng/g) in mayonnaise (A) 2-ethylfuran; (B) Pentanal; (C) E-2-pentenal; (D) Octanal; (E) 2-heptenal; (F) Nonanal; (G) 2,4-heptadienal; (H) Hexanal; Analyzed was performed in duplicate and different letter is represented significantly different ($P < 0.05$) by DMRT.

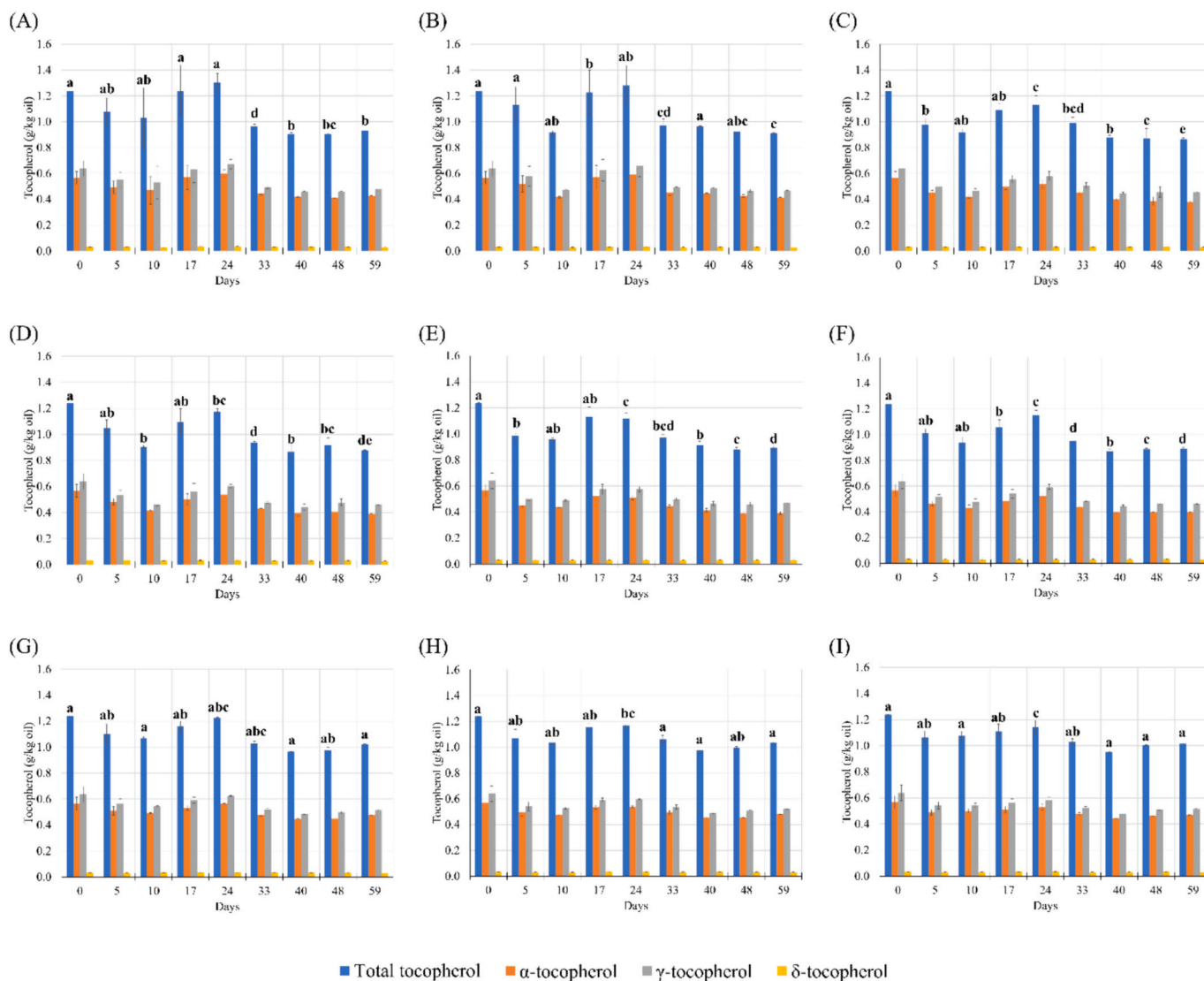


Fig. 5. The loss of tocopherols kinetic rate from day 0 to day 59 in each mayonnaise by (A) Control mayonnaise (M); (B) Mayonnaise with 100 ppm of EDTA (EDTA-M); (C) Control mayonnaise with citric acid (CA-M); (D) Mayonnaise fortified with NaDES1 (NADES1-M); (E) Mayonnaise fortified with NaDES1 enriched with Hawm Gra Dang Nghe rice bran (R + NaDES1-M); (F) Mayonnaise fortified with NaDES1 enriched with Niaw Dum Mor rice bran (B + NaDES1-M); (G) mayonnaise fortified with NaDES2 (NADES2-M); (H) Mayonnaise fortified with NaDES2 enriched with Hawm Gra Dang Nghe rice bran (R + NaDES2-M); (I) Mayonnaise fortified with NaDES2 enriched with Niaw Dum Mor rice bran (B + NaDES2-M); Analyzed was performed in triplicate and different letter is represented significantly different ($P < 0.05$) by DMRT.

could be maintained with more sensitive lipids. The combination of these factors demonstrates the potential and efficacy of NaDES in food models, increasing shelf life and providing an alternative to synthetic antioxidant compounds with additional benefits.

4. Conclusion

The incorporation of NaDES into the mayonnaise matrix was shown to affect its physical properties, especially water activity (a_w) and pH. The changes in physical properties seem to play an important role in reducing the rate of lipid oxidation. Especially, mayonnaises fortified with NaDES2 with a_w at 0.910 and pH at 5.03, effectively reduced the rate of lipid oxidation while maintaining the highest tocopherols content during storage. Selecting the appropriate NaDES formulation and dosage can effectively reduce water activity, while providing opportunity to integrate antioxidants and bioactive compounds extracted from various sources. This strategy not only preserve tocopherols, but also extends the shelf life of mayonnaises by reducing lipid oxidation products, while introducing health-promoting bioactive molecules. The findings of this

study enhance our understanding of NaDES and suggest innovative application in the food industry. Yet, further investigation is required to deepen our understanding and to explore more possibilities.

Ethics statement

For the research presented in this study, no ethics statement is required since neither procedures nor raw materials involved animals or animal-derived products.

CRediT authorship contribution statement

Chatchai Siripattanakulkajorn: Writing – original draft, Methodology, Investigation, Funding acquisition, Conceptualization. **Piraporn Sombutsuwan:** Writing – review & editing, Supervision, Investigation, Conceptualization. **Pierre Villeneuve:** Writing – review & editing, Supervision, Conceptualization. **Bruno Baréa:** Writing – review & editing, Investigation, Conceptualization. **Romain Domingo:** Writing – review & editing, Validation, Investigation. **Marc Lebrun:** Writing – review &

editing, Resources, Methodology. **Kornkanok Aryusuk:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **Erwann Durand:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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