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Development of a novel functional jelly with dieckol-rich extract from *Eisenia bicyclis*: Physicochemical, antioxidant, and sensory characterization

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ABSTRACT

This study aimed to develop a novel functional jelly by incorporating dieckol-rich extracts from *Eisenia bicyclis* (*EB*). In the extraction process, a high dieckol yield (16.5 mg/g biomass) was achieved by response surface optimization (optimum conditions: 55.3 % prethanol A, 70.9 °C, and 87.3 min). Dieckol jellies (DJs) were produced by adding various amounts of the extract with 25, 50, 75, and 100 % of 9.954 mg (recommended daily intake). The antioxidant activity of DJ increased from 0.02 to 0.4 mg ascorbic acid equivalent/mL with increasing dieckol content, and the texture analysis showed increased hardness, adhesiveness, and chewiness in DJs with over 75 % dieckol. The sensory testing indicated that DJ 25 had a superior overall preference, comparable to DJ 0 and higher than DJ 50 − 100. This study confirmed that *EB* is a high-potential source of dieckol, and the developed DJ is expected to have high potential as a novel functional food.

1. Introduction

Phlorotannins are useful bioactive substances that are specifically biosynthesized in brown algae. Among several phlorotannins, including dieckol, eckol, and phlorofucofuroeckol, dieckol has been widely studied as the major and most active compound (Lee & Jeon, 2015). Various studies have reported that dieckol exhibits multiple biological functions, such as antioxidant (Shin et al., 2023), anti-fungal (Lee et al., 2010), anti-aging (Jang et al., 2015), and anti-tumorigenic activities (Oh et al., 2011), which have been demonstrated through in vitro experiments. In addition, sleep-enhancing (Kim et al., 2022), hepatoprotective (Kang et al., 2012), and antiosteoporosis and bone protective effects (Wang et al., 2022) of dieckol have been demonstrated through in vivo experiments. The Ministry of Food and Drug Safety (MFDS) of the Republic of Korea has approved the use of dieckol-rich extracts from brown algae *Ecklonia cava* (*EC*) and *Ecklonia stolonifera* (*ES*) as functional ingredients with sleep-enhancing and hepatoprotective activities, respectively (Korea Ministry of Food and Drug Safety (MFDS), 2018). Although dieckol is attracting attention as an ingredient for functional foods (Yoon et al., 2024), limited types of feedstocks and food formulations and low extraction yields of dieckol hinder practical applications of dieckol-rich extracts (Rajan et al., 2021; Shin et al., 2023).

Previous studies have primarily utilized *EC* as a feedstock for dieckol recovery, followed by *ES* and *Eisenia bicyclis* (*EB*) (Banach et al., 2020). Accordingly, process optimization studies to improve dieckol extraction yield have only focused on *EC*. Park and Lee (2021) optimized ultrasound-assisted extraction of anti-glycation agents (with dieckol) from *EC*, and the results showed an extraction yield of about 1.62 % and a dieckol content of about 44.61 mg/g extract. Shin et al. (2023) optimized the dieckol extraction yield in the maceration process of *EC*, producing approximately 6.4 mg/g biomass under optimized conditions (62.6 % ethanol, 54.2 °C extraction temperature, and 13.2 min extraction time). The dieckol extraction yield from *EB* was about 6.0 mg/g biomass in a non-optimized maceration process (50 % ethanol, room temperature, and 24 h extraction time) (Lee, Heo, et al., 2023). To expand and apply the feedstocks for dieckol recovery, another biomass such as *EB* should be evaluated.

In Korea, various forms such as films, jellies, capsules, tablets, pills, liquids, syrups, and pastes are permitted as types of functional foods (Korea Ministry of Food and Drug Safety (MFDS), 2020). For functional

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foods containing dieckol, the formulation is limited to capsules (Um et al., 2018) or tablets (Lee & Jeon, 2015), thus the development and evaluation of other formulations is required to expand the related markets. Among various formulations, jelly offers a compact size, easy storage, portability, and convenient consumption, with a relatively simple manufacturing process (Ozcan et al., 2024; Tarahi et al., 2023). In the previous studies, various bioactive compounds were utilized as functional ingredients in jelly, and the developed jelly was characterized: antioxidants-rich rosemary extract (Cedeño-Pinos et al., 2020), fermented guava pulp enriched with vitamin B12 (Palachum et al., 2020), and probiotics isolated from fermented gooseberry and Indian scramberry (Kathiresan et al., 2021). In particular, seaweed or its extract (e.g., *Ulva prolifera* and *Gracilaria*) has been incorporated into jelly, highlighting the potential and feasibility of functional jelly development using algae (Kim et al., 2019; Manurung et al., 2018; Manzoor et al., 2024).

This study aimed to develop a novel functional jelly with dieckol-rich extract from edible brown macroalga *EB*. In the extraction process, the major extraction parameters were selected and optimized to prepare dieckol-rich extracts from *EB* with a high dieckol yield. In the food production process, the effect of dieckol content (i.e., extract loading) on the physicochemical, antioxidant, and sensory properties of dieckol jelly (DJ) was investigated to determine the optimum dieckol content for manufacturing superior functional food.

2. Materials and methods

2.1. Materials

For extracting dieckol, *Eisenia bicyclis* (collected in 2023) was sourced from Ulleungdo (Welcome Ulleung, Ulleung-gun, Republic of Korea). Prethanol A (95 %) and Trifluoroacetic acid (99 %) were purchased from Duksan Chemicals (Ansan, Republic of Korea), and Samchun Chemicals (Seoul, Republic of Korea), respectively. Acetonitrile (HPLC grade) was purchased from Daejung Chemicals & Metals (Siheung, Republic of Korea), and Dieckol (HPLC grade) as a standard material was purchased from Aktin Chemical, Inc. (Chengdu, P.R. China). For jelly preparation, the ingredients were purchased from online market, including konjac powder (Chansem Food, Incheon, Republic of Korea), carrageenan (MSC Co., Ltd., Yangsan, Republic of Korea), xanthan gum (Shandong Fufeng Fermentation Co., Ltd., Linyi, China), locust bean gum (Tate & Lyle Italia Spa, Syracuse, Italy), sugar-free plum extract (Chamdeul-ae Bio Food, Sancheong, Republic of Korea), plum flavor (Jinhwang, Incheon, Republic of Korea), erythritol (Sandong sanyuan biotechnology Co., Ltd., Binzhou, China), and sucralose (Sunvision sweet Co., Ltd., Xintai, China). Allulose syrup (with a solid content of 70.4 %) was supplied by Daesang Corporation (Seoul, Republic of Korea). For the antioxidant activity analysis, the standard substance ascorbic acid was purchased from Sigma-Aldrich Co. (St. Louis, USA). 2,2′-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) was purchased from Wako pure chem Co., Ltd. (Osaka, Japan). Ethanol was obtained from Duksan Pure Chemical Co., Ltd. (Seoul, Republic of Korea).

2.2. Biomass preparation

The collected *EB* was immediately washed several times with fresh water to remove odors and then dried in a dry oven (WOF-155, Daihan Scientific Co., Ltd., Seoul, Republic of Korea) at 50 °C for 2 days (Oh et al., 2021). The dried biomass was ground to a size of 450 − 1000 μm and then dried for one more day until the weight became constant. The biomass was stored at 4 °C in a plastic bottle until further analysis. Prior to all experiments where biomass was used, the biomass was dried at 50 °C for 6 h to obtain the extraction results on a dry weight basis.

2.3. Extraction procedures

In order to investigate the effect of solvent (prethanol A) concentration and time on dieckol yield, 2 g of dried biomass was soaked in 20 mL of various concentrations of prethanol A such as 25 %, 50 %, 75 %, and 95 % prethanol A mixed with distilled water (Shin et al., 2023). The extraction was conducted in a 50 mL plastic tube at room temperature. Samples were collected in 1 mL aliquots at 1, 2, 3, 4, 5, 6, and 24 h, and the samples were analyzed to determine the dieckol concentration. The extracts were stored frozen before the analysis. For the RSM modeling, all extractions were performed in a water bath (BHS-2, JOANLAB, Huzhou, China). All experiments were performed in triplicate, and the results are presented as the average values.

2.4. Experimental design and analysis for RSM modeling

In order to statistically optimize the extraction conditions with maximum dieckol yield, a central composite rotatable design of RSM was used. Table 1 shows extraction parameters (such as solvent concentration, temperature, extraction time) and designed range. The range of temperature was designed based on the boiling point of prethanol A (78.5 °C). The ranges of solvent concentration and time were designed after the fundamental experiments (Section 3.1). The experimental response for the RSM modeling was dieckol extraction yield, which is defined as mg of dieckol recovered from g of dried biomass. Based on the experimental response, the RSM modeling was performed using the following formula (Bellebcir et al., 2023):

$$Y = \beta_0 + \sum_{i=1}^{k}\beta_i X_i + \sum_{i=1}^{k}\beta_{ii}X_i^2 + \sum_{i=1}^{k}\sum_{j=i+1}^{k}\beta_{ij}X_iX_j,$$

where Y is the response value (i.e., dieckol extraction yield), β_0 is the offset term, β_i, β_{ii}, and β_{ij} are regression model coefficients, respectively (Abubakar et al., 2023). K is the number of parameters (k = 3 in this study). X_i and X_j indicate the input parameters (A, B, and C in this study). The RSM modeling (regression analysis) was performed using Design-Expert (Stat-Ease Inc., Minneapolis, MN, USA).

After the regression analysis, the analysis of variance (ANOVA) was carried out to assess the statistical significance of the model equation using the Design-Expert software. Then, numerical optimization was conducted based on the model equation in order to obtain the optimum conditions to get maximum dieckol extraction yield. Afterward, the confirmation experiments were carried out to evaluate the RSM model-based optimization results by comparing the model-predicted yield and experimental yield. All experiments were carried out in triplicate, and the results are presented as average values.

2.5. HPLC analysis for quantification of dieckol in EB extract

The dieckol concentration in *EB* extracts were determined by high-performance liquid chromatography-diode array detector system (HPLC-DAD, Hitachi, Tokyo, Japan). For analysis, INNO Column C18 (5 μm, 4.6 mm × 250 mm, Young Jin Biochrom, Seongnam-si, Korea) was used. For solvent, 0.1 % trifluoroacetic acid in water (solvent A) and 0.1 % trifluoroacetic acid in acetonitrile (solvent B) were used. The flow rate was 0.8 mL/min, injection volume was 5 μL, and monitoring wavelength

Table 1
Experimental design for response surface modeling.

Extraction parameter	Unit	Symbol	Coded Level				
			−α[a]	−1	0	1	α[a]
Solvent concentration	%	A	6.36	20	40	60	73.64
Temperature	°C	B	24.77	35	50	65	75.23
Time	min	C	6.14	30	65	100	123.86

[a] where α is the 1.682 in this design.

was 230 nm. The gradient elution was performed (min, % of solvent A:B): 0 min, 90:10 %; 15 min, 70:30 %; 30 min, 50:50 %; 35 min, 40:60 %; 38 min, 90:10 %; 40 min, 90:10 %. To quantify the dieckol concentration, the standard curve was generated by analyzing different concentrations of dieckol standard material. After that, dieckol yield was calculated from dieckol concentration, solvent volume, and biomass weight (dry weight basis), and the results are presented as mg/g biomass (i.e., dried *EB*).

2.6. Preparation of dieckol-containing jelly with different amount of Eisenia bicyclis (EB) extracts

Jelly was prepared with varying amounts of *EB* extract containing dieckol. Based on the recommended daily intake of 9.954 mg dieckol by the Korea MFDS (2018), jellies were prepared to contain 0 %, 25 %, 50 %, 75 %, and 100 % of this amount, designated as DJ 0, DJ 25, DJ 50, DJ 75, and DJ 100, respectively. In preliminary experiments, other ingredients were prepared according to the mixing ratios shown in Table 2. Sweeteners including allulose syrup, erythritol, and sucralose were dissolved in purified water at 80 °C in a water bath (SWB-20 L-3, Major Science, Taoyuan City, Taiwan) for 20 min. Subsequently, gelling agents such as konjac powder, carrageenan, xanthan gum, and locust bean gum were added. The mixture was homogenized at 6000 rpm for 2 min using a homogenizer (T25, IKA, Königswinter, Germany). Then, *EB* extracts, sugar-free plum syrup, and plum flavor were added and evenly dispersed under stirring at 80 °C using a stirrer (HS12-60P, Misung Scientific Co., Ltd., Yangju, Republic of Korea). The final mixture was molded into 20 g portions, cooled at room temperature for 20 min, sealed, and stored at 4 °C for 1 h before analysis.

2.7. Characterization of dieckol-containing jelly with different amount of Eisenia bicyclis (EB) extracts

2.7.1. Measurement of color, pH, and syneresis

The surface color values of the manufactured jellies (2.5 × 6 × 1.5 cm) were measured using a colorimeter CR-300 (Minolta co., Osaka, Japan). Measurements were performed in triplicate for each sample, focusing on lightness (L*), redness (a*), and yellowness (b*). The color values for the standard white plate used were L* = 96.90, a* = 0.24, and b* = 1.97. The results were represented as the average values.

For the pH measurement, 5 g of the sample was placed in 45 mL of distilled water and homogenized for 1 min using a processor (MQ3135, Braun, Kronberg, Germany) following the method of Renaldi et al. (2022) with slight modifications. The mixture was then centrifuged (Super22K, Hanil, Incheon, Korea) at 3000 rpm and 4 °C for 10 min.

Table 2
Ingredient of dieckol-containing jelly with different amount of *Eisenia bicyclis* (*EB*) extracts.

Ingredient	Samples				
	DJ 0	DJ 25	DJ 50	DJ 75	DJ 100
Purified water	77	74	72	65.9	67
Eisenia bicyclis extracts (with about 5 mg/mL of dieckol)	0	2.5	5	7.5	10
Konjac powder	0.4	0.4	0.4	0.4	0.4
Kappa-carrageenan	0.6	0.6	0.6	0.6	0.6
Xanthan gum	0.1	0.1	0.1	0.1	0.1
Locust bean gum	0.4	0.4	0.4	0.4	0.4
Sugar-free plum syrup	13	13	13	13	13
Plum flavor	3	3	3	3	3
Allulose syrup (solid content 70.4 %)	4.6	4.6	4.6	4.6	4.6
Erythritol	0.87	0.87	0.87	0.87	0.87
Sucralose	0.03	0.03	0.03	0.03	0.03
Total	100	100	100	100	100

DJ means dieckol-containing jelly, and 0, 25, 50, 75, and 100 indicate the percentage of the recommended daily intake of dieckol contained in one jelly.

Afterwards, the supernatant was mixed using a stirrer (HS12-60P, Misung Scientific Co., Ltd., Yangju, Korea), and then its pH was measured using a pH meter (S20 Seven easy, Mettler Toledo, Columbus, Ohio, USA).

Syneresis was measured according to the method of Mahdavi et al. (2016). Each sample was cut into uniform sizes (2 × 2 × 1.5 cm), and the initial weight of jelly was measured. The jelly was placed in a petri dish and stored at 4 °C for 5 days, and then the syneresis of jelly was calculated as follows:

$$Syneresis\ (\%) = \frac{Weight\ of\ separated\ liquid\ (g)}{Initial\ weight\ of\ jelly\ (g)} \times 100$$

2.7.2. Evaluation of mechanical properties

The texture of each sample cut into dimensions of 1 × 1 × 1.5 cm was analyzed using a texture analyzer (TA-XT2i, Stable Micro Systems, Surrey, UK). For measurement, a 10 mm cylinder probe was used in the texture profile analysis (TPA) mode and pre, test, and post speeds of 2.0, 2.0, and 10.0 mm/s, respectively. Hardness, cohesiveness, springiness, chewiness, and adhesiveness were analyzed, and the results were represented as the mean of 10 repetitions.

2.7.3. Evaluation of antioxidant activities

ABTS radical scavenging activity was measured according to Miller and Rice-Evans (1996) method. ABTS radicals were prepared by mixing 7 mM ABTS in a water solution with sodium persulfate (2.45 mM in the final solution) and incubating at 30 °C for 24 h in the dark. Afterward, the ABTS solution was diluted in distilled water to achieve the absorbance of the solution at the level of 1.0 ± 0.05 measured at 745 nm. 800 μL of the diluted solution was added to 200 μL of each sample (ethanolic extract from DJ), and the mixture was reacted in the dark for 20 min. The absorbance of the reaction mixture was then measured at 745 nm using the spectrophotometer. The standard curve was established using ascorbic acid as the standard reagent, at concentrations of 0, 0.01, 0.02, and 0.04 mg/mL, resulting in a correlation coefficient (r^2) of 0.98. The ABTS radical scavenging activity of the samples was expressed in terms of mg ascorbic acid eq/mL.

2.8. Sensory evaluation

Sensory evaluation was conducted with 33 general consumers (ages: 20–40 years; 20 males and 13 females). Samples were placed on disposable plates labeled with three-digit random numbers and served with bottled water. Evaluation criteria included appearance, color, flavor, taste, texture, and overall preference, assessed using a seven-point hedonic scale (1 = "extremely dislike" to 7 = "extremely like"). The test followed ethical principles approved by the University's Research Ethics Committee (SMU IRB (ex-2023-008)).

2.9. Statistical analysis

The statistical significance between dieckol yields in each time of 25 % prethanol A-based extraction process was analyzed based on a one-way ANOVA and Tukey's test by SPSS Statistics 29.0 (IBM-SPSS Inc., USA). The experimental data for the characterization of jelly were determined at least three times. The measurement results were subjected to analysis of variance (ANOVA) using SPSS (version 27.0). The significant difference between each jelly was tested at $p < 0.05$ with the Duncan test.

3. Results and discussion

3.1. Effect of solvent concentration and extraction time on dieckol yield from Eisenia bicyclis (EB)

The effects of solvent concentration and extraction time on dieckol yield from biomass were investigated in order to design experimental range for RSM modeling (Table 1). Fig. 1 shows time profiles of dieckol yield from *EB* (mg/g biomass) during maceration extraction using prethanol A solvent. The dieckol extraction capacity was higher in the order of 25 %, 50 %, 75 %, and 95 % prethanol A. The extraction process using water resulted in a dieckol yield of only about 0.4 mg/g biomass after 24 h (data not shown). The poor water solubility of dieckol has been reported (Woo et al., 2021), thus water was not considered as a solvent for the dieckol extraction process in this study. The 95 % solvent showed a dieckol yield of approximately 0.5 mg/g biomass at all time points analyzed. In the water-prethanol A mixture, increasing the percentage of water improved the dieckol yield. Previous studies have shown that binary solvents are more efficient for extracting phenolics than monosolvents, and the mixtures of water and organic solvent can improve the extraction of phenolics by creating a more polar medium (Chaves et al., 2020; Shin et al., 2023; Sonar & Rathod, 2020). In addition, the mixture of water and ethanol can increase the contact surface of biomass and ethanol by weakening the bonds between polyphenolics–protein and polyphenolics–cellulose, improving the extraction yield of phenolics (Hobbi et al., 2021; Rodríguez De Luna et al., 2020). While the maximum dieckol yield (at 1 day) of 75 % solvent was only 2.12 ± 0.22 mg/g biomass, 25 % and 50 % solvent recovered dieckol with yields of 5.32 ± 0.65 and 5.31 ± 0.48 mg/g biomass, respectively. In particular, dieckol extraction from *EB* using 25 % solvent achieved higher dieckol yields up to 5 h compared to 50 % solvent. Overall results suggest that solvent concentration is one of the key extraction parameters, and similar to the present study, Shin et al. (2023) have also reported that solvent (ethanol) concentration had a statistically significant effect on dieckol yield from *E. cava*. Therefore, solvent concentration was reflected in the RSM modeling for dieckol extraction from *EB*. The range of this parameter was set to 6.36–73.64 %, which is expected to result in relatively high yields. On the other hand, the range of extraction times was set to 6.14 − 123.86 min because all intervals between 2 and 24 h were not statistically significant in the 25 % solvent-based process, which had the best dieckol extraction capacity (Fig. 1). In the fundamental experiment of dieckol extraction with 50 % solvent at 50 °C, no further increase in dieckol yield was observed even after 2 h (data not shown).

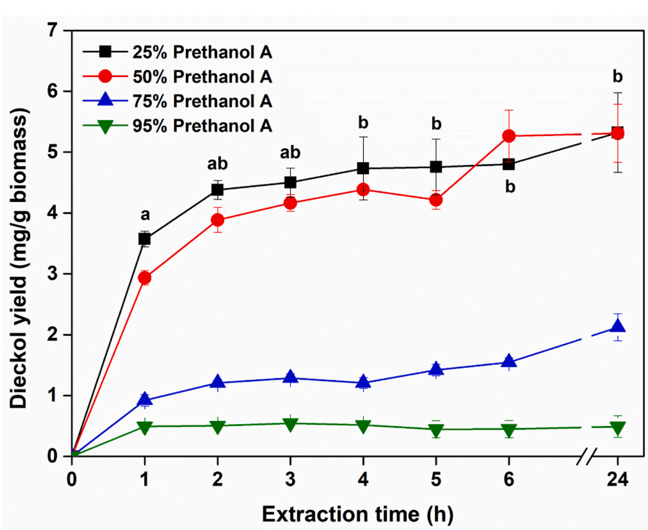

Fig. 1. Effect of solvent concentration and time on dieckol yield from *Eisenia bicyclis* (dry weight basis). Extraction experiments were performed at room temperature. The data is presented as mean values. Error bar indicates standard deviation ($n = 3$). In black line (25 % prethanol A), data with different letters are significantly different ($p < 0.05$).

3.2. Production of dieckol-rich extracts: RSM approach

3.2.1. Development of a regression model for predicting dieckol yield

For the development of the regression model predicting dieckol yield from *EB*, experiments were performed under RSM-designed runs. Table 3 lists the RSM design and experimental results. Based on the results, the model formula (1) was derived by regression analysis as follows:

$$\begin{aligned} \textit{Dieckol yield}\ (mg/g\ biomass) = {} & 12.791 + 1.681\,A + 1.671B + 0.660C \\ & + 1.183AB – 0.068\,AC – 0.00017\,BCE \\ & –1.956A^2 – 0.294B^2 - 1.558C^2, \end{aligned} \tag{1}$$

where Dieckol yield is the model-predicted value, A, B, and C are the input parameters of solvent concentration, temperature, and time, respectively. The equation was expressed based on the coded levels (Table 1). In the coded formula (1), the relative impact of the factors can be identified by comparing the factor coefficients. Terms that have positive signs indicate synergistic effects increasing dieckol yield from *EB*, while terms with negative signs indicate hostile effects (Noor et al., 2022). The predicted values of dieckol yield under the RSM-designed conditions are shown in Table 3.

To evaluate the regression validation, the ANOVA of formula (1) was performed (**Table S1**). The model F-value of 17.66 implies the statistical significance of the model, and there is only a 0.01 % chance that a model F-value this large could occur due to noise. A low *p*-value (generally $<$0.05) of terms implies a significant effect statistically (Moslemi et al., 2023). Therefore, the model terms of A (solvent concentration), B (temperature), C (time), AB, A^2, and C^2 indicate the significant effects, implying the significance of all extraction parameters on dieckol yield from *EB*. In particular, solvent concentration and temperature had significant interaction effects (*p*-value = 0.0098) on dieckol yield. The R^2, adjusted R^2, and predicted R^2 of the model were determined to be 0.9408, 0.8875, and 0.6963, respectively. An R^2 close to 1 means a high correlation between the predicted and experimental values, and since R^2 increases as the number of parameters increases, the adjusted R^2 is provided (Shin et al., 2023). The difference between adjusted R^2 and predicted R^2 indicates model accuracy for predicting response values.

Table 3
Experimental results of dieckol yield under the RSM-designed runs and the predicted value from the developed model.

Run No.	Point type	Coded level			Y: Dieckol yield (mg/g biomass)	
		A	B	C	Experimental [a]	Predicted by the developed model
1	Factorial	−1	−1	−1	5.64	6.09
2	Factorial	1	−1	−1	8.33	7.22
3	Factorial	−1	1	−1	7.26	7.06
4	Factorial	1	1	−1	13.24	12.93
5	Factorial	−1	−1	1	7.84	7.54
6	Factorial	1	−1	1	8.82	8.40
7	Factorial	−1	1	1	8.02	8.52
8	Factorial	1	1	1	15.17	14.11
9	Axial	–α	0	0	5.00	4.43
10	Axial	α	0	0	8.65	10.09
11	Axial	0	–α	0	8.63	9.15
12	Axial	0	α	0	14.42	14.77
13	Axial	0	0	–α	6.86	7.27
14	Axial	0	0	α	9.04	9.49
15	Center	0	0	0	12.44	12.79
16	Center	0	0	0	12.80	12.79
17	Center	0	0	0	11.33	12.79
18	Center	0	0	0	13.69	12.79
19	Center	0	0	0	14.05	12.79
20	Center	0	0	0	12.59	12.79

[a] Extraction experiments were performed in randomized order. Data for modeling was filled in as average values ($n = 3$).

Generally, to predict the response with high accuracy, the difference should be less than 0.2 (Lee, Kim, et al., 2023). In the ANOVA result for the model formula (1), the difference was determined to be less than 0.2, indicating that the model accuracy is high. For a model to fit well, the F-value for lack of fit must be insignificant, and in the model developed, the F-value for lack of fit (p-value = 0.3711) was found to be "not significant". In addition, the adequate precision of the developed model was 13.8904. An appropriate precision of 4 or more indicates that the model is suitable for navigating the designed space (Lee et al., 2022), thus allowing the developed model to appropriately navigate the space. Overall, the ANOVA results proved that the model formula (1) can be used to predict the dieckol yield in statistical optimization.

3.2.2. Effect of extraction parameters on dieckol yield

The single factor effect of solvent concentration, temperature, and time and the interaction effect between solvent concentration and temperature significantly ($p < 0.05$) affect dieckol yield from *EB* (**Table S1**). The effect of extraction parameters on dieckol yield is shown in Fig. 2. For extractions with low concentrations of solvent, temperature-induced changes in dieckol yield were not dramatic, while using higher concentrations of solvent, the dieckol yield improved with increasing temperature (Fig. 2A). The interaction effects of solvent concentration and temperature are clearly shown in Fig. 2B. The effect of solvent concentration on dieckol yield was plotted in different tendencies respective to temperature, demonstrating the interaction effect. In Fig. 1, the dieckol extraction capacity of low concentrations of prethanol A (25 % or 50 %) was high; however, it is important to note that this test was performed at room temperature. RSM modeling identified that an increase in solvent concentration can enhance dieckol yield at relatively high temperatures. The interaction effect of solvent concentration and temperature (i.e., model term AB) had a statistically significant effect (*p*-value = 0.0098) on dieckol yield from *EB* (**Table S1**). In our previous study (Shin et al., 2023), RSM modeling on dieckol yield from *E. cava* (belonging brown algae) was performed, and the results showed no significant interaction effect between solvent concentration and temperature. In contrast, there is a significant interaction effect for dieckol yield from *EB*. The significant interactions between solvent concentration and temperature in the extraction of phenolics have been reported in previous studies (Wang et al., 2017; Yang et al., 2009). This synergistic effect can be attributed to increased solute vapor pressure with rising temperature, which increases solubility (Wang et al., 2014). Fig. 2C and Fig. 2D show the effects of temperature and time and solvent concentration and time, respectively. The dieckol yield from *EB* enhanced as time increased until about 80 min (Fig. 2C). An increase in temperature consistently enhanced the dieckol yield, suggesting that extracting *EB* at a relatively high temperature maximizes yield. On the other hand, increasing the solvent concentration up to 50 % contributed to the enhancement of the dieckol yield, and no drastic

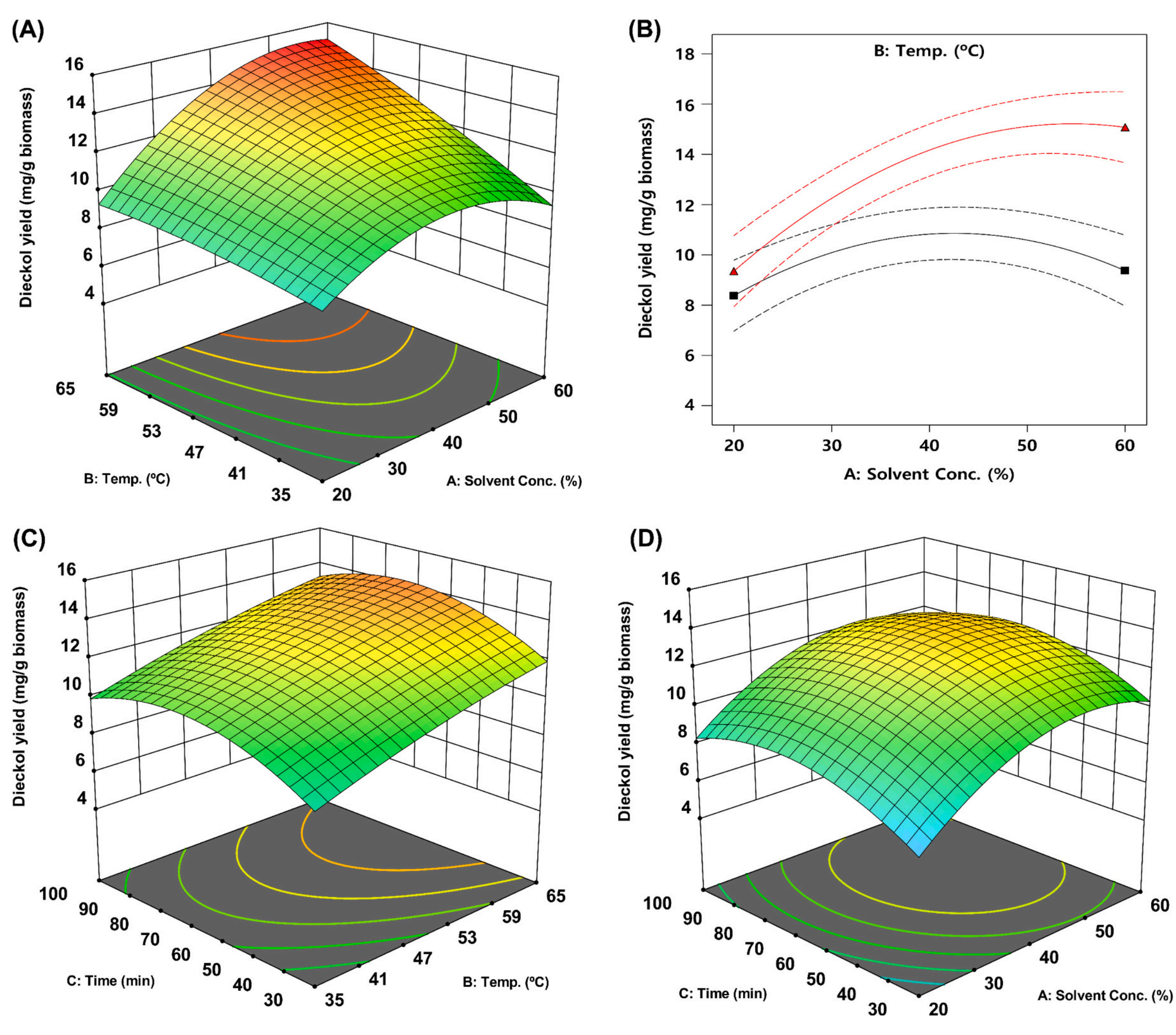

Fig. 2. The effects of parameters in dieckol extraction from *Eisenia bicyclis*. The effects of solvent (prethanol A) concentration and temperature are presented as response surface plot (A) and interaction plot (B) – red line, 65 °C; black line, 35 °C. The effects of temperature and time (C) and solvent concentration and time (D) are presented as response surface plots. The level of the other parameter was fixed at coded level 0. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

improvement in the yield was observed with further increases (Fig. 2D). The influences of these significant parameters are reflected in the model formula (1), thus it is expected to be useful as optimization tool for maximizing dieckol yield from *EB*.

3.2.3. Optimization and experimental validation

To maximize dieckol yield from *EB*, the extraction parameters were optimized by numerical optimization using the model formula (1). The optimization goal was to maximize dieckol yield from *EB* while all parameters were in range. As a result, the optimum extraction conditions were suggested, and the actual experiments were performed for the validation (**Table S2**). After the extractions under four conditions, the actual error values were all low (0.66–5.92 %), demonstrating the prediction accuracy of the RSM model. Finally, the optimum extraction conditions (55.3 % prethanol A, 70.9 °C, and 87.3 min) resulting in the highest dieckol yield were determined. Under the optimum conditions, dieckol yield from *EB* was determined to be approximately 16.5 mg/g biomass. Little work has been undertaken on the design of extraction processes to maximize dieckol yield from brown algae, thus this study highlights the importance and significance of an optimized process for extracting dieckol from *EB*, which resulted in the approximately 2.58-fold recovery of dieckol compared to the optimum process for *E. cava* (Shin et al., 2023).

3.3. Physicochemical characterization of dieckol-containing jelly with different amount of *Eisenia bicyclis* (*EB*) extracts

3.3.1. Color, pH, and syneresis

The color properties, pH, and syneresis of the manufactured jelly containing different extract amount (DJ) are listed in Table 4. The lightness (L*) of DJ 0 (control) was the highest at 54.66, while the experimental groups (DJ 25, 50, 75, and 100) ranged from 32.47 to 38.03, showing a significant decrease with increasing extract content. The appearance of DJ became darker with increasing amounts of the extract (Table 4). The redness (a*) was lowest in DJ 0 at 1.92, significantly increased up to DJ 25, and then decreased with further increases in *EB* extract. The yellowness (b*) was highest in DJ 0 at 12.54, but significantly decreased to 5.88–10.01 with the addition of *EB* extract. The color characteristics of the *EB* extract, compared to the control jelly (DJ 0), showed lower values in L*, a*, and b*as shown in Table 4. Therefore, as the extract content increased in jelly, L* and b* significantly decreased. However, the extract's redness did not proportionally reflect its amount in jelly.

A study by Kim et al. (2010) found that the a* value of jelly with black ginseng extract increased up to 1 % addition and then decreased beyond that level. Similarly, with roselle extract, the L* and b* values decreased with increasing extract amounts, whereas the a* value of the jelly increased up to a certain level (17.5 %) and then decreased, aligning with our results. As the number of pigment particles increases, light scatters in multiple directions, reducing light absorption (Chantrapornchai et al., 1998). According to Wannasin and McClements (2023), higher pigment concentrations reduce light penetration due to increased light scattering, weakening color intensity. This might explain that a* value of extract is not proportional to its amount in the jelly. The color of a product can influence consumer acceptance, affecting taste thresholds and expectations (Gunes et al., 2022). Darker colors in food usually negatively impact consumer choice (Clydesdale, 1993).

The pH of the jelly is shown in Table 4. The pH of the control jelly (DJ 0) at 3.35 is due to the low pH (2.95) of plum syrup, one of the main ingredients. The pH of the jelly increased up to 3.44 with the amount of added *EB* extract, as *EB* extract has a higher pH (5.02) compared to the control. According to Garrido et al. (2015), the gelation and quality of jelly critically depend on soluble solids and pH, with optimal gelation occurring within a pH range of 2.8–3.5. Thus, the pH of all jellies produced in this study falls within the ideal range for proper gelation. Additionally, pH is an important indicator of food stability and preservation (Karastogianni et al., 2016). Foods with a pH below 4.5 can delay spoilage and positively influence gel formation (Karastogianni et al., 2016). Although the addition of *EB* extract slightly increased the pH, it remained below 4.5, suggesting that the jelly's shelf life will be preserved.

Syneresis is the expulsion of liquid from a gel over time, with lower syneresis indicating more stability of the gel's structure (De Alwis & Wijesekara, 2022). The syneresis of jelly showed no significant difference from DJ 0 to DJ 75, ranging from 1.86 to 2.05 % (Table 4). However, DJ 100, with the highest *EB* extract content, showed a significantly lower value at 1.59 %. Akhavan Mahdavi et al. (2016) found that syneresis in jelly with encapsulated *Berberis vulgaris* (barberry fruit) extract

Table 4
Appearance, Hunter's color values, pH, syneresis, and texture properties of dieckol-containing jelly with different amounts of *Eisenia bicyclis* (*EB*) extracts.

Sample	Color values			pH	Syneresis (%)	Texture properties				
	L*	a*	b*			Hardness (N/m^2)	Springiness	Cohesiveness	Chewiness	Adhesiveness
EB extracts	33.19 ± 0.18	0.37 ± 0.40	1.26 ± 0.33	5.02 ± 0.00	–	–	–	–	–	–
 DJ 0	54.66 ± 0.83^{a}	1.92 ± 0.19^{d}	12.54 ± 0.76^{a}	3.35 ± 0.01^{c}	2.05 ± 0.04^{a}	25,465.00 ± 3438.00^{b}	0.87 ± 0.02^{NS}	0.44 ± 0.10^{NS}	0.78 ± 0.27^{b}	1.49 ± 0.15^{b}
 DJ 25	38.03 ± 0.62^{b}	4.67 ± 0.13^{a}	10.01 ± 0.29^{b}	3.36 ± 0.01^{c}	2.00 ± 0.02^{a}	25,847.00 ± 2165.00^{b}	0.88 ± 0.02	0.42 ± 0.03	0.75 ± 0.07^{b}	1.73 ± 1.00^{b}
 DJ 50	35.79 ± 0.64^{c}	3.47 ± 0.13^{b}	6.22 ± 0.56^{c}	3.40 ± 0.00^{b}	1.99 ± 0.03^{a}	28,266.00 ± 1783.00^{b}	0.85 ± 0.01	0.42 ± 0.05	0.79 ± 0.05^{b}	4.79 ± 0.72^{a}
 DJ 75	36.42 ± 0.48^{c}	3.19 ± 0.11^{b}	5.88 ± 0.39^{c}	3.44 ± 0.00^{a}	1.86 ± 0.11^{a}	33,868.00 ± 382.00^{a}	0.88 ± 0.03	0.54 ± 0.00	1.28 ± 0.05^{a}	4.60 ± 0.26^{a}
 DJ 100	32.47 ± 0.27^{d}	2.38 ± 0.22^{c}	6.37 ± 0.49^{c}	3.44 ± 0.01^{a}	1.59 ± 0.19^{b}	34,123.00 ± 637.00^{a}	0.84 ± 0.00	0.50 ± 0.05	1.13 ± 0.14^{a}	4.98 ± 1.36^{a}

DJ means dieckol-containing jelly, and 0, 25, 50, 75, and 100 indicate the percentage of the recommended daily intake of dieckol contained in one jelly. All values are presented as mean ± SD ($n = 3$). Different letters indicate significant differences between values in the same row according to Duncan's multiple range test ($p < 0.05$). NS means not significant.

significantly decreased, indicating enhanced gel strength due to the interaction between the extract and other ingredients. Similar decreases in syneresis were reported in fruit jelly with added fiber powders (wheat, bamboo, psyllium, and apple) (Figueroa & Genovese, 2019) and jelly with theanine extract from green tea (Kim et al., 2021), aligning with our study's trends. Increasing solute concentration within the gel can raise osmotic pressure, hindering moisture movement out of the gel and stabilizing its structure by retaining water (Mizrahi, 2010). These reasons may have contributed to the significantly lower syneresis of DJ 100.

3.3.2. Mechanical properties

The texture of jelly is an important factor in evaluating its quality characteristics (Kopjar et al., 2008). The textural properties of the jelly samples are presented in Table 4. DJ 0, 25, and 50 exhibited hardness ranging from 200 to 222 N/m^2, with no significant differences among them. However, DJ 75 (266 N/m^2) and DJ 100 (268 N/m^2) exhibited a 19.8–34.0 % increase in hardness compared to DJ 0, 25, and 50. This finding aligns with previous studies indicating that higher additive ratios enhance jelly hardness, as seen with aronia juice (Hwang & Shon, 2022) and tomato juice (Hwang & Moon, 2021). Kim et al. (2014) characterized jellies with extracts of four medicinal plants and found that the jelly's strength was significantly higher than the control (without extract), attributing this to phenolic compounds forming cross-links within the gel, enhancing its structural strength (Strauss & Gibson, 2004).

Springiness is defined as the rate of return to original shape after deformation (Nisa et al., 2021), and cohesiveness means a measure of internal bonding strength (Palachum et al., 2023). All samples showed no significant differences in springiness (0.84–0.88) and cohesiveness (0.42–0.54), indicating that *EB* extract content does not affect these properties. Chewiness, reflecting the energy required for mastication to a swallowable state and correlated with hardness (Marti et al., 2014), showed no significant difference between DJ 0 (0.78), DJ 25 (0.75), and DJ 50 (0.79), however chewiness significantly increased in DJ 75 (1.28) and DJ 100 (1.13), exhibiting a similar trend observed in hardness.

Adhesiveness is the force needed to overcome the attraction between food and another material in contact (Marti et al., 2014). The adhesiveness of DJ 50, 75, and 100 ranged from 4.60 to 4.98, significantly higher than those of DJ 0 and 25. Jaiswal et al. (2022) reported that adding more beetroot extract significantly increased jelly adhesiveness, similar to our findings. Dieckol, a phenolic compound (Fukuyama et al., 1989), along with polysaccharides and other phenolic compounds, strengthens the gel's molecular structure through cross-links via hydrogen bonding (Strauss & Gibson, 2004). This likely contributes to increased adhesiveness in gels (Back et al., 2022). Therefore, it is presumed that as the *EB* extract content increased in the jelly, cross-linking formed between the phenol structure of dieckol and polysaccharides, increasing the adhesiveness of DJ. An extreme adhesive jelly may have negative textural characteristics and is less acceptable to consumers (de Souza et al., 2014). DJ 25 showed relatively low adhesiveness, and consumer preferences should be investigated through sensory evaluation of DJ 25 and DJ 50–100.

3.3.3. Antioxidant activities: ABTS cation radical scavenging activity

The ability to scavenge ABTS cation radicals, which are blue/green and can be reduced by antioxidants, is used to evaluate the antioxidant activity of various foods (Floegel et al., 2011). Fig. 3 compares the antioxidant activity of jellies containing different amounts of dieckol. The ABTS radical scavenging capacity of the jelly increased from 0.02 to 0.40 mg ascorbic acid equivalent/mL with increasing dieckol content. The ABTS radical scavenging activity of *EB* extract is linked to dieckol, a marker compound in the phlorotannin class (Kwon et al., 2013). Antioxidant activity in functional foods with brown algal extracts is attributed to phlorotannins, including dieckol and phlorofucofuroeckol-A (Kwon et al., 2013). Nakamura et al. (1996) found that the antioxidant activity of phlorotannins from brown algae *EB* increased with higher concentrations. Kadam et al. (2015) reported that adding brown seaweed (*Ascophyllum nodosum*) extract to gelatin films increased DPPH radical scavenging activity up to 20 times.

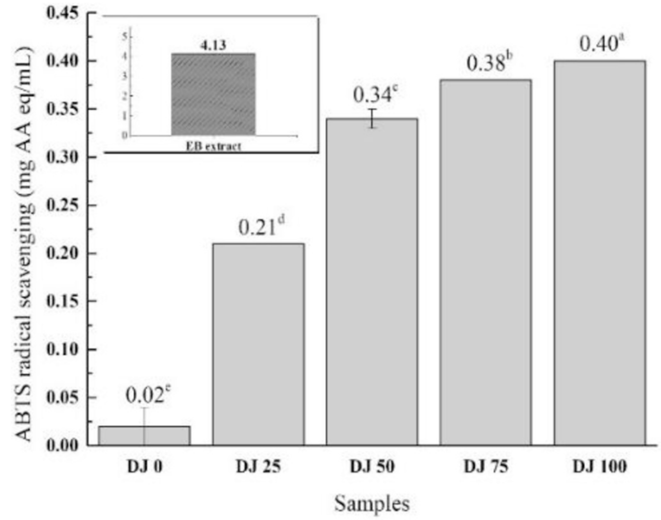

Fig. 3. ABTS cation radical scavenging activity of dieckol-containing jelly with different amount of *Eisenia bicyclis* (*EB*) extracts. DJ means dieckol-containing jelly, and 0, 25, 50, 75, and 100 indicate the percentage of the recommended daily intake of dieckol contained in one jelly. All values are presented as mean ± SD ($n = 3$). Different letters indicate significant differences between values in the same row according to Duncan's multiple range test ($p < 0.05$).

Shibata et al. (2008) compared the antioxidant activities of phlorotannin indicators (eckol, phlorofucofuroeckol A, dieckol, and 8,8′-bieckol) with terrestrial polyphenols (catechin and EGCG) and vitamins (ascorbic acid and α-tocopherol), finding that phlorotannin indicators, except eckol, had antioxidant capacities 2–10 times greater than vitamins. Phlorotannins in brown seaweed have phenolic hydroxyl groups that bind to reactive oxygen species (ROS), reducing their activity and interrupting the chain reaction causing cell damage (Rajan et al., 2021; Zheng et al., 2022). Thus, phlorotannin components with high antioxidant capacity have significant potential in medicine, food, and health products (Zheng et al., 2022). In this study, increased *EB* extract content enhanced the antioxidant activity of the DJs, suggesting that the increased antioxidant activity of DJ was due to various bioactive compounds (including dieckol) in *EB* extract. This indicates that jelly with *EB* extract could be a functional food and highlights the potential of using dieckol-containing *EB* extract in various food development fields in the future.

3.4. Sensory evaluation

The addition of functional ingredients can alter sensory characteristics such as color, flavor, aroma, and texture of the original product, potentially affecting consumer satisfaction (Gunes et al., 2022). Therefore, determining the optimal amount of addition through sensory evaluation is crucial for product development. The sensory evaluation results of developed jelly are presented in Fig. 4. Appearance preference was significantly higher in DJ 0 (5.28) compared to the experimental groups, with no significant difference among samples with dieckol (3.88–4.53). Color preference was highest in DJ 0 at 5.47, decreasing as the extract content increased, but there was no significant difference between the DJ 50, 75, and 100.

Akelom et al. (2022) reported a decrease in overall liking with an increase in *Moringa oleifera* leaf extract content in jelly, indicating that darker-colored products due to extracts can affect to reduce consumer acceptance (Priyanto & Nisa, 2016; Wulandari et al., 2020). Kumar et al.

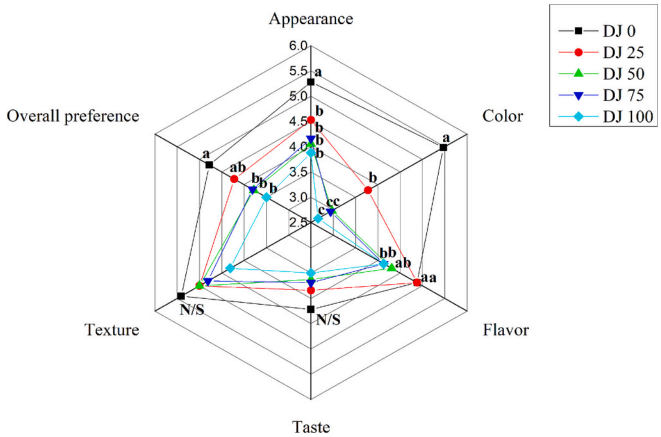

Fig. 4. Sensory evaluation of dieckol-containing jelly with different amount of *Eisenia bicyclis* (*EB*) extracts DJ means dieckol-containing jelly, and 0, 25, 50, 75, and 100 indicate the percentage of the recommended daily intake of dieckol contained in one jelly. All values are presented as mean ± SD ($n = 3$). Different letters indicate significant differences between values in the same row according to Duncan's multiple range test ($p < 0.05$). NS means not significant.

(2018) prepared and characterized biscuits with seaweed (*Caulerpa racemosa*) powder (0, 1, 5, and 10 %), and the 5 % group showed significantly lower scores in all sensory attributes, primarily due to changes in exterior color. Flavor preference was the highest in DJ 0, 25, and 50 (4.31–4.88), while DJ 75 and 100 had significantly lower preferences. Taste and texture did not show significant differences among samples, with ranges of 3.50–4.22 and 4.31–5.41, respectively. Overall preference was highest in DJ 0 and DJ 25 at 4.78 and 4.22, respectively, and decreased in DJ 50, 75, and 100. Based on sensory evaluation results, adding dieckol to the jelly at 25 % of the recommended daily intake (DJ 25) appears to be the most appropriate level. In this experiment, the *EB* extract containing dieckol in jelly had the most adverse impact on appearance and color, reducing overall preference. To develop jelly with higher dieckol content, improving the color of the extract and mitigating the distinctive seaweed flavor are necessary.

4. Conclusions

This study aimed to develop a novel functional jelly with dieckol-rich extract from edible brown macroalga *EB*. The significant extraction parameters were found to be solvent concentration, temperature, and time ($p < 0.05$), and in particular, a significant interaction effect between solvent concentration and temperature was observed. The optimum dieckol extraction process for *EB* achieved a yield of approximately 16.5 mg dieckol/g biomass (dry weight basis), which was 2.58-fold higher than the yield from the earlier reported optimal process for *E. cava*. A novel functional jelly, DJs with the extract containing 25, 50, 75, and 100 % of 9.954 mg dieckol produced, and key findings showed that increasing *EB* extract content in the DJ enhanced antioxidant activity but also increased adhesiveness and hardness, and the DJ 25 showed the best sensory properties. Future research needs to explore ways to balance functional benefits with sensory satisfaction and expand the application of seaweed-derived bioactive compounds in food products.

Ethical statement

The sensory studies in this study followed ethical principles approved by the University's Research Ethics Committee (SMU IRB [ex-2023-008]). The purpose, process, risks, and benefits of the study were adequately explained to all participants and their informed consent was obtained. Participation was voluntary and participants could withdraw from the study at any time.

CRediT authorship contribution statement

Su-Bin Lim: Writing – original draft, Software, Formal analysis. **Jeongho Lee:** Writing – original draft, Software, Formal analysis. **Yoon-Hee Yang:** Methodology, Formal analysis. **Hyerim Son:** Methodology, Formal analysis. **Hah Young Yoo:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Jung-Ah Han:** Writing – review & editing, Resources, Project administration, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2024.102044.

Data availability

Data will be made available on request.

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