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Bio-versus Chemical Approaches to Produce Maltodextrin (de 9 – 12) for Potential Applications in Functional Foods and Pharmaceuticals

Tam Minh Le*, Nhung Thi Tran
Ho Chi Minh City University of Technology and Education, Vietnam

*Corresponding author. *Email: tamlm@hcmute.edu.vn*

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ABSTRACT

Maltodextrin is recognized as a food and pharmaceutical additive that is safe for direct human consumption. The physical and chemical properties of maltodextrin strongly depend on its DE (dextrose equivalent) index. Maltodextrin is a hydrolyzed product derived from starch that has gained numerous industrial applications since the last few decades. Currently, there is an increasing demand for modified starch products. Unfortunately, the relevant production process remains inefficient, leading to relatively low product quality and performance. This paper reports the results of parameterized production processes of maltodextrin with DE values ranging from 9 to 12, achieved by hydrolyzing cassava starch. Two approaches were conducted in this work, including hydrolysis with the catalysis of α-amylase enzyme and HCl acid. In the α-amylase enzyme method, various factors were investigated, such as starch content, hydrolysis time, enzyme concentration, and hydrolysis temperature. In the HCl acid method, process parameters were studied, including starch content, HCl acid concentration, reaction time, and temperature. For both approaches, the DE index of maltodextrin was selected as the objective function, and it was found to be influenced by several process conditions. Utilizing a full Design of Experiment (DoE) plan, a regression equation was developed to illustrate the influence of these factors. From the regression equation, the optimal conditions for the production of desired maltodextrin were derived and compared between the two hydrolysis methods.

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1. Introduction

Starches have a particularly significant nutritional role because, during the digestive process, they are hydrolyzed into glucose. This compound serves as the primary source of calories for humans. Starch is essential in the food industry owing to its physicochemical properties. It is frequently employed as a viscosity agent for liquid foods, a stabilizing agent for colloidal foods, and a binding and thickening agent that imparts hardness and elasticity to various food products [1], [2]. Additionally, starch powder is widely applied in treating wastewater, creating hydrophobic coatings in the production of emulsion explosives, and serving as a binder in paint technology [3], [4]. Modified starch has been developed since the early 19th century, and these modifications contribute to enhancing the value of starch [5]–[7].

An important process of starch is its hydrolysis by either acids or enzymes. Acids can hydrolyze starch in its original granular form or in its gelatinized or paste form, whereas enzymes can effectively hydrolyze starch only when it is in the gelatinized form. Both acids and enzymes share the similarity of breaking down starch molecules by cleaving the α-D-(1,4) glycoside bond. This reaction is characterized by a rapid decrease in viscosity and the production of sugar. Modified starch is also referred to as transformed starch. Natural starch is subjected to denaturing methods such as physical, enzymatic, or chemical processes to alter its physical and chemical properties [1]–[4]. In today's food technology, modified starch finds widespread use as a co-gelling agent, thickener, stabilizer, or emulsifier. In the pharmaceutical field, modified starch serves as a drug carrier, and it has applications in other industries [6], [7]. Currently, there is a growing demand for modified starch products, including dextrin, syrup, and especially maltodextrin with a DE index ranging from 9 to 12. Researching and optimizing the

starch modification process to meet the increasing market demand at a competitive price is of utmost importance.

Starch modification methods can be classified into three categories: physical, chemical, and enzymatic transformations. In the first method, which involves the use of acidic agents, certain bonds between starch molecules and within starch molecules are broken. This process results in a reduction in molecular size and yields starch with new properties. In industrial production, starch is typically dispersed in an inorganic acid solution with a concentration of 1–3%. The mixture is then thoroughly stirred at a temperature of 50–55 °C for 12–14 hours, followed by neutralization, filtration, and drying. Acid-modified starch exhibits several distinctive properties compared to its original form, including reduced affinity for iodine, lower specific viscosity, and higher osmotic pressure due to its decreased mass. The average molecule size is smaller, and when gelatinized in hot water, the particles swell less. In warm water with a temperature below the gelatinization temperature, its solubility is higher, the gelatinization temperature is increased, and the alkalinity index is also elevated [7]–[10]. In the second method involving enzymes, starch molecules are randomly broken into low-molecular dextrins or gradually shortened into two glucose units, or maltose is released. However, this enzymatic process also alters the properties of the starch solution. Among enzymes, amylase is a physiologically significant enzyme and is commonly known by its trade name, i.e. diastase. Amylase is present in both plants and animals and belongs to the group of hydrolytic enzymes, catalyzing the breakdown of intramolecular glucoside bonds in polysaccharides with the involvement of water. When amylase hydrolyzes starch, it can produce a crucial product, maltodextrin [11], [12].

According to the definition of the US Food and Drug Administration (FDA), maltodextrins are polysaccharides consisting of α-D-glucose radicals linked together by 1-4 glycosidic bonds. They are products of incomplete starch hydrolysis (by enzymes or acids), with dextrose equivalents (DE) ranging from 4 to 20 [13]. The properties of maltodextrin depend on the DE index. The product may exist in the form of a white powder or a concentrated solution. Maltodextrin has a simple sugar structure, which stimulates appetite, facilitates digestion, and delivers nutrients without affecting blood sugar balance. Thanks to these properties, maltodextrin finds extensive use in food products designed for children, the elderly, and the ill. It is even added to cow's milk to replace lactose. Maltodextrin is used in the production of fresh cakes, gum candy, marshmallows, and snack cakes. It serves the purpose of stabilizing the product shape, preserving flavor, and preventing candy from melting. In bread production, it helps prevent tooth decay and is suitable for individuals with gum disease, blood pressure concerns, and diabetes [14], [15]. Additionally, maltodextrin is employed in the production of numerous beverages [16]. It acts as an essential oil carrier, stabilizer, and flavor preserver in the production of items like chocolate and coffee. Herbal extracts are concentrated and mixed with spray-dried maltodextrin to create a fine powder, forming a membrane to preserve the fruit. Notably, maltodextrin is widely used in the pharmaceutical industry. It can be found in dietary plans, serving as a tonic and a food supplement to enhance drug solubility, aiding patient absorption. In pill production, maltodextrin functions as an excipient for tablet filling and as a capsule membrane. The addition of maltodextrin protects enzymes from reacting with the substrate or being oxidized [16]. In summary, maltodextrin has a wide range of applications in functional foods and pharmaceuticals depending on its DE index. In this work, we conduct hydrolysis of cassava starch to produce maltodextrin with DE in a range of 9–12. The study encompasses hydrolysis conducted through both acidic and enzymatic pathways. The results provide comprehensively understand between the advantages and disadvantages between the two applied methods in production of a special range DE of maltodextrin.

2. Materials and Methods

2.1. Materials

Tapioca starch has the trade name of tapioca starch from Tai Ky Food Flour Joint Stock Company (TAKYFOOD). Tapioca starch has a moisture content of 12%. It was stored at room temperature and was kept away from direct sunlight. Besides, other chemical reagents were also used including $CuSO_4$ (99%, Xinglong), D-glucose (Analytical reagent, Quangdong Sci-Tech Co), HCl (32%, Technology Shanghai Limited), and NaOH (99%, Shangdong).

2.2. Methods

In the screening step, only one parameter was alternatively changed while the rest were kept constant at common levels. The studied parameters included catalyst concentration, starch content, reaction time, and temperature. For example, in the HCl hydrolysis method, to investigate the influence of acid concentration, experiments were conducted with acid concentrations of 2%, 4%, 6%, 8%, 10%, and 12%. This involved mixing 20g of cassava starch with 40ml of HCl at the mentioned concentrations in an Erlenmeyer flask and placing the reactor in a 55 °C for 6 hours. The solution was filtered to measure dry matter concentration, viscosity, and DE (dextrose equivalent) index. During the hydrolysis process, it was essential to regularly stir the solution to prevent starch from settling at the bottom, which could lead to inaccurate results. Similar procedure was applied for other parameter investigation. Likewise, enzyme was replaced for HCl in enzymatic hydrolysis of which particular levels of investigated factors will be shown in the experimental results. The common levels for both methods were summarized as follows:

- Weight of cassava starch: 40 g.
- Powder/water ratio is ¼.
- pH= 6.5÷7.0.
- Added suitable amount of $CaCl_2$ solution.
- Enzyme concentration 0.1% in enzymatic hydrolysis (or HCl concentration at 2%).
- Performing gelatinization at 75 ^oC for 30 minutes.
- Reaction time: 10 minutes in enzymatic method (or 4 hours in HCl method).

The measurements include the determination of Brx dry matter concentration, viscosity, and the DE (dextrose equivalent) index. Among these, the DE value is of particular interest. Dextrose equivalent (abbreviated as DE) represents the amount of reducing sugar, expressed in grams of pure anhydrous D-glucose as a percentage of the dry matter. The DE value, during starch hydrolysis, indicates the degree to which starch has been hydrolyzed into reducing sugars. In DoE investigation, DE was selected as the objective function.

DE index was determined by Lane-Eynon method which was well described in the literature [17]–[19].

3. Results and Discussion

3.1. Hydrolysis with α-amylase

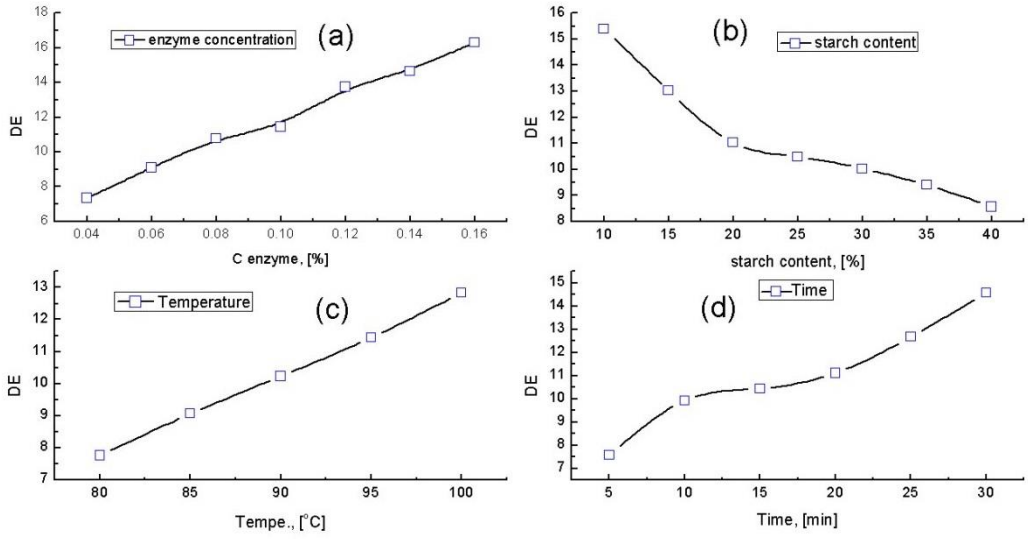

Figure 1. *DE variation under influences of (a) enzyme concentration, (b) starch content, (c) hydrolysis temperature and (d) hydrolysis time*

The Fig. 1 illustrates the effects of enzyme concentration, starch content, hydrolysis time, and hydrolysis temperature. First, an increment in enzyme concentration leads to an increase in the DE index. As shown in Fig. 1a, higher enzyme concentrations expedite the liquefaction process but also escalate costs. Conversely, lower enzyme concentrations slow down liquefaction and prolong the process. Within the range of 0.06% to 0.10% enzyme concentration, the DE index falls within the desired range of 9 – 12. Second, concerning starch content as seen in Fig. 1b, increasing this factor results in a decrease in the DE index. Excessive starch content can impede enzyme-starch interaction, reducing the rate of the hydrolysis reaction and leading to a lower DE index. Third, under similar conditions of starch content, temperature, and enzyme concentration, higher temperatures accelerate the reaction rate, yielding higher DE indexes as seen in Fig. 1c. The temperature range of 95 – 100 °C is considered as ideal for enzyme operation, resulting in the desired DE index. Finally, Fig. 1d reveals that extended hydrolysis times correspond to higher DE values obtained. It was also produced lower viscosity, and improved filtration efficiency. Short hydrolysis times are associated with low DE indexes and high viscosity, making the filtration process challenging.

From these screening steps of the above four parameters, temperature was fixed at 95 ºC for all hydrolysis. The rest three parameters were further investigated in a matrix of a full DoE plan with enzyme concentration (Z_1/x_1), starch content (Z_2/x_2), and hydrolysis time (Z_3/x_3) as shown in Table 1.

Table 1. *The 2^3 DoE factorial design plan for enzymatic hydrolysis of starch*

Number	**Real variable**			**DoE variable**			**Objective function**
N	Z_1, (%)	Z_2, (%)	Z_3,(min)	x_1	x_2	x_3	DE
1	0.12	35	25	+1	+1	+1	12.03
2	0.12	35	10	+1	+1	-1	11.15
3	0.12	15	25	+1	-1	+1	11.89
4	0.12	15	10	+1	-1	-1	11.24
5	0.06	35	25	-1	+1	+1	9.55
6	0.06	15	25	-1	-1	+1	10.65
7	0.06	35	10	-1	+1	-1	8.99
8	0.06	15	10	-1	-1	-1	9.86
Central composite							
9	0.09	25	17.5	0	0	0	11.25
10	0.09	25	17.5	0	0	0	10.95
11	0.09	25	17.5	0	0	0	11.33

Regression model was obtained as the following equation after validation and elimination the influenced parameters according to Fisher's criteria with 95% confidence interval which similarly done as the literature [20].

$$y = 10.554 + 0.864x_1 + 0.316x_3 + 0.369x_1x_2 \quad (1)$$

To optimize the production of maltodextrin from cassava starch using the α-amylase enzyme method at a temperature of 95°C, the ideal conditions for producing maltodextrin with a DE index of 9 to 12 include an enzyme concentration of 0.12%, starch content of 35%, and a hydrolysis time of 10 minutes. Comparable results of the influence of temperature and reaction time have been reported by Toratane et al. [21].

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Table 2. *Results of the optimal parameters of the maltodextrin production process using the α-amylase*

Real variable	DoE variable	DoE value	Exp. value
Z_1	x_1	+1	0.12
Z_2	x_2	+1	35
Z_3	x_3	-1	10
DE	Y(%)	11.47	11.46

3.2. Hydrolysis with catalysis HCl

There are several parameters that affect the hydrolysis of starch using acidic agent HCl. As depicted in Fig. 2a, the DE increases as a function of HCl used because a higher acid concentration results in more H^+ ions which participated in starch hydrolysis. This allows greater contact with starch molecules and more thorough starch breakdown, leading to a higher DE index. However, when the acid concentration is too high (greater than 10%), maltodextrin may develop a brown color that impacts the product's appearance. Therefore, in the production of maltodextrin using the acid method, a maximum acid concentration of 10% was considered. Secondly, in Fig. 2b, elevated temperatures lead to increased reaction rates, higher viscosity, and a higher DE index. However, higher temperatures also cause maltodextrin to take on a darker brown color and reduce efficiency. Therefore, the chosen production temperature was 55 °C, as at this temperature, the DE index of maltodextrin falls within the desired range of 9 to 12. Thirdly, as seen in Fig. 2c, longer hydrolysis times are associated with higher viscosity and an increased DE index. Prolonged hydrolysis times allow H^+ ions more interaction with starch, resulting in more extensive starch chain cleavage. Finally, Fig. 2d, higher starch content leads to lower viscosity and DE values. This suggests that with higher starch content, there is less contact between starch and acid, which reduces the ability to hydrolyze starch and results in a lower DE index. Nevertheless, using too low of a starch content can compromise performance and increase production costs. Similar effects relating to starch content has been reported by Sievert et al. [22].

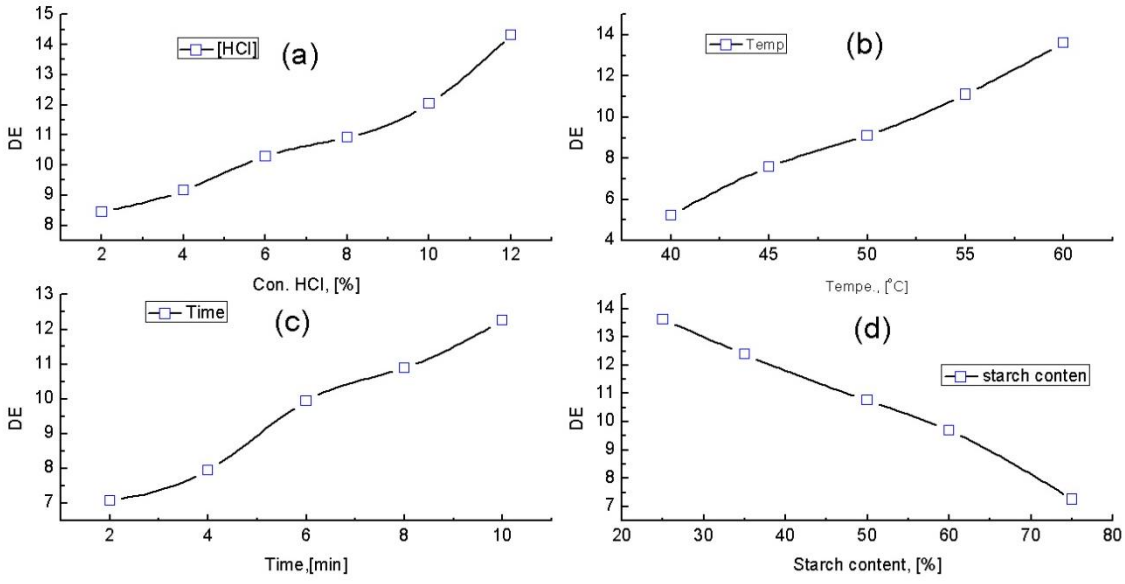

Figure 2. *HCl method for starch hydrolysis in consideration effects of HCl concentration, starch content, temperature and time of hydrolyzation.*

Afterwards, three parameters were selected for DoE investigation with experimental plan as listed in Table 3. From the total factor experimental planning method, calculating the coefficients' variances and eliminating non-influential factors resulted in the regression equation:

$$y = 9.631 - 0.724x_2 + 0.716x_3 + 0.281x_1x_2 - 0.444x_2x_3 \quad (2)$$

The regression equation reveals that two factors i.e. starch content and hydrolysis time, significantly influence the DE index. Specifically, starch content is inversely proportional to the DE index, while hydrolysis time is directly proportional to the DE index. Among these factors, starch content has the most pronounced impact on the objective function. To increase the DE index, reducing the starch content is the most effective approach. The regression equation provides a straightforward way to adjust the conditions for process improvement. The optimal conditions are outlined in Table 4, in which HCl acid concentration of 4%, starch content relative to the acid of 25%, and a hydrolysis time of 10 hours are considered as optimal conditions for producing maltodextrin (9–12 DE index) from cassava starch using the HCl acid method at a temperature of 55°C.

Table 3. *The 2^3 DoE full factorial design plan for HCl hydrolysis of starch*

Exp. N	Real variable			DoE variable			OF
	Z_1, (%)	Z_2, (%)	Z_3, (hours)	x_1	x_2	x_3	Y(%)
1	8	75	10	+1	+1	+1	9.97
2	8	75	4	+1	+1	-1	8.57
3	8	25	10	+1	-1	+1	12.25
4	8	25	4	+1	-1	-1	9.06
5	4	75	10	-1	+1	+1	8.39
6	4	25	10	-1	-1	+1	11.78
7	4	75	4	-1	+1	-1	9.04
8	4	25	4	-1	-1	-1	9.33
Central composite							
9	6	50	7	0	0	0	10.36
10	6	50	7	0	0	0	10.57
11	6	50	7	0	0	0	10.25

Table 4. *Results of the optimal parameters of the maltodextrin production process using HCl catalyst*

Real variable	DoE variable	DoE value	Exp. value
Z_1	x_1	-1	4
Z_2	x_2	-1	25
Z_3	x_3	+1	10
DE	Y(%)	11.79	11.81

4. Conclusions

This paper presents an investigation of the factors affecting the dextrose equivalent index of maltodextrin produced from cassava starch using two hydrolysis methods, i.e. HCl acid and α-amylase enzyme. The Design of Experiment method was employed to derive regression equations after validation and the elimination of less significant parameters as well as their interactions. To produce maltodextrin with a DE index ranging from 9 to 12, the results can be summarized and compared as follows. In the acid method, the optimal conditions include an HCl concentration of 4%, starch content of 25%, and a hydrolysis time of 10 hours at 55 °C. In contrast, for the enzyme method, the ideal parameters were found at a set of enzyme concentration of 0.12%, starch content of 35%, and a hydrolysis time of 10 minutes at 95°C. Therefore, the enzymatic method was proven to be more effective due to shorter time required and higher starch content being handled. Moreover, the bio-engineering approach allows for easy adjustment of the DE value according to specific requirements. Last but not least, enzyme method does not lead to product discoloration and equipment corrosion as faced in maltodextrin production via the acid method.

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Conflict of Interest

The authors declare no conflict of interest.

REFERENCES

[1] D. Saha and S. Bhattacharya, "Hydrocolloids as thickening and gelling agents in food: a critical review," *J. Food Sci. Technol.*, vol. 47, no. 6, pp. 587–597, Dec. 2010.
[2] A. Apriyanto, J. Compart, and J. Fettke, "A review of starch, a unique biopolymer—Structure, metabolism and in planta modifications," *Plant Sci.*, vol. 318, p. 111223, 2022.
[3] A. Zarski, K. Bajer, and J. Kapuśniak, "Review of the Most Important Methods of Improving the Processing Properties of Starch toward Non-Food Applications," *Polymers*, vol. 13, no. 5, p. 832, 2021.
[4] S. A. Osemeahon, O. Maitera, and B. Dimas, "Influence of starch addition on properties of methylolated urea/starch copolymer blends for application as a binder in the coating industry," *J. Toxicol. Environ. Health Sci.*, vol. 7, no. 5, pp. 44-51, 2015.
[5] C. O. Moore, J. V. Tuschhoff, C. W. Hastings, and R. V. Schanefelt, "Application of starches in foods," *Food Sci. Technol.*, pp. 575-591, 1984.
[6] P. Bhatt *et al.*, "Structural Modifications and Strategies for Native Starch for Applications in Advanced Drug Delivery," *Biomed Res. Int.*, vol. 2022, p. 2188940.
[7] P. Wang and X. Xu, "Modified Starches and the Stability of Frozen Foods," in *Starch in Food*, 2018, pp. 581-593.
[8] J. Compart *et al.*, "Customizing Starch Properties: A Review of Starch Modifications and Their Applications," *Polymers*, vol. 15, no. 16, p. 3491, 2023.
[9] R. C. S. Thys *et al.*, "The effect of acid hydrolysis on the technological functional properties of pinhão (Araucaria brasiliensis) starch," *Ciênc. Tecnol. Aliment.*, vol. 33, pp. 89-94, 2013.
[10] P. Chen *et al.*, "Effect of acid hydrolysis on the multi-scale structure change of starch with different amylose content," *Food Hydrocolloids*, vol. 69, pp. 359-368, 2017.
[11] T. Munegumi, M. Inutsuka, and Y. Hayafuji, "Investigating the Hydrolysis of Starch Using α-Amylase Contained in Dishwashing Detergent and Human Saliva," *J. Chem. Educ.*, vol. 93, no. 8, pp. 1401-1405, 2016.
[12] S. Ozdemir *et al.*, "A Novel Raw Starch Hydrolyzing Thermostable α-Amylase Produced by Newly Isolated Bacillus mojavensis SO-10: Purification, Characterization and Usage in Starch Industries," *Braz. Arch. Biol. Technol.*, vol. 61, p. e18160399, 2018.
[13] Federal Register, vol. 63, no. 58, pp. 14608-14611 (1998, Mar. 26), *Rules and Regulations*. [Online]. Available: https://www.govinfo.gov/pkg/html
[14] A. Parikh, S. Agarwal, and K. Raut, "A Review of applications of maltodextrin in pharmaceuticals industry," *Int. J. Pharm. Bio Sci.*, vol. 4, no. 4, pp. 67-74, 2014.
[15] Z. Saavedra *et al.*, "Technological Application of Maltodextrins According to the Degree of Polymerization," *Molecules*, vol. 20, no. 12, pp. 21067-21081, 2015.
[16] S. V. Borges, "Application of maltodextrin in green corn extract powder production," *Powder Technol.*, vol. 263, pp. 89-95, 2014.
[17] N. T. Hien, "Improving technology, production equipment, and the application of Maltodextrin from corn starch in the pharmaceutical and food industries," (in Vietnamese), Institute of Food Industry, Hanoi, Vietnam, 2006.
[18] *Hydrolyzed products from starch - Determination of reducing power and titratable acidity of Dextrose - Lane and Eynon standard titration method*, (in Vietnamese), Vietnam Standard TCVN 10376:2014, Vietnam, 2014.
[19] C. S. James, *Analytical chemistry of foods*, U.K.: Springer Science & Business Media, 2013.
[20] T. L. Minh, N. H. Dong, and D. N. Tan, "Design of Experiment Method – An Application in Improving Quality of Magnesium Hydroxide Produced from Natural Seawater Bittern," *J. Chem. Eng. Res. Updates*, vol. 8, pp. 48-59, 2021.
[21] T. Munegumi *et al.*, "Investigating the Hydrolysis of Starch Using α Amylase Contained in Dishwashing Detergent and Human Saliva," *J. Chem. Educ.*, vol. 93, no. 8, pp. 1401-1405, 2016.
[22] D. Sievert, Y. Pomeranz, "Enzyme resistant starch. I. Characterization and evaluation by enzymatic, thermomechanical, and microscopic methods," *Cereal Chemistry*, vol. 66, pp. 342-347, 1989.

Dr. -Eng. Tam Minh Le. (*tamlm@hcmute.edu.vn*) PhD at Max Planck Institute for Dynamics of Complex Technical Systems, Magdeburg, Germany. Postdoctoral research at Vietnam Academy of Science and Technology. Editorial Board Member of JCERU journal. Expertise in the fields of advanced materials, enantiomer separation, crystallization, chromatography, process analytical technology (PAT), chemical and bio-process engineering. ORCID: https://orcid.org/0000-0002-5372-346X

Dr. Nhung Thi Tran. (*nhungtt@hcmute.edu.vn*) has received a B.E. degree in chemical engineering from Ho Chi Minh city University of Technology, Vietnam, in 2010; a M.E. degree in chemical engineering in Gachon University, Korea, in 2012; and a Ph.D degree in materials science and engineering in Nanyang Technological University, Singapore, in 2017. She is currently a lecturer at Ho Chi Minh city University of Technology and Education, Vietnam. ORCID: https://orcid.org/0000-0003-0352-9463