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ABSTRACT

This study investigated the production of process flavorings by direct extrusion method using a model system consisted of 80% (w/w) D-xylose and glycine, alanine, or valine at 20% (w/w). The extruder’s barrel temperatures were controlled at 65, 80, and 50 °C. The aroma compounds in the obtained products were analyzed by gas chromatography-mass-spectrometry (GC-MS). Some properties relevant for food application including solubility, water absorption index, and oil absorption capacity were also determined. Characteristics and color profiles of the extruded flavorings were similar to the process flavorings obtained by conventional method (reflux method). Process flavorings using valine presented the highest L* value (lightness, 59.65), water absorption index, water solubility index, and oil absorption capacity at 3.26 and 1.76 g/g sample, and 2.72 g oil/g, respectively. Use of glycine resulted in the highest frequency of just-about-right for the taste (60%), while alanine gave the highest frequency for odor (53.3%). The extruded flavorings exhibited sweet, sour, and meaty flavor with 13 and 12 volatile compounds from glycine and alanine. When valine, the higher molecular weight amino acid in the bitter group was used, 37 volatiles with more complex flavor profiles were found, among of which, the compound 5-methyl-2-phenylhex-2-enal, which exhibits a chocolate odor, was also present. Sweet and meaty process flavorings could be produced by direct extrusion using D-xylose and glycine, alanine, and valine, with low moisture content at only 5% by weight. The information obtained about their physical, taste and sensory properties, volatile profiles, are beneficial for further use in industrial applications.

Keywords: Maillard reaction, process flavorings, extrusion process, gas chromatography-mass spectrometry (GC-MS), amino acid, D-xylose
1. Introduction

Thermally generated flavors are often described as “process flavoring,” which are widely used to deliver flavor characteristics of thermally treated food products such as meat, confectionery, bread, nuts, desserts, and beverages (Weenan, 1998). A non-enzymatic browning reaction called “Maillard reaction” plays a significant part in the food industry and food research due to the formation of color and flavor compounds (Yu et al., 2017). During this reaction, the carbonyl group of reducing sugar condenses with an amine group of amino acid, leading to Amadori products or Amadori rearrangement products (ARPs), (Cui et al., 2018). The reactions lead to furfural compounds and Strecker degradation products and many flavors and aroma compounds such as heterocyclic, aliphatic compounds, and high molecular weight-red or brown color like melanoidin pigments (Hodge et al., 1972; Martins and van Boekel, 2003; Katayama, Tatemichi, and Nakajima, 2017).

Generally, pentose sugars react faster than hexose sugars because of their OH group on the ring, which affects the ring stability, and their ability to participate in the Maillard reaction (Laroque et al., 2008). Methionine and cysteine are the two most crucial amino acids for savory and meat flavor generation. Some amino acids in the sweet and bitter taste group, such as valine, glycine, and alanine, are used to generate sweet confectionery products such as chocolate-like flavor (Granvogl et al., 2012). Strecker aldehydes such as acetaldehyde (pungent, yogurt-like odor, odor threshold 25 mg/L) and 2-methyl propanal (malty, green odor, odor threshold 1 mg/L) were identified from the Maillard reaction using alanine and valine as the amino acid, respectively (Rychlik et al., 1998). Crafack et al. (2014) reported the changing of many volatile compounds such as aldehydes, furans, ketones, pyrroles and other volatile compounds from Maillard reaction and initial intermediate compounds that produced from peptides and reducing sugars in the chocolate production. The type of amino acid is the main point to greatly influence the aroma profile of cocoa and chocolate. Chocolate-like flavor can
be produced from leucine, tyrosine, serine, or valine, mixed with tannic acid, fructose, and propylene glycol, heated at 120°C, with stirring for 30 min (Reineccius, 1995) before spray drying the process flavorings obtained.

For the past five decades, many thermal process flavorings have been produced using various techniques such as reflux at atmospheric pressure, vacuum oven drying, oven drying, and extrusion (Parker, 2015). Sweet-like flavor for food and beverage applications are mainly manufactured via the Maillard reaction using the conventional reflux process, which is time-consuming and challenging to handle since it needs drying after the heating process. Alternative methods with higher efficiency with shorter time for the production to produce process flavorings are required. On the other hand, the extrusion process combines various unit operations such as mixing, cooking, and texturizing into a single continuous process (Fichtali and Voort, 1989), generating very low amount of effluent and wastes. The extrusion technique has been extensively exploited in food industries due to its high generation potential and low cost per outcome unit (Colonna et al., 1984; Korkerd et al., 2016). Reducing sugars and amino acids in the extruder immediately undergo the Maillard reactions (Maga and Kim, 1989). There are few reports in the literature on the manufacturing of meat process flavor by extrusion (Darrington, 1987; Baek et al., 2001). In addition, to the best of our knowledge, there are no reports on the use of extrusion to produce sweet flavorings. Using an extruder instead of the conventional method to produce process flavorings including sweet flavorings would improve the cost-benefit and would increase the production efficiency for a commercial scale application.

Based on the above, this research aimed to produce process flavorings by the reaction between xylose and three selected amino acids in the sweet and bitter groups, namely glycine, alanine, and valine, using the extrusion process. Since pentose sugar exhibits higher reactivity than hexose sugar, D-xylose, which is a typical pentose sugar used in many food products, was
chosen to be used in this study. Some properties of the extruded flavoring compounds required for food application such as solubility, water absorption, oil absorption capacity, and sensory evaluation were determined. The volatile profile of the products obtained were analyzed by gas chromatography-mass spectrometry. The information obtained in this study will support the development of an efficient process to obtain flavoring compounds required for food applications.

2. Materials and methods

2.1. Chemicals

Food grade amino acids, glycine (99%), and alanine (98%), were obtained from Mighty International Co., Ltd. (Thailand), while valine (98%) was purchased from Shanghai Kyowa Amino Acid Co., Ltd. (China). Xylose (98%) was bought from NP Chemical Supply Co., Ltd. (Thailand).

2.2. Production of process flavorings

Three amino acids, namely valine, glycine, and alanine, were added to xylose at the amino acid: xylose ratio of 80:20 (w/w) and the feed moisture was adjusted at 5% (wet weight basis). The mixture was mixed in a mixer (Kenwood KM230, England) for 25 min before being fed into a single screw extruder (Brabender 19/20DN, Germany) with a 19.1 mm screw diameter and a barrel length of 386 mm. The extrusion was operated using the method of Sakunwaropat et al., (2019) modified to a screw speed of 30 rpm, feed rate 50 g/min, die diameter of 1 mm, and with temperature in zones 1, 2, and 3 at 65, 80, and 50 °C, respectively. The extruded process flavorings were dried at 50 °C for 3 h before being ground into powder using a hammer mill (BOSCH MCM3501M, Germany) and passed through a sieve (60 mm mesh size). The powder samples were packed in brown glass bottles and kept at -5 °C for
further analysis. The conceptual image of process flavorings production via Maillard reaction by direct extrusion is shown in Fig. 1.

2.3. Properties of the extruded process flavorings

2.3.1. pH and color

Before and after extrusion, the sample pH was measured following the AOAC method (1990). For analysis, the sample was suspended in water and the pH was measured using a pH meter (Model 510, Cyberscan, Netherland). The color of the sample was measured using a colorimeter (Color Quest XE, USA). In the CIE Lab system, $L^*$ is lightness on a 0 to 100 scale from black and white; $a^*$ (+) red or (-) green; and $b^*$ (+) yellow or (-) blue.

2.3.2. Sensory test

Taste and odor descriptions of the extruded process flavorings were carried out by fifteen trained panelists (6 males and 9 females, aged between 25 and 50 years) at the Department of Research and Development of Mighty International Co., Ltd. This company creates, develops, manufactures, and markets a wide variety of high-quality savory flavors, seasoning, and condiments for customers in the food industry for more than 30 years. The panelists had been trained based on the taste descriptor set, which consisted of 1% of the following samples boiling in water: sweet, sucrose; sour, citric acid; salty, sodium chloride; bitter, caffeine; and umami, monosodium glutamate. The taste and odor descriptions were done according to Wong et al., (2008). One percent of the sample was dissolved in boiling water. Then, 20 mL of the sample was served at room temperature and at 60 °C. The samples were coded with 3-digit numbers and served in a randomized manner. The detection and evaluation of the flavoring solution used a simple descriptive test.
The taste or aroma intensity was evaluated using a “just-about-right rating” (JAR) modified from Hoppert et al., (2013) with corresponding checked boxes label with “weak” (left), “just-about-right” (midpoint), and “strong” (right), using a commercial chocolate product as a reference. Values of 1, 2, or 3 were assigned to each category in the JAR assessments, respectively.

2.3.3. Water absorption index (WAI) and water solubility index (WSI)

WAI and WSI were determined according to Franklin et al., (2017). One gram of the sample was suspended in 10 mL of distilled water, stirred in a vortex mixer for 5 min, and centrifuged at 3,500 g for 15 min. The supernatant was evaporated and dried at 102 ºC to a constant weight. WAI and WSI were calculated according to Eq. (1) and (2), as follows:

\[
\text{Water absorption index (WAI)} = \frac{W_g}{W} \quad (1)
\]

\[
\text{Water solubility index (WSI)} = \frac{W_s}{W} \quad (2)
\]

where ‘W’ is the weight of dry solids in the original sample (g), ‘W\_g’ is the weight of sediment (g), and ‘W\_s’ is the weight of dissolved solids in the supernatant (g).

2.3.4. Oil absorption capacity (OAC)

The oil absorption capacity was determined by mixing 100 mg of the sample with 1 mL of refined cooking palm oil (Morakot Industries Public Company Limited, Thailand). After 30 min of mixing, the sample was centrifuged at 3,000 g, 4 ºC for 10 min. The supernatant was carefully removed with a pipette, and OAC was expressed as g oil/ g sample.
2.4. Gas chromatograph-mass spectrometer (GC-MS) analysis

The volatile compounds in the process flavorings were analyzed using a gas chromatography-mass-spectrometry, GC-MS (5977B MSD; Agilent Technologies Inc., Santa Clara, CA, USA). For the analysis, 1 g of the extruded process flavorings was placed in a 20 mL vial containing 5 mL of distilled water, with headspace extraction temperature and time of 60 °C and 40 min, respectively, before injection into the GC column. The headspace solid-phase microextraction (HS-SPME) with a Teflon-lined septa, 50/30 µm divinylbenzene/carboxen/polydimethylsiloxane (DVB/CAR/PDMS) fiber (Supelco Inc., Bellefonte, PA) was used. The injector was set at 250 °C with desorption time at 5 min using a GC capillary column HP-5MS (30 m × 0.25 mm i.d. × 0.25 µm film thickness) with a split ratio of 1:1. The oven temperature was maintained initially at 40 °C for 5 min, then programmed to increase from 40 to 250 °C at a rate of 4 °C/min. Electron-impact mass spectra were generated at 70 eV, with m/z scan range from 40 to 500. The MS source temperature was set at 250 °C. The volatile compounds were identified according to NIST 14 mass spectra libraries (Yang et al., 2015) installed in the GC-MS equipment.

2.4.1. Identification of volatile compounds

Mass spectrum and linear retention index (LRI) with reference compounds were used to compare in the identification. LRI was calculated taking into account the retention times of standard alkane (C5-C19) series, run under the same GC-MS conditions, and compared to those of authentic or published compounds reported in the literature (Dool and Kratz, 1963), according to Eq. (3)

\[ \text{LRI} = 100N + 100n(tR,a− tR,N) / (tR(N+n) − tR,N) \] (3)

where N is the carbon number of the lower alkane and n is the difference in carbon number of the two n-alkanes that bracket the component; tR, a, tR, N, and tR(N+n) are the retention times.
of the unknown (or test) component, the lower alkane, and the upper alkane, respectively (Dool and Kratz, 1963). The odor description of each compound was reported based on the database of the website http://www.thegoodsentscompany.com/ and published literature data. Peak area integration was done to indicate the relative concentration of each peak.

2.5. Data analysis

All experiments were performed in triplicate. Analysis of variance (ANOVA) was used to analyze statistical significance using SPSS 17 for Windows (SPSS Inc., Chicago, Ill. USA). The data were subjected to one-way ANOVA, and comparison of means was made by Duncan’s multiple range test (DMRT) for a significance level of ($p < 0.05$).

3. Results and discussion

3.1. Appearance and color

The process flavorings obtained by extrusion presented similar moisture content in the range of 0.23-0.33%. The pH of the initial mixture of D-xylose and each amino acid ranged between 5.90 – 5.96, and reduced to 4.29 - 4.56 after the extrusion. The color and appearance of the process flavorings after extrusion are shown in Table 1. The color values show that the extent of pigment formation from xylose and each amino acid occurred in the extruder in the order valine > glycine > alanine, with L* values of 59.68 ± 1.05, 49.24 ± 0.05 and 27.22 ± 0.34, respectively. The cross-linking of peptides or free amino acids with reducing sugar and condensation by Maillard reaction leads to the formation of brown high-molecular-weight compounds or melanoidins (Manzocco et al., 2000). Melanoidins are brownish colorants comprised of nitrogenous polymers and copolymers of high molecular weight (Cammerer and Kroh, 1995; Martins and Boekel, 2003). Yellow color as $b^*$ of the products resulting from the use of glycine (17.87 ± 0.04) and valine (22.86 ± 0.19) was much higher than that of alanine.
The color of process flavorings is related to the condensation of carbonyl or aromatic compounds with amino acid, polymerized, and yielded the cross-linking heterogeneous melanoidin (Mohsin et al., 2018). Generally, essential compounds such as heterocyclic and reductone are formed from fission reaction and dehydration of Maillard reaction intermediates (Coleman and Chung, 2002). Many flavoring compounds and melanoidins that contribute to the brown color and desirable aroma or odor in food are formed from such reactions (Coleman and Chung, 2002; Cui et al., 2017).

3.2. Taste and odor description

The taste description of the process flavorings from D-xylose with glycine and alanine evaluated by the trained panelists presented sweet and sour with an additional meaty taste for glycine (Table 1). Sweet and acidic aroma were found from glycine and alanine, with cocoa aroma for glycine, while valine exhibited a bitter taste with burnt, pungent and explicit chocolate odor. Wong et al. (2008) studied the aroma produced from glucose and amino acids by boiling each mixture in acidic conditions at 100 °C and observed that glycine and valine presented a pleasant caramel-like odor while alanine produced a fruity and flowery odor. In fact, the use of different amino acids and reducing sugar affects the functional properties, odor and color of the product since many intermediates and reaction compounds are formed during the Maillard reaction (Aljahdali and Carbonero, 2019; Starowicz and Zielin´ski, 2019). In addition, volatile compounds are concatenated to flavor, which subsequently influences acceptance and aroma components (Zhang et al., 2019)

Concerning the JAR assessment taste evaluation, a higher frequency for too weak was observed for alanine (Fig. 2). Glycine resulted in the highest just-about-right for the taste (60%). In terms of odor, alanine use indicated the highest just-about-right frequency (53.3%), followed by glycine, and valine at 33.3, and 20 %, respectively. Valine presented a too strong
taste of its bitterness and too strong odor. However, the panelists indicated that the use of valine gave typical chocolate flavor. Further study on the mixture ratio and extrusion conditions would be useful to help improving the flavor profiles of the obtained products.

3.3. Physicochemical properties

Physicochemical properties are essential characteristics to be considered for processing of food flavoring compounds before consumption, such as for mixing, cooking, and frying purposes. As can be seen in Fig. 3, the use of different amino acids significantly affected the WAI, WSI, and OAC of the extruded flavoring compounds \((p < 0.05)\). Valine gave the highest WAI, WSI and OAC, corresponding to 3.26, 1.76 g/g sample, and 2.72 g oil/g, respectively. The products from alanine presented lower WSI and OAC. The results could be due to the different intermediates and products obtained during the reaction in the extruder with various amino acids. Amadori rearrangement products are the initial reaction intermediates that can be decomposed into \(\alpha\)-dicarbonyls, which are highly reactive intermediate products and react further rapidly into a higher complex mixture containing volatile flavor compounds and browning pigments (Troise et al., 2016; Wang and Ho, 2010). The water solubility of these pigments depends on their molar mass and polarity. Those compounds with low molar mass are soluble in ethanol, chloroform and partially soluble in organic solvents of low polarity.

3.4. Identification of volatile compounds of the extruded process flavorings

The volatile compounds in the process flavorings obtained from D-xylose and each amino acid are presented in Tables 2-4. In total, 13, 12, and 37 volatile compounds were identified in the products obtained from glycine, alanine, and valine, respectively. The main compounds with glycine and alanine included aldehyde, alcohol, ester, phenol, and furfural compounds, as shown in Tables 2 and 3. Acetaldehyde was detected from D-xylose and
alanine. This result is in agreement with Grandvogl et al., (2012), who reported the acetaldehyde as the Strecker aldehyde formed in a dry-or low-moisture Maillard system. Acetaldehyde was produced from the breakdown and dehydration of moieties starting from Amadori rearrangement products at the intermediate stage of the 2,3-enolisation of Amadori product (Parker, 2015). The alcohol compound contributed to citrus, fresh floral, oily, and sweet odor, 2-ethyl-1-hexanol was detected in both products from glycine and alanine. Tetradecane with the mild waxy odor was found using glycine and valine, while 3-mercaptophexyl acetate with the floral and fruity odor was detected in those of alanine and valine.

Three compounds were found in all products, namely, nonanal, 2-amino-4-methylbenzoic acid, and furfural. Nonanal exhibits waxy, aldehydic, rose, fresh orris, orange peel, and fatty odor, while 2-amino-4-methylbenzoic acid exhibits fruity, concord grape, musty with a floral, powdery nuance. A relatively higher concentration (higher peak area) of furfural (non-S-containing compound) was found in the reaction using alanine and valine, compared to that of glycine. Furfural gave sweet woody, almond fragrant, baked bread-like odor. The carbonyl-amine condensation reaction between pentoses sugar such as D-xylose and amino acid led to a highly reactive C5 moiety, some of which ends up as 2-furfural compounds via hydrolytic and \( \alpha \)-dicarbonyl cleavage reaction of 3-deoxyhexosulose (Yaylayan and Keyhani, 2000). Furfural and furanone were produced from the oxidation and fragmentation of the specific intermediates of 1,2-enaminol and \( \alpha \)-dicarbonyls (Cui et al., 2019).

This is the first time the production of process flavorings from D-xylose with glycine, valine or alanine by extrusion method is reported in the literature. Yu et al., (2018) investigated the volatile compounds from the Maillard reaction of glucose and glycine by a high-intensity ultrasound method using a continuous high-intensity ultrasound processing system and thermal processing in a shaking water bath. The results showed that pyrazines were the main
compounds in both products with flavor description as caramel, cocoa, nutty, and roasted odor. However, the production of the sweet flavorings required a very short time for the reaction. Though the volatile compounds obtained were not the same, the product obtained by extrusion in the present study exhibited some similar odor descriptions, including cocoa, nutty, and roasted aroma.

More aldehyde compounds were identified in the process flavorings using valine (7 compounds) especially the 5-methyl-2-phenylhex-2-enal which indicates aldehydic, bitter, cocoa, nut skin green, sweet, chocolate, fruity, rancid butter, while the use of glycine and alanine showed only 1, and 3 aldehyde compounds, respectively, as shown in Table 2-4. Two Strecker aldehydes, namely 2-methylpropanal and 3-methylbutanal, were also detected with the use of valine. Grandvogl et al., (2012) identified a series of semi-stable intermediates in the form of oxazolines in a low-moisture system that released the Strecker aldehydes, 2-methylpropanal from the Maillard reaction between reducing sugar and valine. Among these compounds, the compound 2-methylpropanal has a malty flavor with an odor threshold of 0.5 μg/Kg.

About 8 ester compounds were detected from the reaction between D-xylose and valine while only 3 and 2 ester compounds were found with the use of glycine, and alanine, respectively. Esters could be formed during the Maillard reaction. It might be generated from the transformation of alcohols to esters, promoted by hydrogen bonds using oxygen as the oxidant. The high temperature, high pressure, low moisture and shear force used during extrusion could generate oxygen in the gap of screw of the extruder. Several alcohol compounds were generated and led to oxidative self-esterification of these compounds to esters in the system. In fact, direct transformation of alcohols into esters can be achieved using catalysts based on metals such as iridium (Yamamoto et al., 2011) and gold (Miyamura et al., 2010). Liu et al., (2018) reported the transformation of alcohols to esters. The compound 1-
ethyl-3-methylimidazolium acetate could effectively catalyze this kind of reaction using oxygen as an oxidant without any other catalysts or additives. The oxidative self-esterification of benzylic alcohols or aliphatic alcohols and cross-esterification between benzyl alcohols and aliphatic alcohols could all be achieved with high yields. Furans, ketones, and the Strecker aldehydes were aroma-active compounds and important flavor precursors in the Maillard reaction system. Cho et al., (2010) reported that volatile compounds produced from glucose and valine in aqueous systems using a drying oven were heterocyclic compounds, pyrazines, furans, and furan derivatives. Heterocyclic compounds containing oxygen, nitrogen, sulfur, or combinations of these atoms are significant to thermally processed foods (Nursten, 2000). The results showed that using higher molecular weight (MW) of valine delivered more complex flavor profiles of the process flavorings produced. Valine is the amino acid in the bitter group, with MW (117.10 g) higher than glycine (75.10 g) and alanine (89.10 g), which are in the group of sweet amino acids (Damodaran and Parkin, 2017).

3.5. Correlation between sensory test, taste, and odor description

Fig. 4 summarized the volatile compounds detected by GC-MS in each processing flavoring prepared from D-xylose and three different amino acids. The results indicated good correlation between the taste and odor descriptions expressed by the trained panelists (Table 1). The reaction between D-xylose and glycine, and alanine gave compounds with sweet and sour flavors, while D-xylose and valine exhibited rather bitter taste with more complex flavors and more detected volatile compounds. To be noted, the panelists described more intense flavors and taste likely to be used for cocoa or chocolate for the extruded process flavoring from D-xylose and valine. In addition, volatile compounds are concatenated to flavor, which subsequently influences acceptance and aroma components (Zhang et al., 2019). The information obtained about the physical properties, sensory properties as well as
the volatile compounds in each model in this study could facilitate the fabrication of flavoring compounds with required taste and odor by modifying the type or ratio of amino acids.

3.6. Possible pathway for the formation of volatile compounds

The mechanisms of the Maillard reaction are very complicated with several compounds and many pathways involved. The reaction pathways and effect of different conditions on the formation of intermediate and final products have been studied in the conventional method, aqueous solution, by many researchers (Yu et al., 2017; Grandvogl et al., 2012; Yaylayan and Keyhani, 2000; Moshin et al., 2018). However, to the best of our knowledge, there is no report on the pathway for the formation of volatile compounds from D-xylose and amino acids in the low moisture content, or the dry system before. The reactions occurred under the extrusion were very rapid with low moisture, around only 5% (w/w) of the feed raw substrates.

The possible formation pathways of some volatile compounds found in glycine, alanine, and valine are presented in Fig. 5,6,7, respectively. The reaction in the early stage and intermediate stage of three amino acids might be the same. Nevertheless, intermediate compounds could be different. It might be possible that after feeding the mixed substrates into the extruder, high temperature in the barrel of the extruder could melt sugar and leading to open the chain of D-xylose structure (compound 1, Fig. 5,6,7) (Shahidi et al., 2014). The nucleophilic attack of the amino group, typically the α-amino group of amino acid (compound 2, Fig. 5-7) on the reducing sugar (Parker, 2015) led to the formation of glycated product (Schiff-base) (compound 4, Fig. 5,6,7), which further re-arranged to Amadori product (compound 5, Fig. 5,6,7) (Boateng & Yang, 2021). Using different initial amino acid as precursor led to the generation of different Amadori products or intermediate compounds.
The compounds 1-L-glycine- 1-deoxy-D-xyllose (glycine-Amadori), 1-L-alanine- 1-deoxy-D-xylose (alanine-Amadori), and 1-L-valine- 1-deoxy-D-xylose (valine-Amadori) were probably formed from the same pathways as described in Fig. 5,6,7 (compound 5).

The reaction possibly continued to undergo a 1,2-enolisation reaction, which could have been started from Amadori rearrangement products (APRs) (compound 6, Fig. 5,6,7). ARPs exist in equilibrium with their keto-enol tautomers (Parker, 2015). Enolisation was followed by loss of water, and hydrolysis of the imine to release the amino acid (intact) and the 3-deoxypentosulose (compound 9, Fig. 5,6,7), (Weenen, 1998). The polymerization and dehydration of 3-deoxypentosulose (compound 9, Fig. 5,6,7) produced furfural (compound 10, Fig. 5,6,7) (Brands and van Boekel, 2001). Furfural was detected in the process flavoring compounds obtained from all amino acids. The chemical 3-deoxypentosulose could also undergo fragmentation reactions to form highly reactive short-chain intermediates (Parker, 2015) such as methylglyoxal (compound 11, Fig. 7) and 2-hydroxyethanal or glycolaldehyde (compound 12, Fig. 7).

For Strecker degradation and final stages, the reaction between D-xylose and valine, which yielded the highest number of volatile compounds, was selected as shown in Fig. 7. It is possible that valine (compound 13, Fig.7) reacted with dicarbonyl (compound 9, Fig.7) and lost water via carbonyl-amine condensation, then leading to Schiff base of the compound (compound 14, Fig.7).

In dry or low moisture reaction, it is possible that 3-methylbutanal (compound 15, Fig. 7) and amino aldehyde sugar have been generated from the structure of Schiff base compound (Route 1, Fig. 7) by the substitution of free amino acids in the system and hydrolysis reaction (Granvogl et al., 2012). The compound 2-methylpropanal (compound 16, Fig. 7) and aminoaldehyde (compound 17, Fig. 7) could be formed by decarboxylation of Schiff base of the compound (compound 14, Fig. 7, route 2) with rearrangement of the structure and hydrolysis.
route 2.1). After rearrangement form of structure in route 2, it is possible that the compound 2-methylpropanol (compound 18, Fig. 7) with amino-aldehyde (compound 17, Fig. 7) have also been generated (route 2.2). The compounds 2-methylpropanol and 3-methylbutanal are important Strecker aldehydes that were detected only in process flavorings from the use of valine (Granvogl et al., 2012). However, there are several possible pathways for the formation of 5-methyl-2-phenylhex-2-enal. The 3-methylbutanal (compound 15, Fig. 7) might react with phenyl acetaldehyde (compound 19, Fig. 7), lose water and form 5-methyl-2-phenylhex-2-enal (compound 20, Fig. 7). This compound was the key volatile compound that exhibited the specific chocolate flavor from the use of valine. The flavor wheel of process flavorings from D-xylose with valine produced via Maillard reaction by direct extrusion is presented in Fig. 8.

4. Conclusions

In this study, processes flavoring compounds were successfully produced from D-xylose in combination with three different amino acids: glycine, alanine, or valine. All products obtained gave a brownish color and could be dissolved in water at room temperature. The product from D-xylose and valine exhibited the highest WAI, WSI, and OAC. The use of sweet glycine and alanine gave process flavorings with sweet, sour, and meaty flavor with 13, 12 identified volatile compounds from glycine and alanine. The flavorings from valine exhibited a more complex flavor character: bitter taste flavor with 37 identified volatile compounds. The findings of the present study allow concluding that it is possible to produce sweet and meat flavoring product from amino acid and reducing sugar via the Maillard reaction using the extrusion method. This open up new opportunities for the food industry since, when compared to the conventional method used to produce flavorings, extrusion is a continuous, high temperature-short time, and more environmentally friendly alternative with almost no effluent formed during the process. Moreover, the extruded product’s flavor profiles may help
predict the potential flavor properties from D-xylose and the different amino acids. Moreover, the flavor profiles obtained from the study could help predicting the potential flavor properties from D-xylose and some other amino acids or nitrogen sources. The addition of other ingredients to react or adjust the odor or taste profiles of the products would potentially facilitate the innovation for many flavoring products with existing and new requirements, by this high efficient processing method.

CRediT authorship contribution statement

Sirinapa Sasanam: Methodology, Software, Data Curation, Writing - Original Draft.

Benjawan Thumthanaruk: Project administration, Supervision. Savitri Vatanyoopaisarn: Project administration, Supervision. Chureerat Puttanlek: Resources, Validation

Dudsadee Uttapap: Resources, Validation. Vasan Rattananupap: Funding acquisition, Project administration.

Solange I. Mussatto: Supervision, Writing - Review & Editing. Vilai Rungsardthong: Conceptualization, Supervision, Writing - Review & Editing.

Declaration of competing interest

The authors have declared no conflicts of interest for this article.

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Savitri Vatanyoopaisarn https://orcid.org/0000-0002-1465-5839

Solange I. Mussatto https://orcid.org/0000-0002-7182-6198
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https://doi.org/10.1088/1755-1315/301/1/012053.


<table>
<thead>
<tr>
<th>Substrate</th>
<th>Appearance</th>
<th>Color</th>
<th>Taste description</th>
<th>Odor description</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>Initial Feed</td>
<td>Extruded product</td>
<td>L*</td>
<td>a*</td>
</tr>
<tr>
<td>Xylose + Glycine</td>
<td></td>
<td></td>
<td>49.2 ± 0.0&lt;sup&gt;b&lt;/sup&gt;</td>
<td>9.9 ± 0.0&lt;sup&gt;a&lt;/sup&gt;</td>
</tr>
<tr>
<td>Xylose + Alanine</td>
<td></td>
<td></td>
<td>27.2 ± 0.3&lt;sup&gt;c&lt;/sup&gt;</td>
<td>6.8 ± 0.1&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>Xylose + Valine</td>
<td></td>
<td></td>
<td>59.6 ± 1.0&lt;sup&gt;a&lt;/sup&gt;</td>
<td>8.5 ± 0.1&lt;sup&gt;b&lt;/sup&gt;</td>
</tr>
</tbody>
</table>

Means in the same column followed by the same letter are not significantly different at p< 0.05 by Duncan’s multiple range test.
Table 2 Identification of volatile compounds of process flavoring from xylose and glycine by extrusion

<table>
<thead>
<tr>
<th>No.</th>
<th>Volatile compound</th>
<th>Odor description</th>
<th>Odor type</th>
<th>LRI</th>
<th>Peak area</th>
<th><strong>Process flavorings</strong></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Aldehyde compounds (1)</td>
<td>Nonanal</td>
<td>Waxy, aldehydic, rose, fresh orris, orange peel, fatty peely</td>
<td>Aldehydic</td>
<td>1108</td>
<td>923700</td>
<td>Gly, Ala, Val</td>
</tr>
<tr>
<td>Alcohol compounds (3)</td>
<td>2-ethyl-1-hexanol</td>
<td>Citrus, fresh floral, oily, sweet</td>
<td>Citrus</td>
<td>1033</td>
<td>401869</td>
<td>Gly, Ala</td>
</tr>
<tr>
<td></td>
<td>5-methyl-2-propan-2-ylcyclohexan-1-ol</td>
<td>Cooling mentholic, minty</td>
<td>Mentholic</td>
<td>1176</td>
<td>73381</td>
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</tr>
<tr>
<td></td>
<td>3,7-dimethyloctan-3-ol</td>
<td>Floral lily, bois de rose, woody, lilac, tea, oily</td>
<td>Floral</td>
<td>1100</td>
<td>36500</td>
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<tr>
<td>Acid compound (1)</td>
<td>2-amino-4-methylbenzoic acid</td>
<td>Fruity, concord grape, musty with a floral powdery nuance</td>
<td>Fruity</td>
<td>928</td>
<td>284909</td>
<td>Gly, Ala, Val</td>
</tr>
<tr>
<td>Ester compounds (3)</td>
<td>Butyl 2-ethylhexanoate</td>
<td></td>
<td>*</td>
<td>1293</td>
<td>291043</td>
<td>Gly, Val</td>
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<tr>
<td></td>
<td>2-methylpropyl 3-hydroxy-2,2,4-trimethylpentanoate</td>
<td>Acidic and sharp, cheese-like, sour milky, tobacco, with fruity nuances</td>
<td>Acidic</td>
<td>1355</td>
<td>111829</td>
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</tr>
<tr>
<td></td>
<td>3-hydroxy-2,4,4-trimethylpentyl-2-methylpropanoate</td>
<td></td>
<td>*</td>
<td>1376</td>
<td>198312</td>
<td></td>
</tr>
<tr>
<td>Furans non-S-containing compound (1)</td>
<td>Furfural</td>
<td>Sweet, woody, almond, fragrant baked bread</td>
<td>Bready</td>
<td>851</td>
<td>34351</td>
<td>Gly, Ala, Val</td>
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<tr>
<td>Furans O-containing compound (1)</td>
<td>Tetrahydrofuran</td>
<td>Ether-like odor</td>
<td>*</td>
<td>627</td>
<td>10468</td>
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<td>Hydrocarbon compounds (2)</td>
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<td>1280</td>
<td>7744</td>
<td></td>
</tr>
<tr>
<td>Alkanes</td>
<td>Tetradecane</td>
<td>Mild waxy</td>
<td>*</td>
<td>1399</td>
<td>12468</td>
<td>Gly, Val</td>
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<tr>
<td>Phenols compounds (1)</td>
<td>Phenol,4,6-di (1,1-dimethylethyl)-2-methyl-</td>
<td></td>
<td>*</td>
<td>1685</td>
<td>16208</td>
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</tr>
</tbody>
</table>

* Means no odor was detected or no report on their odor description/odor profile.

LRI and MS were used as the identification methods.

** Volatile compounds were founded when the listed amino acids were used.
Table 3 Identification of volatile compounds of process flavoring from xylose and alanine by extrusion

<table>
<thead>
<tr>
<th>No.</th>
<th>Volatile compound</th>
<th>Odor description</th>
<th>Odor type</th>
<th>LRI</th>
<th>Peak area</th>
<th>**Process flavorings</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><em>Strecker aldehyde (1)</em></td>
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<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>Acetaldehyde</td>
<td>Pungent, ethereal, fresh fruity, musty</td>
<td>Ethereal</td>
<td>530</td>
<td>774755</td>
<td></td>
</tr>
<tr>
<td></td>
<td><em>Aldehyde compounds (3)</em></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1-ethylpyrrole-2-carbaldehyde</td>
<td>Burnt, roasted, smoky, waxy, aldehydic, rose, fresh orris, orange peel</td>
<td>Roasted</td>
<td>1059</td>
<td>439060</td>
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</tr>
<tr>
<td>3</td>
<td>Nonanal</td>
<td>Waxy, aldehydic, rose, fresh orris, orange peel, fatty peely orange peel, green melon</td>
<td>Aldehydic</td>
<td>1108</td>
<td>329889</td>
<td>Gly, Ala, Val</td>
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<tr>
<td>4</td>
<td>Decanal</td>
<td>Waxy, fatty, citrus orange peel, green melon</td>
<td>Waxy</td>
<td>1210</td>
<td>79809</td>
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<td></td>
<td><em>Alcohol compounds (1)</em></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>2-Ethyl-1-hexanol</td>
<td>Citrus, fresh floral, oily, sweet</td>
<td>Citrus</td>
<td>1033</td>
<td>308295</td>
<td>Gly, Ala</td>
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<tr>
<td></td>
<td><em>Acid compounds (3)</em></td>
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<tr>
<td>6</td>
<td>2-amino-4-methylbenzoic acid</td>
<td>Fruity, concord grape, musty with a floral, powdery nuance</td>
<td>Fruity</td>
<td>928</td>
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<td>Octanoic acid</td>
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<td>1283036</td>
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<td>8</td>
<td>2-methylpropanoyl 2-methylpropanoate</td>
<td>Acidic, dairy, fruity</td>
<td>Acidic</td>
<td>1800</td>
<td>10455</td>
<td>Ala, Val</td>
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<tr>
<td></td>
<td><em>Ester compounds (2)</em></td>
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</tr>
<tr>
<td>9</td>
<td>3-Mercaptohexyl acetate</td>
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<td>Fruity</td>
<td>1260</td>
<td>94190</td>
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<td>10</td>
<td>Propanoic acid, 2-methyl-, butyl ester</td>
<td>Pungent, acidic and dairy-like</td>
<td>Acidic</td>
<td>1376</td>
<td>77851</td>
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<td>11</td>
<td>Furfural</td>
<td>Sweet, woody, almond, fragrant baked bread</td>
<td>Bready</td>
<td>851</td>
<td>2231437</td>
<td>Gly, Ala, Val</td>
</tr>
<tr>
<td></td>
<td><em>Amine compounds (1)</em></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>12</td>
<td>N,2-Dimethyl-1-propanamine</td>
<td>*</td>
<td>*</td>
<td>626</td>
<td>933902</td>
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</tbody>
</table>

* Means no odor was detected or no report on their odor description or odor profile.

RI and MS were used as the identification methods.

** Volatile compounds were founded when the listed amino acids were used.
Table 4 Identification of volatile compounds of process flavoring from xylose and valine by extrusion

<table>
<thead>
<tr>
<th>No.</th>
<th>Volatile compound</th>
<th>Odor description</th>
<th>Odor type</th>
<th>LRI</th>
<th>Peak area</th>
</tr>
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<tbody>
<tr>
<td></td>
<td>Strecker aldehydes (2)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>2-methylpropanal</td>
<td>Fresh aldehydic</td>
<td>Aldehydic</td>
<td>620</td>
<td>50009017</td>
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<tr>
<td>2</td>
<td>3-methylbutanal</td>
<td>Fruity, dry green</td>
<td>Fruity</td>
<td>707</td>
<td>14760642</td>
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<tr>
<td></td>
<td>Aldehyde compounds (7)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>5-methyl-2-phenylhex-2-enal</td>
<td>Aldehydic, bitter</td>
<td>Chocolate</td>
<td>1495</td>
<td>221655</td>
</tr>
<tr>
<td>4</td>
<td>2-methylhex-2-enal</td>
<td>*</td>
<td>Sharp, irritating</td>
<td>876</td>
<td>111987</td>
</tr>
<tr>
<td>5</td>
<td>2-ethylhex-2-enal</td>
<td>Sharp, irritating</td>
<td>Sharp, irritating</td>
<td>898</td>
<td>1578514</td>
</tr>
<tr>
<td>6</td>
<td>Nonanal</td>
<td>Waxy, aldehydic</td>
<td>Aldehydic</td>
<td>1108</td>
<td>4015487</td>
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<tr>
<td>7</td>
<td>(Z)-5-methyl-2-propan-2-ylhex-2-enal</td>
<td>Herbal lavender, woody, green</td>
<td>Herbal</td>
<td>1117</td>
<td>209637</td>
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<tr>
<td>8</td>
<td>2-(2,6,6-trimethylcyclohexen-1-yl) acetaldehyde</td>
<td>Camphoraceous, cooling, woody, oily and fruity</td>
<td>Camphorous</td>
<td>1236</td>
<td>868462</td>
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<tr>
<td>9</td>
<td>2,4,6-cycloheptatrien-1-one, 2-hydroxy-4-(1-methylethyl)-</td>
<td>Phenolic, woody, mossy</td>
<td>Woody</td>
<td>1301</td>
<td>3333008</td>
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<tr>
<td></td>
<td>Alcohol compounds (2)</td>
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<td></td>
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<td></td>
</tr>
<tr>
<td>10</td>
<td>3,7,11,15-tetramethylhexadec-1-en-3-ol</td>
<td>Mild floral, herbal, green</td>
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<td>1345</td>
<td>458286</td>
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<tr>
<td>11</td>
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<td>Spicy</td>
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<td>38049</td>
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<td></td>
<td>Acid compound (2)</td>
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<tr>
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<td>Undecanoic acid</td>
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<td>Fruity, concord grape, musty with a floral powdery nuance</td>
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<td>*</td>
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<td>1047500</td>
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<td>16</td>
<td>4-(2,2,6-trimethylcyclohexyl) butan-2-one</td>
<td>Sweet, floral, balsam, rose, woody</td>
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<tr>
<td>17</td>
<td>2-butanone, 4-(2,6,6-trimethyl-1-cyclohexen-1-</td>
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<tr>
<td>18</td>
<td>5,9-undecadien-2-one, 6,10-dimethyl-(E)</td>
<td>Fresh green, fruity, waxy, rose, woody, magnolia tropical</td>
<td>Floral</td>
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<td>42773</td>
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<tr>
<td>No.</td>
<td>Volatile compound</td>
<td>Odor description</td>
<td>Odor type</td>
<td>LRI</td>
<td>Peak area</td>
</tr>
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<td>-----</td>
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<td>----------------------------------------------------------------------------------</td>
<td>-----------</td>
<td>------</td>
<td>-----------</td>
</tr>
<tr>
<td>19</td>
<td>2-methylbutanoyl 2-methylbutanoate</td>
<td>Acidic, fruity, dirty, cheesy, fermented</td>
<td>Fruity</td>
<td>1076</td>
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<td>20</td>
<td>1-azabicyclo [2.2.2] octan-3-ol</td>
<td>Odorless</td>
<td>*</td>
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<tr>
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<td>3-mercaptopheoxy acetate</td>
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<td>Fruity</td>
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<tr>
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<td>Butyl 2-ethylhexanoate</td>
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<td>*</td>
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<td>Isopentyl 3-hydroxy-2-methylene</td>
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<td>*</td>
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<td>Propanoic acid, 2-methyl-, octyl ester</td>
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<td>Benzoic acid, 4-tert-butyl-, propyl ester</td>
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<td>*</td>
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<tr>
<td>27</td>
<td>Furfural</td>
<td>Sweet, woody, almond, fragrant baked bread</td>
<td>Bready</td>
<td>851</td>
<td>1955657</td>
</tr>
<tr>
<td>28</td>
<td>1H-pyrrole</td>
<td>Sweet, warm, nutty, ethereal</td>
<td>Nutty</td>
<td>948</td>
<td>114615</td>
</tr>
<tr>
<td>29</td>
<td>4-methyl-2-(2-methylprop-1-enyl)-3,6-dihydro-2H-pyran</td>
<td>Green, weedy, cortex, herbal, narcissus, celery</td>
<td>Green</td>
<td>1170</td>
<td>69406</td>
</tr>
<tr>
<td>30</td>
<td>N,N,2,4-tetramethyl benzenamine</td>
<td>*</td>
<td>*</td>
<td>1242</td>
<td>764619</td>
</tr>
<tr>
<td>31</td>
<td>2-methoxy-N,N-dimethylaniline</td>
<td>*</td>
<td>*</td>
<td>1179</td>
<td>10898613</td>
</tr>
<tr>
<td>32</td>
<td>Bicyclo</td>
<td>Spicy, minty, camphoreous</td>
<td>Camphoreous</td>
<td>1019</td>
<td>73830</td>
</tr>
<tr>
<td>33</td>
<td>Tetradecane</td>
<td>Mild, waxy</td>
<td>*</td>
<td>1398</td>
<td>18567</td>
</tr>
<tr>
<td>34</td>
<td>Heptadecane</td>
<td>Fuel-like</td>
<td>Fuel-like</td>
<td>1699</td>
<td>64734</td>
</tr>
<tr>
<td>35</td>
<td>2,5-dimethylhexa-2,4-diene</td>
<td>*</td>
<td>*</td>
<td>1174</td>
<td>236581</td>
</tr>
<tr>
<td>36</td>
<td>6-propyl-1,2,3,4-tetrahydrophenanthrene</td>
<td>*</td>
<td>*</td>
<td>1376</td>
<td>1541425</td>
</tr>
<tr>
<td>37</td>
<td>Alloaromadendrene</td>
<td>Woody</td>
<td>Woody</td>
<td>1430</td>
<td>112168</td>
</tr>
</tbody>
</table>

* Means no odor was detected or no report on their odor description or odor profile.

LRI and MS were used as identification methods.

** Volatile compounds were founded when the listed amino acids were used.
Figure 1. Production of process flavorings from valine and D-xylose by direct extrusion method using a single screw extruder.
Figure 2. Percentage of responses or frequency on process flavorings produced from D-xylose and each amino acid via direct extrusion using just-about-right scales for taste (a) and odor (b). The bars and error bars represent the means, and standard deviation (mean ± SD) with different letters (a, b, c) in the same category (weak, just-about-right, and strong) mean significantly different (p<0.05).
**Figure 3.** Some properties of process flavorings produced from D-xylose and each amino acid via direct extrusion; WAI: water absorption index (g/g), WSI: water solubility index (g/g), OAC: oil absorption capacity (g oil/g). The bars and error bars in the same category (weak, just-about-right, and strong) represent the means, and standard deviation (mean ± SD) with different letters (a, b, c) mean significantly different (p<0.05).
Figure 4. Volatile compounds found in the process flavorings prepared from D-xylose with three different amino acid, glycine, alanine, and valine by direct extrusion.
**Figure 5.** Scheme illustration of possible pathways of the volatile compound, furfural, detected from the reaction between D-xylose and glycine during the extrusion.
Figure 6. Scheme illustration of possible pathways of the volatile compound, furfural, detected from the reaction between D-xylose and alanine during the extrusion.
**Figure 7.** Scheme illustration of possible pathways of some volatile compounds detected from the reaction between D-xylose and valine during the extrusion.

*Volatile compound detected from process flavorings.*
Figure 8. Flavor wheel of process flavorings from D-xylose with valine produced via Maillard reaction by direct extrusion.
Conflict of Interest and Authorship Conformation

• All authors have participated in (a) conception and design, or analysis and interpretation of the data; (b) drafting the article or revising it critically for important intellectual content; and (c) approval of the final version.

• This manuscript has not been submitted to, nor is under review at, another journal or other publishing venue.

• The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.