Downloaded from orbit.dtu.dk on: Mar 21, 2025

DTU Library

Development of a water solubility model of extruded feeds by utilizing a starch gelatinization model

Cheng, Hongyuan; Wang, Hao; Ma, Shifeng; Xue, Min; Li, Junguo; Yang, Jie

Published in:
International Journal of Food Properties

Link to article, DOI:
10.1080/10942912.2022.2046055

Publication date:
2022

Document Version
Publisher's PDF, also known as Version of record

Link back to DTU Orbit

Citation (APA):
Cheng, H., Wang, H., Ma, S., Xue, M., Li, J., & Yang, J. (2022). Development of a water solubility model of extruded feeds by utilizing a starch gelatinization model. *International Journal of Food Properties*, *25*(1), 463-476. https://doi.org/10.1080/10942912.2022.2046055

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

INTERNATIONAL JOURNAL OF FOOD PROPERTIES
2022, VOL. 25, NO. 1, 463–476
https://doi.org/10.1080/10942912.2022.2046055

Taylor & Francis
Taylor & Francis Group

OPEN ACCESS Check for updates

Development of a water solubility model of extruded feeds by utilizing a starch gelatinization model

Hongyuan Cheng[a,b], Hao Wang[a], Shifeng Ma[a], Min Xue[a], Junguo Li[a], and Jie Yang[a]

[a]Institute of Feed Research, Chinese Academy of Agricultural Sciences, Beijing, China; [b]National Food Institute, Technical University of Denmark, Kgs. Lyngby, Denmark

ABSTRACT

This study aimed to estimate the effects of extrusion variables on extruded aquafeed water solubility using a starch gelatinization model. A new equation was derived from a classical starch gelatinization degree model to calculate aquafeed water solubility. After considering the data of five formulations, the average absolute deviation (AAD) of the new equation was 7.97% for the extrudate water solubility and 2.24% for the water solubility index. In the most cases, water solubility and water solubility index increased with an increase in the process screw speed and temperature but decreased with a decrease in water content. On the other hand, when the moisture content increased from 0.2 to 0.3 kg/kg, the water solubility increased from 4.91 to 5.22% (wt) for a typical recipe in the low moisture content extrusion process. The new equation established a way to quantitatively elaborate the relationship between the extrudate water solubility and extrusion process variables.

ARTICLE HISTORY
Received 17 August 2021
Revised 27 January 2022
Accepted 19 February 2022

KEYWORDS
Extrudate water solubility; Model; Starch gelatinization; Extrusion; Water solubility index

Introduction

The water solubility of extruded aquafeed is an important parameter for understanding aquatic animal digestion and managing water quality in aquaculture practices. In aquaculture practices, the discharge of soluble contents from aquafeed can lead to eutrophication and destruction of the natural ecosystem in the receiving water body.[1] In recirculating aquaculture systems (RAS), the solubility or leaching of soluble nutrients from aquafeed is an important impact factor for RAS operation.[2,3] In the RAS, the soluble nutrient and dust either dissolved or dropped from the aquafeed directly affect water quality, bio-filter function, and lifespan of the RAS. Thus, understanding aquafeed water solubility and its correlation with aquafeed processing variables is essential in aquaculture practices and aquafeed production.

The extrusion cooking method is the most common way to produce aquafeeds. In the production, recipe formulation and extrusion process variables are the two important aspects of the physical qualities of aquafeed, such as bulk density, hardness, durability, and water solubility.[4] The formulation of aquafeed is often composed of fishmeal, wheat/corn flour, soybean meal, etc., depending on the raw material supply and nutrition requirements for aquatic animals. Thus, many ingredient options may be available for a recipe in terms of a designed nutrition profile for a type of fish. The extrusion process is an experience/skill-based process. It is often difficult to set up a set of optimal extrusion process variables for frequently changing recipes. To address these challenges, it is necessary to search for a quantitative model to describe the effects of the extrusion process variables and the recipe formulations on the physical quality parameters of the aquafeed products, such as water solubility.

CONTACT Hongyuan Cheng chenghongyuan@caas.cn Institute of Feed Research, Chinese Academy of Agricultural Sciences, Beijing, 100081, China and National Food Institute, Technical University of Denmark, Kgs. Lyngby, 2800, Denmark; Jie Yang yangjie02@caas.cn Institute of Feed Research, Chinese Academy of Agricultural Sciences, Beijing, 100081, China

© 2022 Hongyuan Cheng, Hao Wang, Shifeng Ma, Min Xue, Junguo Li and Jie Yang. Published with license by Taylor & Francis Group, LLC.
This is an Open Access article distributed under the terms of the Creative Commons Attribution License (http://creativecommons.org/licenses/by/4.0/), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

The effects of extrusion process variables on extrudate water solubility have been investigated by researchers. Oikonomou and Krokida (2011) collected the literature data for the water absorption index (WAI) and water solubility index (WSI) of different grain products.[5] Later, they proposed an empirical power-law model to correlate the WAI and WSI based on their data collection.[6] The standard deviations of the WSI model prediction are 1.2–27.3 for six products and two mixtures. Ganjyal et al. (2003) illustrated the relationship between WSI and process variables using a neural network method for waxy maize starch extrusion.[7] However, the neural network model requires a large amount of data to train the model coefficients to predict the extrudate water solubility well. Lei et al. (2005) developed an SME-Arrhenius model to correlate the WSI and extrusion variables in a rice flour extrusion process.[8] Independent variables were selected based on statistical significance from the experimental data.[8] Polynomial models, such as response surface methodology (RSM), are often applied to correlate the extrudate water solubility and the extrusion process variables. These models typically have linear and quadratic terms to describe the effects of process parameters on water solubility. In practice, the correct selection of a polynomial model is challenging. The obtained polynomial model is only limited to that particular studied case.[9]

In an extrusion process, recipe nutrients, such as protein, starch, non-starch polysaccharides, fat, fiber, minerals and water, undergo thermal and mechanical treatment (cooking). After cooking, the protein molecules (either soluble or insoluble) change their orientation and network, and are aggregated. The water solubility of protein molecules is often reduced.[10] Meanwhile, starch is gelatinized in water and becomes partly soluble in cool water. At higher extrusion temperatures, the extent of gelatinization leads to an increase in starch water solubility.[11] The water solubility of dietary fiber also increases to some extent.[12,13] The water solubility of fat does not change during the extrusion process. Some vitamins are degraded. The water solubility of processed materials changes based on the solubility of different nutrients. Among these changes, starch gelatinization and starch solubility are the most important. Therefore, we used the starch gelatinization model to estimate the effects of extrusion variables on extrudate water solubility.

This study aimed to estimate the effects of extrusion variables on extruded aquafeed water solubility using a starch gelatinization model. RSM was applied to evaluate the effects of extrusion process parameters (moisture content, screw speed) on the water solubility and WSI of the extruded feed. The starch gelatinization theory and model were considered as the baseline to calculate the extrudate water solubility behaviors under different extrusion process conditions.

Materials And Methods

Recipe formulation

Five commercial aquafeed recipes were used in this study. The ingredients and proximate nutritional information of the recipes are listed in Table 1. To prepare the recipes, five batches (100 kg each) of raw materials for each recipe were mixed for 10 min using a twin-shaft paddle mixer to ensure uniform mixing and finely ground (0.180 mm sieve).

Experimental design

The central composite design was used to investigate the effects of die temperature, moisture content, and screw speed on the water solubility and WSI of the five diets. There were a total of 20 test points, including eight cube points, six star points, and six repeated center points. For the CCD0 diet, the moisture content, screw speed and die temperature all varied between 22–32% (wt), 180–300 rpm, and 80–130°C, respectively. For CCD20 and CCD25 diets, the moisture content, screw speed and the die temperature varied between 22–32% (wt), 190–290 rpm, and 90–140°C, respectively. For the Ma-05 diet, the moisture content was set between 24–32% (wt), the screw speed varied between 190–290 rpm, and the die temperature varied between 110–150°C. For the Ma-01 diet, the moisture content was

Table 1. Recipe formulations and nutrient compositions (%, dry matter).

	Recipe name				
Ingredient, %	CCD0	CCD20	CCD25	Ma-01	Ma-05
Fish meal [1]	0	20	25	30	28
Wheat flour	15	15	15	11.9	5
Soybean meal	54	39	39	0	0
Soybean protein concentrate	18	4.6	0	30	23
Cottonseed protein concentrate	0	6.4	6	8.3	23
Wheat gluten		3	3	5	5
Soybean oil	12	10	10	9	9.5
Additive	1	2	2	0.8	1.5
Cassava				5	5
Total	100	100	100	100	100
Crude protein	38.03	41.21	41.06	48.81	50.90
Crude lipid	13.53	13.09	13.43	12.11	12.84
Crude fiber	22.41	16.87	16.22	6.78	7.01
Starch content	10.70	11.19	11.19	13.05	8.13
Moisture	8.58	8.55	8.69	4.80	4.73
Others	6.75	9.09	9.41	14.45	16.39
Total	100	100	100	100	100

[1] Fish meal: Shandong Chishan Fish meal Factory (Rongcheng, China); Wheat flour: Nankou Flour Mill (Beijing, China); Soybean meal, wheat gluten and soybean oil: Bohai Oil Co., Ltd (Qingdao, China); Soybean protein concentrate: Yihai Kerry Investment Co. Ltd (China); Cottonseed protein concentrate: Xinjiang Jinlan Plant Protein Co. Ltd (China); Cassava: Haid Group Co., Ltd (Guangdong, China).

changed between 20–36% (wt), the screw speed was set between 180–300 rpm, and the die temperature varied between 100–150°C. The processing parameters are listed in Tables 2 and 3. The experiments were conducted in a random order.

Extrusion

The extrusion trials were performed in a twin-screw extruder (MY 56 × 2, Muyang, Yangzhou, China), equipped with a pre-conditioner. The screw diameter of the extruder was 56 mm, and the length to diameter ratio was 20:1. The flow rate of the materials was kept constant at 70 kg/h using a screw feeder. The temperature in the pre-conditioner was greater than 95°C, and the moisture content of the materials was adjusted by adding steam and water. The barrel temperatures in the different zones were controlled by adding either water or steam. Two dies were used in the investigation. The processing parameters were set according to the experimental design shown in Tables 2 and 3. When the processing parameters reached the set-up values and stabilized for 15 min, the samples were collected. Each treatment was repeated three times. The extruded feeds were air-dried at room temperature (25°C) for 48 h until the final moisture content was 4–6% (wt). The samples were stored at 4°C until further analysis.

Water solubility and water solubility index (WSI) of extrudates

The water solubility and WSI of the extruded feeds were determined following the methods of Ma et al. (2021) and Wang et al. (2021).[14,15]

Response surface methodology (RSM)

Experimental data for a central composite design were fit to a second-order polynomial (Eq. 1) by multiple linear regression using the Design Expert (Version 8.0.6, STAT-EASE Inc., Minneapolis, USA). In the models, Y_i is the estimated response, b_0 is the intercept, b_i,b_{ii} and b_{ij} are the regression coefficients for the linear, quadratic and interaction term, respectively, X is the predictor variable and ε is the residual (error).

Table 2. Extrusion process variable and extrudate water solubility measurement results (means ± SEM).

	Recipe name														
	CCD0					CCD20					CCD25				
No.	Td, °C [1]	H_2O, %	Ns, rpm	S, %	WSI, %	Td, °C	H_2O, %	Ns, rpm	S, %	WSI, %	Td, °C	H_2O, %	Ns, rpm	S, %	WSI, %
1	90	24	204	4.40 ± 0.11	14.85 ± 0.22	100	24	210	9.90 ± 1.62	16.38 ± 0.19	130	24	210	7.50 ± 0.33	18.65 ± 0.31
2	90	30	204	4.13 ± 0.12	14.68 ± 0.08	100	30	210	6.09 ± 0.05	16.84 ± 0.12	130	24	270	7.33 ± 0.96	17.70 ± 0.09
3	120	24	204	4.18 ± 0.17	14.87 ± 0.13	100	24	270	11.12 ± 0.17	17.55 ± 0.08	100	24	210	6.28 ± 0.82	19.30 ± 1.13
4	120	30	204	3.29 ± 0.08	14.47 ± 0.09	100	30	270	6.67 ± 0.10	18.17 ± 0.26	130	30	270	9.00 ± 0.28	17.51 ± 0.39
5	90	24	276	4.54 ± 0.15	15.33 ± 0.19	130	24	210	9.51 ± 0.08	16.44 ± 0.17	115	27	240	6.97 ± 0.28	18.34 ± 0.62
6	90	30	276	3.67 ± 0.09	14.91 ± 0.14	130	30	210	6.74 ± 0.25	17.14 ± 0.09	100	24	270	8.32 ± 0.29	18.11 ± 0.83
7	120	24	276	4.85 ± 0.06	15.11 ± 0.16	130	24	270	9.44 ± 1.20	17.81 ± 0.15	100	30	270	7.99 ± 0.18	16.97 ± 0.33
8	120	30	276	3.57 ± 0.13	14.99 ± 0.09	130	30	270	6.89 ± 0.21	18.56 ± 0.12	115	27	240	7.45 ± 0.21	17.48 ± 0.47
9	105	22	240	6.17 ± 0.55	15.19 ± 0.28	115	27	240	7.86 ± 0.15	18.23 ± 0.17	130	30	210	7.72 ± 0.63	16.93 ± 0.09
10	105	32	240	4.51 ± 0.06	15.00 ± 0.08	115	27	240	8.16 ± 0.23	18.60 ± 0.22	115	27	240	6.89 ± 0.19	18.21 ± 0.24
11	80	27	240	3.80 ± 0.09	14.97 ± 0.08	115	27	240	8.02 ± 0.25	19.06 ± 0.46	100	30	210	7.21 ± 0.38	16.69 ± 0.32
12	130	27	240	4.03 ± 0.16	14.87 ± 0.07	115	27	240	8.70 ± 0.42	18.10 ± 0.16	115	27	240	7.53 ± 0.54	17.07 ± 0.35
13	105	27	180	2.89 ± 0.13	14.90 ± 0.10	115	22	240	13.47 ± 1.25	19.39 ± 0.08	115	27	290	7.30 ± 0.76	18.49 ± 0.08
14	105	27	300	3.99 ± 0.09	14.55 ± 0.20	115	32	240	6.94 ± 0.16	16.87 ± 0.07	115	27	240	7.04 ± 0.15	17.38 ± 0.47
15	105	27	240	4.19 ± 0.23	14.85 ± 0.25	115	27	190	7.08 ± 0.33	17.00 ± 0.35	115	22	240	8.43 ± 0.65	18.90 ± 0.57
16	105	27	240	4.21 ± 0.06	14.73 ± 0.12	115	27	290	9.10 ± 0.10	20.21 ± 0.03	140	27	240	8.14 ± 1.12	18.50 ± 0.58
17	105	27	240	4.24 ± 0.24	14.56 ± 0.05	90	27	240	6.81 ± 0.19	16.71 ± 0.18	115	27	190	5.94 ± 0.19	17.91 ± 0.20
18	105	27	240	4.26 ± 0.09	15.01 ± 0.20	140	27	240	7.03 ± 0.11	18.37 ± 0.50	115	27	240	7.00 ± 0.28	18.15 ± 0.41
19	105	27	240	4.41 ± 0.15	15.10 ± 0.08	115	27	240	8.19 ± 0.82	18.25 ± 0.13	115	32	240	7.94 ± 0.38	17.87 ± 0.17
20	105	27	240	4.30 ± 0.12	14.65 ± 0.16	115	27	240	8.49 ± 0.36	18.54 ± 0.22	90	27	240	6.26 ± 0.55	19.14 ± 0.55

[1] Td: die temperature, °C, H_2O: moisture content, % (wt), Ns: screw speed, rpm, S: pellet water solubility, % (wt).
WSI: pellet water solubility index, % (wt).

Table 3. Extrusion process variable and extrudate water solubility measurement results (means ± SEM).

	Recipe name									
	Ma-05					Ma-01				
No.	Td, °C [1]	H_2O, %	Ns, rpm	S, %	WSI, %	Td, °C	H_2O, %	Ns, rpm	S, %	WSI, %
1	118	25.6	210	4.91 ± 0.04	13.03 ± 0.25	110	23	204	3.69 ± 0.07	n.a.[2]
2	142	25.6	210	8.40 ± 0.34	13.28 ± 1.54	140	23	204	3.17 ± 0.12	n.a.
3	118	25.6	270	4.97 ± 0.03	13.58 ± 0.74	110	23	276	3.75 ± 0.08	n.a.
4	142	25.6	270	5.62 ± 0.13	14.08 ± 0.07	140	23	276	3.73 ± 0.05	n.a.
5	118	30.4	210	4.35 ± 0.28	13.45 ± 0.39	110	33	204	3.62 ± 0.08	n.a.
6	142	30.4	210	4.48 ± 0.13	13.94 ± 0.13	140	33	204	3.75 ± 0.05	n.a.
7	118	30.4	270	4.78 ± 0.24	13.70 ± 0.22	110	33	276	4.77 ± 0.07	n.a.
8	142	30.4	270	4.73 ± 0.06	13.99 ± 0.01	140	33	276	3.73 ± 0.05	n.a.
9	130	24	240	4.93 ± 0.17	14.24 ± 0.03	125	20	240	5.35 ± 0.08	n.a.
10	130	32	240	4.59 ± 0.34	14.38 ± 0.07	125	36	240	3.89 ± 0.03	n.a.
11	130	28	190	5.05 ± 0.11	13.58 ± 0.68	125	28	180	3.42 ± 0.05	n.a.
12	130	28	290	5.03 ± 0.09	13.03 ± 0.15	125	28	300	3.76 ± 0.06	n.a.
13	110	28	240	4.44 ± 0.16	13.47 ± 0.26	100	28	240	3.06 ± 0.08	n.a.
14	150	28	240	5.22 ± 0.29	13.72 ± 0.04	150	28	240	3.59 ± 0.02	n.a.
15	130	28	240	5.63 ± 0.16	13.63 ± 0.17	125	28	240	3.81 ± 0.13	n.a.
16	130	28	240	5.75 ± 0.15	13.74 ± 0.13	125	28	240	3.75 ± 0.08	n.a.
17	130	28	240	5.45 ± 0.13	13.72 ± 0.17	125	28	240	4.06 ± 0.03	n.a.
18	130	28	240	5.68 ± 0.11	13.43 ± 0.12	125	28	240	4.07 ± 0.05	n.a.
19	130	28	240	5.45 ± 0.13	13.63 ± 0.13	125	28	240	3.99 ± 0.08	n.a.
20	130	28	240	5.68 ± 0.14	13.72 ± 0.11	125	28	240	3.61 ± 0.02	n.a.

[1] Td: die temperature, °C, H_2O: moisture content, % (wt), Ns: screw speed, rpm, S: pellet water solubility, % (wt), WSI: extrudate water solubility index, % (wt).

[2] n.a.; no available. The WSI data are missing for Ma-01 as they were not measured in the original work.

Table 4. The equations in terms of coded factors and variance analysis results of the regression models (Recipe CCD0, CCD20 and CCD25).

Recipe	CCD0				CCD20				CCD25			
Item	S		WSI		S		WSI		S		WSI	
	CE[a]	*P*-value	CE	*P*-value	CE	*P*-value	CE	*P*-value	CE	*P*-value	CE	*P*-value
Intercept	4.27		14.88		8.24		17.91		7.14		17.97	
Die temperature (X_1)	−0.03	0.5617	−0.04	0.5354	−0.06	0.5628	0.28	0.2269	0.36	0.0289	−0.10	0.5738
Moisture content (X_2)	−0.45	<0.0001	−0.10	0.0879	−1.80	<0.0001	−0.13	0.5802	0.12	0.4076	−0.54	0.0065
Screw speed (X_3)	0.18	0.0093	0.07	0.279	0.39	0.0035	0.78	0.0028	0.46	0.0091	−0.02	0.899
X_1X_2	−0.13	0.1117	-	-	0.37	0.0198	-	-	0.16	0.4023	-	-
X_1X_3	0.16	0.057	-	-	−0.21	0.1362	-	-	−0.21	0.2732	-	-
X_2X_3	−0.12	0.1246	-	-	−0.05	0.7006	-	-	0.02	0.9001	-	-
X 2 1	−0.16	0.0173	-	-	−0.49	0.0006	-	-	0.09	0.5413	-	-
X 2 2	0.35	<0.0001	-	-	0.67	<0.0001	-	-	0.44	0.01	-	-
X 2 3	−0.32	0.0002	-	-	−0.08	0.4528	-	-	−0.12	0.4097	-	-
Model[b]		<0.0001				<0.0001		0.0144		0.0323		0.0443
Lack of Fit[c]	0.0070			0.5172	0.2390			0.0188	0.0316			0.2943
Model R-squared[d]	0.9460			0.2366	0.9766			0.4733	0.7584			0.3880
Adeq Precision Precision[e]	20.028			4.322	28.165			7.190	6.877			6.370

[a] Coefficient Estimate of independent variables in the final equation.

[b] The fitness of the model was evaluated and the interactions between the independent and dependent variables were identified by variance analysis ($P > 0.05$).

[c] Lack of Fit: This is the variation of the data around the fitted model. If the model does not fit the data well, the test will show significant.

[d] Measure of the amount of variation around the mean explained by the model.

[e] Adeq Precision measures the signal to noise ratio. A ratio greater than 4 is desirable.

$$Y_i = b_0 + \sum_{i=1}^{n} b_i X_i + \sum_{i=1}^{n} b_{ii} X_{ii}^2 + \sum_{i=1}^{n} \sum_{j=i+1}^{n} b_{ij} X_i X_j + \varepsilon \tag{1}$$

Water solubility of extrudates

In an extrusion cooking process, the changes in water solubility for a processed recipe are determined by the recipe ingredients and extrusion process variables. In terms of the recipe, the water solubility changes of the recipe are the sum of the contributions of each ingredient or nutrient: protein, starch, non-starch polysaccharides, minerals, fat, and ash. In this process, the water solubility of the mixed materials is modified by thermal and mechanical energy, especially for proteins, starch, fiber, and vitamins. Native starch granules are insoluble in cold water. After cooking or heating with excess water, the starch granules start to absorb water, swell, and gelatinize.[16] Subsequently, gelatinized starch is partly soluble in water. Meanwhile, protein molecules aggregate and form a new network. Thus, the water solubility of the protein decreased after extrusion.[10,17] The amount of insoluble dietary fiber in a recipe also decreases after extrusion. Thus, the water solubility of the extrudate is a combination of the solubility of protein, starch, and fiber. To simplify the description of the solubility development of the three nutrients individually, we assume that the overall solubility development of the processed recipe can be represented by starch gelatinization development. The theory and model of starch gelatinization represent the water solubility development of raw materials during the extrusion process; the water solubility development of a recipe in an extrusion process can be approximated by a starch gelatinization model.

Cai and Diosady (1993) investigated the kinetics of wheat starch gelatinization and proposed a pseudo-first-order rate equation to predict the degree of starch gelatinization in an extrusion process.[18] Using the starch gelatinization degree equation, Cai and Diosady (1993) developed another equation to correlate the WSI of extrudates.[19] In their work, the WSI was related to the residence time, mechanical and thermal energy input in an extrusion process through experimental data regression analysis. From the chemical reaction concept, we may develop a model to represent the water solubility of processed raw materials in an extrusion process.

Water solubility model

Based on the work of Cai and Diosady (1993)[18,19] and the assumptions discussed above, a new equation was derived as follows. The rate of disappearance of ungelatinized starch is described as:

$$\frac{d(1-f)}{dt} = -k_g(1-f)^m \tag{2}$$

where f is the degree of gelatinization, t is the residence time of the raw materials in the extrusion process, k_g is the gelatinization rate constant, and m is the reaction order. Following the suggestion of Cai and Diosady (1993),[18] we assume that starch gelatinization in the aquafeed extrusion process is a pseudo-first-order reaction. By setting $m = 1$, when $t = 0, f = 0$, and $t = t, f = f$, Eq. (2) is derived as follows:

$$ln(1-f) = -k_g t \tag{3}$$

$$k_g = k_0 exp\left(\frac{-\Delta E}{RT}\right) \tag{4}$$

where ΔE is the thermal activation energy (kJ/mol), T is the temperature (K), R is the ideal gas constant. During the extrusion process, starch gelatinization occurs in a shearing environment. Cai and Diosady (1993) considered the effect of shear stress on gelatinization and proposed a method to calculate the thermal activation energy that takes into account both thermal and shear stress effects.[18] Using this principle, a relationship between the extrudate water solubility and extrusion process variables can be established. The solubility degree (S_d) is defined as:

$$S_d = \frac{S_0 - S}{S_0} \tag{5}$$

where S_0 is the solubility of the raw materials before extrusion cooking, and S is the solubility after extrusion cooking. By replacing the degree of gelatinization f with S_d in Eq. (2)–(3), we have:

$$\frac{d(1 - S_d)}{dt} = -k_g(1 - S_d) \tag{6}$$

$$ln(1 - S_d) = -k_g t \tag{7}$$

Replacing the S_d in Eq. (7) with Eq. (5), we obtain:

$$ln(S) = -k_g t + ln(S_0) \tag{8}$$

Eq. (8) can be used to correlate the extrudate water solubility and the extrusion process variables. However, we developed a method for calculating the initial water solubility (S_0) of a recipe, gelatinization constant k_g, and time t. S_0 is the initial solubility of the raw materials before extrusion cooking. The main contribution to S_0 is the soluble nutrients (protein, minerals, and non-starch polysaccharides) in a recipe. In practice, S_0 can be measured using a specific recipe. Subsequently, the value of S_0 can be estimated. Because the initial solubility, S_0 is a constant for a specific recipe in an extrusion process, and thus it does not influence the calculation of solubility development in the extrusion process. Therefore, the value of S_0 does not change the basic characteristics of the equation in terms of the description of the solubility development of a recipe during extrusion. In the application of the equation, the initial solubility measurement (S_0) must be measured for every specific recipe. An alternative method is to estimate the initial solubility (S_0) from the typical solubility values of various ingredients. However, the water solubility of a specific ingredient may have different values, as it may come from different agricultural areas or environmental conditions. As can be seen, equation (8) can maintain its basic behavior for solubility development calculation if S_0 is removed from the equation. To avoid the initial solubility measurements for every recipe and estimation errors for the initial water solubility of a recipe, S_0 is neglected from the equation. Thus, we obtain:

$$lnS = -k_g t \tag{9}$$

where S is the extrudate water solubility. In the calculation of the gelatinization constant k_g, Cai and Diosady (1993) split the thermal activation energy (ΔE) into thermal and mechanical contribution terms and developed methods to estimate them.[18] In this study, we do not split the thermal activation energy (ΔE) into thermal and mechanical contribution terms. The thermal activation energy effect was calculated using the classical Arrhenius equation $k = Ae^{E/RT}$. In addition, the moisture content contribution or effect is added to calculate the k_g because starch gelatinization is not an ideal first-order chemical reaction process. Therefore, the k_g is calculated as:

$$k_g = k_f exp\left(\frac{\Delta E}{RT} + k_1 M\right) \tag{10}$$

where M is the moisture or water content in an extrusion process (kg/kg), T is the die temperature (K), and k_f and k_1 are the model coefficients. Furthermore, the residence time (t) in Eq. (9) is estimated as follows:

$$t = k_t \left(\frac{1}{N_s}\right)^{\alpha} \tag{11}$$

where N_s is the screw speed of an extruder, rpm

Set $k_0 = k_f k_t$ then

$$lnS = -k_0 N_s^{-\alpha} exp\left(\frac{\Delta E}{RT} + k_1 M\right) \tag{12}$$

where S represents the extrudate water solubility (kg/kg), N_s is the screw speed of an extruder (rpm), ΔE is the thermal activation energy (kJ/mol), T is the die temperature (K), R is the ideal gas constant, M is the moisture content in an extrusion process (kg/kg), and k_0, α, ΔE, k_1 are the experimental determined parameters. As shown in Eq. (12), the extrudate water solubility is an interaction of three parameters: shear stress (N_s), thermal-mechanical input ($\Delta E/RT$) and moisture content (M). The WSI of a sample has a similar measurement to the water solubility, but a different value for the same sample. Therefore, we suggest that Eq. (12) can also be used to predict the WSI of extruded pellets.

Results

Water solubility and WSI measurement results

The experimental results of water solubility and WSI and the corresponding extrusion process variables for the five studied recipes are given in Table 2–3. The water solubility values of the five studied diets vary from 2.89–13.47% and the values of WSI data range from 13.03–18.65%. The extrusion process has an obvious impact on the water solubility of the extrudates for all the five recipes that were studied.

RSM model results

The RSM model could well fit the water solubility of the feeds, except for the Ma-01 diet (Table 4–5). The processing parameters had significant effects on the water solubility ($P < .05$). However, the RSM could not well correlate the WSI of the feeds (Table 4–5).

Table 5. The equations in terms of coded factors and variance analysis results of the regression models (Recipe Ma-01 and Ma-05).

Recipe	Ma-01		Ma-05			
Item	S		S		WSI	
	CE[a]	*P*-value	CE	*P*-value	CE	*P*-value
Intercept	3.89		5.12		13.61	
Die temperature (X_1)	−0.04	0.7728	0.41	0.0386	0.14	0.0836
Moisture content (X_2)	−0.07	0.6324	−0.45	0.0256	0.10	0.2175
Screw speed (X_3)	0.17	0.2402	−0.15	0.3834	0.05	0.4865
X_1X_2	−0.05	0.8028	−0.51	0.0451	0.00	0.9820
X_1X_3	−0.08	0.6481	−0.38	0.1162	0.01	0.9532
X_2X_3	0.07	0.7219	0.43	0.0820	−0.13	0.2094
X_1^2	−0.21	0.1425	-	-	−0.03	0.7273
X_2^2	0.25	0.0947	-	-	0.23	0.0118
X_3^2	−0.12	0.3956	-	-	−0.13	0.1072
Model[b]	0.4429			0.0270		0.0942
Lack of Fit[c]	0.0242		0.0555		0.0511	
Model R-squared[d]	0.5246		0.7723		0.7143	
Adeq Precision Precision[e]	3.978		8.313		6.218	

[a] Coefficient Estimate of independent variables in the final equation.

[b] The fitness of the model was evaluated and the interactions between the independent and dependent variables were identified by variance analysis ($P > 0.05$).

[c] Lack of Fit: This is the variation of the data around the fitted model. If the model does not fit the data well, the test will show significant.

[d] Measure of the amount of variation around the mean explained by the model.

[e] Adeq Precision measures the signal to noise ratio. A ratio greater than 4 is desirable.

Model regression with experimental data

Eq. (12) was used to fit the experimental water solubility and WSI data of different recipes. A Scipy optimization library in a free and open-source Python programming language (Python software foundation) was used to carry out the regression. In the regression, the objective function was set as

$$\text{obj.} = min\left(\sum_{i=1}^{n}\left(\frac{S_i^{cal} - S_i^{exp}}{S_i^{exp}}\right)^2\right) \tag{13}$$

where obj. is the objective function of the regression, S_i is the extrudate water solubility or the WSI, superscripts “exp” and “cal” represent the experimental and calculated values, respectively, n is the number of experimental runs. In the regression, the model coefficients were determined individually for each recipe. The absolute average deviations of the regression for the five recipes are presented in Table 6. The model coefficients are listed in Table 7–8. Comparisons between the experimental data and the fitted solubility data are shown in Fig. 1–2. In Table 6, the absolute average deviation AAD% is calculated as

$$\text{AAD\%} = \frac{1}{n}\sum_{i=1}^{n}\frac{\left|S_i^{exp} - S_i^{cal}\right|}{S_i^{exp}} \tag{14}$$

where S_i is the extrudate water solubility or the WSI, superscripts “exp” and “cal” represent the experimental and calculated values, respectively, n is the number of experimental runs. The AAD % values of the water solubility data were all less than 10% for the studied 5 recipes (Table 6). The AAD % values of the WSIs were all less than 5%. These AAD % values can be used in engineering applications.

Table 6. Absolute average deviation (AAD%) of experimental data fitting with Eq. (12).

No.	Recipe name	AAD% of fitting water solubility data	AAD% of fitting WSI
1	CCD0	8.22	1.04
2	CCD20	6.97	3.36
3	CCD25	6.59	2.81
4	Ma-01	9.41	NA*
5	Ma-05	8.68	1.74
Average		7.97	2.24

*NA: not available

Table 7. Model coefficients obtained from experimental water solubility data regression.

	Recipe name				
Coefficient	CCD0	CCD20	CCD25	Ma-01	Ma-05
k_0	4.64	3.65	5.00	−0.04	0.58
α	−0.11	−0.15	−0.20	−0.02	0.07
ΔE, kJ/mol	−27.56	−105.95	186.61	6.57	359.00
k_1	1.05	2.72	−0.19	3.55	1.23
obj.	0.03	0.03	0.02	0.25	0.03

Table 8. Model coefficients obtained from experimental WSI data regression.

	Recipe name			
Coefficient	CCD0	CCD20	CCD25	Ma-05
k_0	2.08	3.91	1.57	1.97
α	−0.02	−0.20	0.00	−0.02
ΔE, kJ/mol	−12.21	98.41	−36.02	70.54
k_1	0.12	0.15	0.58	−0.29
obj.	0.00	0.01	0.01	0.00

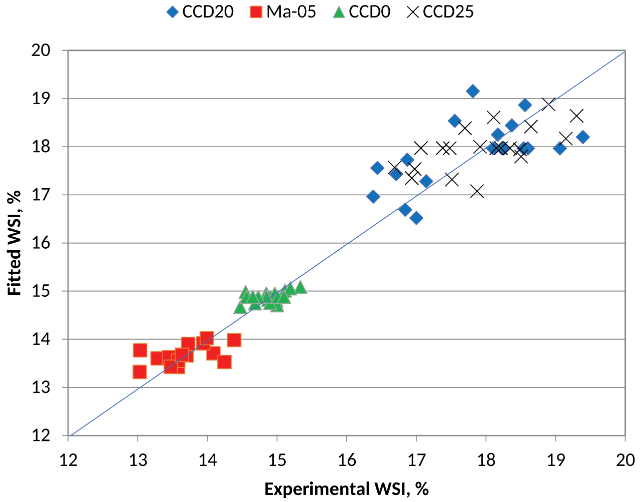

Figure 1. Comparison of experimental and fitted WSI data for 4 aquafeed recipes. The WSI data are missing for Ma-01 as they were not measured in the original work.

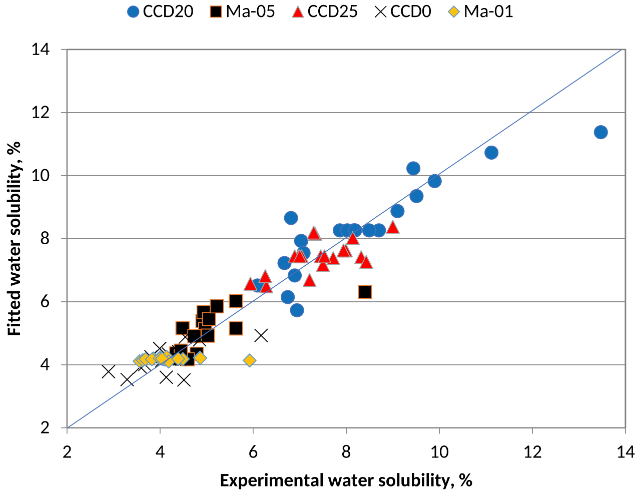

Figure 2. Comparison of experimental and fitted water solubility data for 5 aquafeed recipes.

Fig. 1–2 shows that the deviations of the fitting are evenly distributed. The proposed Eq. (12) can satisfactorily predict either the extrudate water solubility or the WSI data for the studied samples. Observing the water solubility value distributions in Figure 1, we can see that the WSI data are split into group 1: recipe CCD0, group 2: recipe Ma-05, and group 3: recipe CCD20 and CCD25, which are mainly caused by the different formulations.

Effects of extrusion process variables on extrudate water solubility and WSI

Based on Eq. (12), the effects of the extrusion process variables on the extrudate water solubility can be numerically investigated for the five recipes. In this investigation, we only selected recipes CCD20 and Ma-05 to demonstrate the calculation because the two recipes represent the lower and the higher water solubility limits in the five diets. The study results are presented in Fig. 3–6. Other recipes can also be studied using the reported model coefficients in Table 7–8. The pellet water solubility and WSI values increased with an increase in screw speed (Fig. 3–6). The effects of water content (X_w) changes were calculated by the model at three different values, i.e., $X_w = 0.20, 0.25, 0.30$ kg/kg, respectively, which are marked at the left corner in Fig. 3–6. As shown in Fig. 3–5, when X_w increases from 0.2 to 0.3, the extrudate water solubility and WSI decrease. The calculation results show that when X_w increased from 0.2 to 0.3, the WSI increased in recipe Ma-05. For the recipe CCD20, an increase in the extrusion process temperature caused a decrease in the pellet water solubility (Figure 3) and an increase in the WSI (Figure 4). For recipe Ma-05, the extrudate water solubility and WSI (Fig. 5–6) increase with an increase in the process temperature.

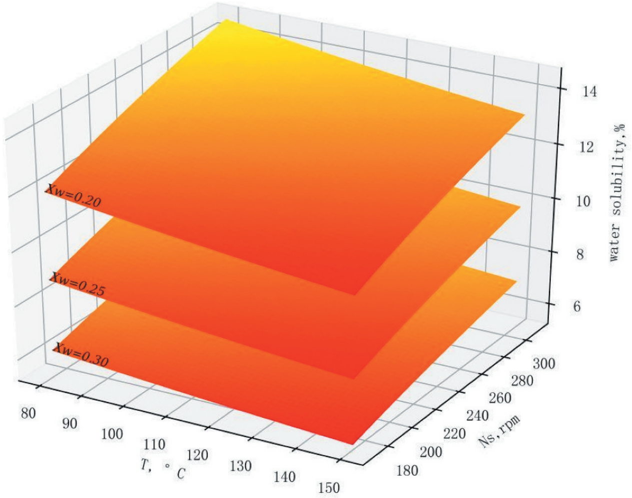

Figure 3. Effects of extrusion process variables on extrudate water solubility, recipe CCD20. T: die temperature, °C, N_s: screw speed, rpm, X_w: moisture content, kg/kg.

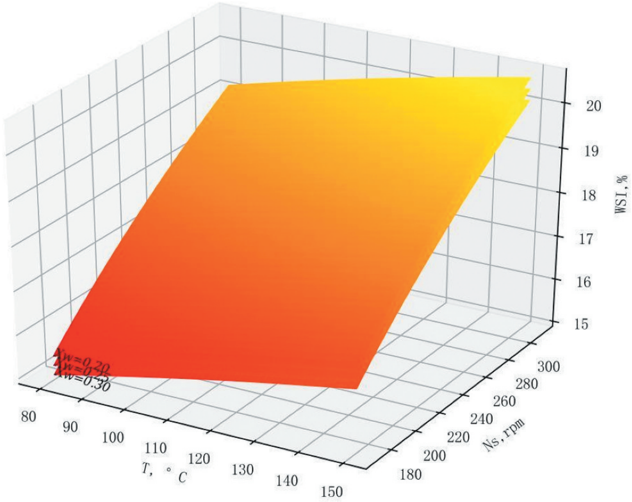

Figure 4. Effects of extrusion process variables on extrudate WSI, recipe CCD20. T: die temperature, °C, N_s: screw speed, rpm, X_w: moisture content, kg/kg.

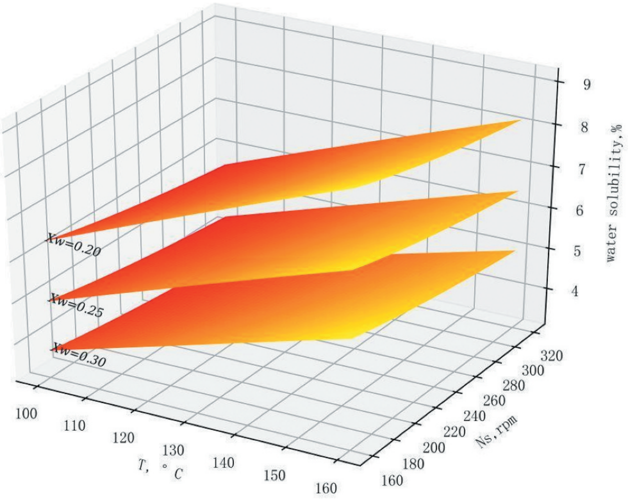

Figure 5. Effects of extrusion process variables on extrudate water solubility, recipe Ma-05. T: die temperature, °C, N_s: screw speed, rpm, X_w: moisture content, kg/kg.

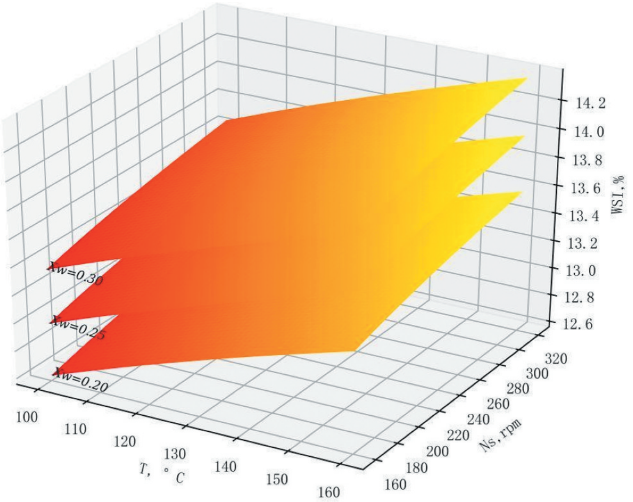

Figure 6. Effects of extrusion process variables on extrudate WSI, recipe Ma-05. T: die temperature, °C, N_s: screw speed, rpm, X_w: moisture content, kg/kg.

Discussion

In the aquafeed extrusion process, the recipe characteristics and extrusion process variables are the two impact factors on the variations of aquafeed pellet properties. In this study, we could not set a direct correlation between the data groups and the recipe ingredients and their content information. Through the recipe viscous property, we may find a relationship between the extrudate water solubility and the recipe characteristics in future investigations.

In this study, the pellet water solubility and WSI values of the two studied recipes increased with an increase in screw speed. A high screw speed could increase the heat input through viscous dissipation, thereby increasing starch gelatinization, further increasing the water solubility and WSI.[20] We observed that the water solubility of the feeds decreased with the increase of moisture content. Consistent with a previous study, Umar et al. (2013)[21] reported that the water solubility of extruded aquafeed decreased with an increase in moisture content from 20% to 40%. As water is a solvent, and the water-soluble components in the material can act as a binder to improve the water stability of feed. However, when X_w increased from 0.2 to 0.3, the WSI increased in recipe Ma-05 in this study, which may be due to the higher dextrinization of the starch.[22] From the experimental data in Table 3, it can also be seen that when the water content increased from 28% (No. 13) to 32% (No. 10), the WSI increased from 13.47% to 14.38% for recipe Ma-05. The calculated WSI development for Ma-05 was mostly consistent with the trend of the experimental observations. The effects of temperature on the pellet water solubility and WSI were different for the recipes. The extrudate water solubility and WSI in Ma-05 increase with the die temperature. Ding et al. (2005)[23] also found that soluble starch increased with an increase in extrusion temperature, which resulted from the increased degree of starch gelatinization. Thus, the increased WSI and water solubility with the increase in die temperature in this study were due to the improved starch gelatinization. However, the extrudate water solubility in CCD20 decreased with the increase in extrusion temperature, which may be related to the denser structure and resistance to immersion in water.

The model in Eq. (12) is built from the chemical reaction kinetics theory based on the degree of starch gelatinization. Based on this development, the model builds its physical and chemical foundation and is independent of the studied extrusion process, recipes, and extruder mechanical geometric conditions. We observed that the model can fit the experimental water solubility of the five different recipes without changing its model expression, which was more reliable than RSM model. The model prediction results may be applied to explain the extrudate water solubility behaviors in different extrusion processes for the same or similar recipes. The proposed model may also be extended to predict the water solubility of other starch-based extrudates, such as breakfast extrudates. However, the water solubility of breakfast extrudates may not be critical from a consumer point of view. In the model development, the effects of starch differences (e.g., amylose and amylopectin ratio) on the trail results were not investigated. This may limit the model to computed water solubility in different cases.

Conclusion

A new model was developed to estimate aquafeed water solubility based on the starch gelatinization degree concept and a quantitative equation for its development in extrusion process. The results show that the established model can estimate the extrudate water solubility and WSI. In the experimental data regression, the total average AAD % values for the five studied recipes were 7.97% for the extrudate water solubility and 2.24% for the extrudate WSI. The effects of the extrusion process variables on extrudate water solubility and WSI were investigated. It was found that the extrudate water solubility and WSI tended to increase with an increase in the extrusion process screw speed. The new model can estimate the water solubility without changing its mathematical formulation. The model expression does not need to be changed when it is used to calculate the water solubility of different recipes in different trials. Thus, the mathematical format of the model is independent of recipe formulations.

Acknowledgments

This study was supported by the National Key R&D Program of China (2019YFD0900200 and 2018YFD0900400); National Natural Science Foundation of China (32172981 and 31902382); The Agricultural Science and Technology Innovation Program of CAAS, China (CAAS-ASTIP-2017-FRI-08); China Postdoctoral Science Foundation (2021M703544); Nordforsk project number (104310) and Norwegian Research council (327109).

The authors declare that they have no conflict of interest.

Disclosure statement

No potential conflict of interest was reported by the author(s).

References

[1] Verdegem, M. C. J. Nutrient Discharge from Aquaculture Operations in Function of System Design and Production Environment. *Rev. Aquac.* 2013, *53*, 158–171. DOI:10.1111/raq.12011.

[2] Schumann, M.; Brinker, A. Understanding and Managing Suspended Solids in Intensive Salmonid Aquaculture: A Review. *Rev. Aquac.* 2020, *124*, 2109–2139. DOI:10.1111/raq.12425.

[3] Kong, W.; Huang, S.; Yang, Z.; Shi, F.; Feng, Y.; Khatoon, Z. Fish Feed Quality Is a Key Factor in Impacting Aquaculture Water Environment: Evidence from Incubator Experiments. *Sci. Rep.* 2020, *10*(1), 187. DOI: 10.1038/s41598-019-57063-w.

[4] Sørensen, M. A Review of the Effects of Ingredient Composition and Processing Conditions on the Physical Qualities of Extruded High-energy Fish Feed as Measured by Prevailing Methods. *Aquac. Nutr.* 2012, *183*, 233–248. DOI:10.1111/j.1365-2095.2011.00924.x.

[5] Oikonomou, N. A.; Krokida, M. K. Literature Data Compilation of WAI and WSI of Extrudate Food Products. *Int. J. Food Prop.* 2011, *14*(1), 199–240. DOI: 10.1080/10942910903160422.

[6] Oikonomou, N. A.; Krokida, M. K. Water Absorption Index and Water Solubility Index Prediction for Extruded Food Products. *Int. J. Food Prop.* 2012, *15*(1), 157–168. DOI: 10.1080/10942911003754718.

[7] Ganjyal, G. M.; Hanna, M. A.; Jones, D. D. Modeling Selected Properties of Extruded Waxy Maize Cross-Linked Starches with Neural Networks. *J. Food Sci.* 2003, *68*(4), 1384–1388. DOI: 10.1111/j.1365-2621.2003.tb09654.x.

[8] Lei, H.; Fulcher, R. G.; Ruan, R.; van Lengerich, B. SME-Arrhenius Model for WSI of Rice Flour in a Twin-Screw Extruder. *Cereal Chem. J.* 2005, *825*, 574–581. DOI:10.1094/CC-82-0574.

[9] Yolmeh, M.; Jafari, S. M. Applications of Response Surface Methodology in the Food Industry Processes. *Food Bioprocess Technol.* 2017, *10*(3), 413–433. DOI: 10.1007/s11947-016-1855-2.

[10] Lin, D.; Lu, W.; Kelly, A. L.; Zhang, L.; Zheng, B.; Miao, S. Interactions of Vegetable Proteins with Other Polymers: Structure-Function Relationships and Applications in the Food Industry. *Trends Food Sci. Technol.* 2017, *68*, 130–144. DOI: 10.1016/j.tifs.2017.08.006.

[11] Ali, S.; Singh, B.; Sharma, S. Effect of Processing Temperature on Morphology, Crystallinity, Functional Properties, and in Vitro Digestibility of Extruded Corn and Potato Starches. *J. Food Process. Preserv.* 2020, *44* (7). DOI: 10.1111/jfpp.14531.

[12] Honců, I.; Sluková, M.; Vaculová, K.; Sedláčková, I.; Wiege, B.; Fehling, E. The Effects of Extrusion on the Content and Properties of Dietary Fibre Components in Various Barley Cultivars. *J. Cereal Sci.* 2016, *68*, 132–139. DOI: 10.1016/j.jcs.2016.01.012.

[13] Naumann, S.; Schweiggert-Weisz, U.; Martin, A.; Schuster, M.; Eisner, P. Effects of Extrusion Processing on the Physiochemical and Functional Properties of Lupin Kernel Fibre. *Food Hydrocoll.* 2021, *111*, 106222. DOI: 10.1016/j.foodhyd.2020.106222.

[14] Ma, S.; Wang, H.; Li, J.; Xue, M.; Cheng, H.; Qin, Y.; Blecker, C. Effect of the Ratio of Wheat Flour and Cassava and Process Parameters on the Pellet Qualities in Low Starch Feed Recipe Extrusion. *Anim. Feed Sci. Technol.* 2021, *271*, 114714. DOI: 10.1016/j.anifeedsci.2020.114714.

[15] Wang, H.; Ma, S.; Yang, J.; Qin, Y.; Cheng, H.; Xue, M.; Li, J.; Li, J. Optimization of the Process Parameters for Extruded Commercial Sinking Fish Feed with Mixed Plant Protein Sources. *J. Food Process. Eng.* 2021, *44*(1), e13599. DOI: 10.1111/jfpe.13599.

[16] Xiao, J.; Zhong, Q. Suppression of Retrogradation of Gelatinized Rice Starch by Anti-Listerial Grass Carp Protein Hydrolysate. *Food Hydrocoll.* 2017, *72*, 338–345. DOI: 10.1016/j.foodhyd.2017.06.016.

[17] Guy, R.;, Ed. Extrusion Cooking Technologies and Applications. *CRC Press.*, 2001.

[18] Cai, W.; Diosady, L. L. Model for Gelatinization of Wheat Starch in a Twin-Screw Extruder. *J. Food Sci.* 1993, *58* (4), 872–875. DOI: 10.1111/j.1365-2621.1993.tb09380.x.

[19] Cai, W.; Diosady, L. L. Modeling of Expansion and Water Solubility Index of Wheat-Starch during Extrusion-Cooking. *Acta Aliment.* 1993, *22*, 181–192.

[20] Guha, M.; Ali, S. Z.; Bhattacharya, S. Twin-Screw Extrusion of Rice Flour without a Die: Effect of Barrel Temperature and Screw Speed on Extrusion and Extrudate Characteristics. *J. Food Eng.* 1997, *32*(3), 251–267. DOI: 10.1016/S0260-8774(97)00028-9.

[21] Umar, S.; Kamarudin, M. S.; Ramezani-Fard, E. Physical Properties of Extruded Aquafeed with a Combination of Sago and Tapioca Starches at Different Moisture Contents. *Anim. Feed Sci. Technol.* 2013, *183*(1–2), 51–55. DOI: 10.1016/j.anifeedsci.2013.03.009.

[22] Williams, M. A.; Horn, R. E., and Rugala, R. P. Extrusion: An in-Depth Look at a Versatile Process. II.[Cooking Process and Its Effect upon Foods and Additives]. *Food. Eng.* 1977, *49*(10), 87–89.

[23] Ding, Q.-B.; Ainsworth, P.; Tucker, G.; Marson, H. The Effect of Extrusion Conditions on the Physicochemical Properties and Sensory Characteristics of Rice-Based Expanded Snacks. *J. Food Eng.* 2005, *66*(3), 283–289. DOI: 10.1016/j.jfoodeng.2004.03.019.