Mathematical Modelling of a Process-Economic of Protein Hydrolyzate Production from Lupine Flour

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Abstract: The aim of the present work is to perform the mathematical modelling and computer simulation of the lupine flour hydrolysis technology, which includes the calculations and also the partial comparison of different process variants. Lupine flour is deemed as a source of valuable protein with a substantial content of essential amino acids which has a significant impact on the prevention of various diseases in a human diet. The proposed mathematical model for the production process enables to accomplish the optimal operating conditions while the specific costs for the production hydrolysis unit comprises the three main processes for the implementation of the hydrolyzate production, i.e. the chemical reaction and its kinetics, the separation of solids from the reaction mixture by filtration and the thickening of the final product. The results have shown that in most of the simulation cases, the economic optimum occurs and thus the application of the model is valuable for the practical purposes of protein hydrolyzate production.

Key-Words: mathematical modelling, simulation, lupine flour, hydrolysis, protein, specific costs

1 Introduction

Proteins are important macronutrients with a source of essential amino acids and energy. Lupine or lupine flour is considered as a nontraditional source of proteins, soluble dietary fibre and, unlike cereals, lupine has a low content of starch. Lupine contains over 30 % protein with a very high content of valuable essential amino acids, i.e. arginine, leucine, isoleucine, lysine, and simultaneously is classed among the gluten-free food. On the other side, lupine is not considered as a source of anti-nutritional factors such as trypsin inhibitors and saponins. From the energy source point of view, it competes with cereals and because of its high level of proteins with oil seed meals. By lupine consuming is enhanced by the capacity to supplement other food components to reach an overall balance of nutrients. [1, 2]. Proteins produced by the hydrolysis of these valuable sources may also elicit potent anticancer, antimicrobial, hypocholesterolemic, antihypertensive,

antithrombotic, and antimicrobial effects [3, 4].

The basic process of hydrolysis technology of a solid substrate (e.g. defatted finely ground lupine flour) is the chemical reaction itself occurring in the reactor of the production unit. The final composition of the reaction mixture is defined by the selected reaction conditions, i.e. presence and type of temperature, catalyst, pressure, hydrodynamic conditions in a given process reactor, substrate concentration and reaction time. The convenient choice of the reaction conditions significantly affects the selectivity of the entire process. As a result, only some of the components of the input substrate can be hydrolyzed. Thus, the feedstock can be refined, or valuable fractions can be obtained from the feedstock with acceptable selectivity. In our case, it is possible to use hydrolysis to remove starch from the input substrate or to get a protein hydrolyzate (in other words, to "extract" the protein fraction from the feedstock). The technological arrangement of both processes is similar; only the form of the final product differs. In the case of "refining" hydrolysis, the main output product is a refined solid phase, when using "extraction" hydrolysis, the main obtained output product is a hydrolyzate.

The pattern of the production hydrolysis unit is shown in Figure 1. The figure presents three main processes of the technology, namely the chemical reaction itself - hydrolysis of the solid substrate, separation of solids from the reaction mixture by filtration, and final concentration of the hydrolyzate (i.e. filtrate). The process of the filtrate concentrating is indicated by only one apparatus in the scheme, but in practice, depending on the final dry matter requirement of the resulting hydrolyzate, a combination of, e.g. evaporator and spray dryer can be used. In the scheme, the main components of the individual process streams, which are used for proposal and formation of the mathematical model of the hydrolysis unit, are also indicated.



Fig. 1: Scheme of the production node of the hydrolysis unit including the list of considered components.

Based on the experimental measurements, the preparation of protein hydrolyzate, i.e. "extraction" hydrolysis, is a promising option. Therefore, the aim of this work is focused on the simulation calculations of this process variant. The process-economic mathematical model of the production process is a very useful tool for achieving this goal. But the mathematical model itself is also suitable for simulating and optimizing "refining" hydrolysis since the process layout is similar to that described above.

2 Mathematical Model of the Production Loop of the Hydrolysis Unit

According to the production diagram depicted in Figure 1, a mathematical model was designed, which includes the components that are summarized in the scheme. The input substrate itself is a very complex substance, but for the purposes of the mathematical model it is advisable to simplify its composition and only take into account the proportion of individual components involved in the reaction, the inert fraction, which is unchanged, the water, and the resulting product - i.e. the component that has reacted.

2.1 Basic Economic Model

In terms of the practical application of the mathematical model for the development and design of the industrial process, it is fundamental to involve economic parameters, as these determine the success and economic viability of the proposed technology. In this case, we will consider the cost of performing the chemical reaction and the cost of the resulting product concentrating to the desired level. Due to the process layout, the cost of filtration can be neglected. In particular, the cost of thickening the resulting product, i.e. the hydrolyzate, is expected to be determinative, but, as will be shown below, due to the complex rheological behavior of the reaction mixture, the cost of the implementation of reaction also occupies a significant portion of the total operating costs. Depreciation and wages are not included - they depend on the specific production capacity and production unit.

The cost for the realization of the chemical reaction is determined by the heat required to heat the reaction mixture and the electrical energy needed to homogenize the reaction mixture. The cost of maintaining the desired reaction temperature is negligible when the reactor is well insulated. However, the cost of homogenizing the reaction mixture depends on the dwell time in the reactor. The designed mathematical model works with a batch reactor, therefore it is also suitable for smallscale production. Let us add that due to the heterogeneity of the reaction system (solid substrate suspended in the liquid phase), the stirring intensity significantly increases the reaction rate and stirring of the batch further prevents the formation of any deposits, and for these reasons, we will only consider the variant of a stirred reactor.

From the discussion of the technology economy it follows that the main part of the operating costs (N) of the hydrolyzate preparation consists of the cost of electricity (N_E) needed to drive the stirrer of the hydrolysis reactor and the cost to achieve the desired concentration of hydrolyzate (N_O), which is in practice achieved by evaporation of water from the filtrate (see Figure 1):

$$N = N_E + N_O \tag{1}$$

The costs of energy are determined by the product of the electromotor stirrer input power *P*, the reaction time τ and the unit price of electric energy K_E . The costs associated with the evaporation of water, it means the concentration of the product, are given by the multiplying of the heat of vaporization of water ΔH_{vyp} , the amount of evaporated water m_{BD} and the unit price of thermal energy K_P . Therefore, the operating costs can be expressed as follows:

$$N = K_E \cdot P \cdot \tau + K_p \cdot H_{vyp} \cdot m_{BD}$$
(2)

The cost of electricity increases with increasing reaction time, but prolonging the reaction time leads to an increase in the product concentration in the reaction mixture, which reduces the price of the thickening. As a result, the major part of the operating expenses, depending on the reaction time, is minimal. The purpose of optimization is to find the minimum, i.e. the optimal reaction time at which the sum of costs is minimal. From a practical point of view, the specific costs are crucial. As the main product is thickened hydrolyzate, the following applies to the specific costs:

$$N_m = \frac{N}{m_P} \tag{3}$$

By optimizing the process according to specific costs, suitable operating conditions can be determined. The dependence of the total costs N is more complicated (2), because the input power is a function of stirring intensity and reactor design, the total cost of the reaction is further influenced by the initial composition of the reaction mixture and other factors. For this reason, in the following chapters we describe in detail the mathematical model used to calculate total costs and discuss individual implemented functional dependencies.

2.2 Kinetic Model of the Batch Hydrolysis Reactor

It follows from the experimental data that the hydrolysis of the input substrate can be modeled with acceptable accuracy using first order kinetics. The mass balance of the hydrolyzable portion of the substrate (component "S" of process streams) in the batch reactor is the following:

$$k. a_{S,N}. (1-x). m_{RS} = a_{S,N}. m_{RS} \frac{dx}{d\tau}$$
(4),

where k denotes the hydrolysis rate constant valid for a given substrate, x stands for the reaction conversion, m is the stream weight and a is the mass fraction. The subscripts denote the balanced component in accordance with the technological scheme (Figure 1).

Mass fractions instead of the usual concentrations were used in differential equation (4) for the reactant balance because this record takes into account both the experimental measurements and the kinetic data evaluation. The reaction time estimation depending on the desired conversion and vice versa are received by the integration of equation (4):

$$\tau = -\frac{1}{k} \ln (1 - x) \tag{5}$$

$$x = 1 - e^{-k\tau} \tag{6}$$

From the mass balance it is possible to easily calculate the output composition of the reaction mixture, assuming that there is no evaporation of the volatile components of the reaction mixture:

$$a_{H,RS} = a_{S,N} \cdot x \tag{7}$$

$$a_{S,RS} = a_{S,N} \cdot (1-x) \tag{8}$$

$$a_{AQ,RS} = a_{AQ,N} \tag{9}$$

$$a_{I,RS} = a_{I,N} \tag{10}$$

Due to the massive molar excess of water in the reaction phase it is justified to neglect water consumption for hydrolysis, as is clear from Eq. 9.

2.3 Mathematical Model of Reactor Batch Homogenization

The aim of this part of the mathematical modelling of the batch reactor is to estimate the input power of the electric motor needed to achieve the desired hydrodynamic mode of the reaction mixture. For the characterization of the hydrodynamic mode we used dimensionless Reynolds criterion (Re_M), which takes the following form for stirring [5]:

$$\operatorname{Re}_{M} = \frac{n \cdot d^{2} \cdot \rho}{\eta} \tag{11}$$

where *n* stands for the stirrer rotational speed, *d* is the stirrer diameter, ρ stands for the reaction mixture density and η denotes the reaction mixture dynamic viscosity.

The input power of the motor required to achieve the specified hydrodynamic mode can be calculated from the dimensionless input power criterion *Po* [5], which is also often referred to as the Euler's Criterion [6]:

$$Po = \frac{P}{\rho \cdot n^3 \cdot d^5}$$
(12)

The dependence of *Po* on *Re* for various stirrer types can be found in the literature (see Figure 2 as an example). From this dependency, it is evident that in a specific range of the Reynolds criterion of stirring, the value of the input power criterion is constant; this area is called automodel regime [6]. Depending on the type of stirrer, the automodel regime range starts at approximately at *Re_M* equal to 1000. The full turbulent mode usually occurs when the *Re_M* value of 10000 is reached in the entire reactor volume; considerably wide area between 10 < *Re_M* <10000 is the transition region [5].

Due to the heterogeneity of the reaction system, it is desirable to work in the turbulent region, or at least in the transition region with the highest stirring intensity. If the so-called kinetic mode is reached, the reaction rate is independent of the hydrodynamic conditions (the chemical reaction is the ratedetermining step), if the stirring intensity is lower (transition or diffusion area), the observed reaction rate is significantly affected or directly controlled by mass transfer [7]. However, the mass transfer rate is also highly dependent on the stirring intensity [6]. Consequently, it is desirable to ensure sufficient stirring in the industrial scale.

For mathematical modelling and simulation of the production process, we will practically in all cases work in the automodel regime and the input power criterion *Po* will act as a constant in the mathematical model according to the type and construction of the stirrer.

By modifying relations (11) and (12), we get the final relation for the calculation of the electromotor input power:

$$P = \frac{\text{Po} \cdot \text{Re}_{M}^{3} \cdot \eta^{3}}{\rho^{2} \cdot d}$$
(13)

It can be seen from relation (12) that the input power significantly depends on the hydrodynamic conditions and the viscosity of the reaction system. It is also considerably influenced by the chosen stirrer and reactor design. For the possibility of variation of the reactor volume in the simulation calculations, constants are introduced into the mathematical model, the product of which with the reactor diameter gives the required geometric dimension, e.g. the level of the reaction mixture in the reactor or stirrer diameter *d*.



Fig. 2: Dependence of input power criterion Po on Re – container with 4 radial baffles; 1 - turbine stirrer with straight vertical blades and separating disc, 2 - turbine stirrer with wing blades, 3 - six-blade stirrer with straight, inclined blades, 4 - propeller stirrer with constant helix, 4a - like 4, but no baffles.

To calculate the input power of the electromotor, it is necessary to estimate the viscosity of the reaction system. Aqueous suspensions of solids containing a hydrolysable fraction (mainly starch) represent very complex systems in terms of quantitative description of their rheological behaviour [8-10]. A certain complication is a fact that these are usually non-Newtonian fluids [8, 9] and, besides, due to the ongoing hydrolysis, the viscosity changes during the reaction. The viscosity of the reaction system is, of course, further dependent on the concentration of the solid substrate in the suspension [9, 11]. A detailed description of such a system requires precise knowledge of rheological behaviour, which is dependent on the specific composition of the input substrate and specific conditions.

A detailed study of this issue is extraordinarily demanding on special instrumentation, and its mathematical apparatus is not yet reliably known. Therefore, for optimization and simulation of the influence of individual parameters on the modelled system, we made the following simplification - we considered the dependence of viscosity on the shear rate negligible for the reasons mentioned; therefore, we further assumed to be working with a Newtonian system.

To express the effect of the initial composition of the reaction mixture on its viscosity, we proceed from experimental data measured in [9] for rice flour suspension in water. The following graph (Figure 3) shows the dependence of the reaction mixture viscosity on the concentration of the solid substrate and the fit of the experimental data by a mathematical model, which is further used to estimate the viscosity of the reaction system. This dependence is non-linear, i.e. increase in solid phase concentration results in a steep increase in viscosity. The viscosity was measured in samples after partial hydrolysis of the hydrolysable fraction, with gelatinization of the starch fraction. Therefore, the use of this data represents a reasonable qualified estimate of the real behaviour of the modelled system.

By the synthesis of these relations, we will attain the mathematical model of the hydrolysis reactor of the main production unit. Figure 4 shows the dependence of the considered cost of the reaction (i.e. the cost of electricity N_E) on the conversion of the reaction. As expected, this dependency is nonlinear and increases with increasing conversion, since longer reaction times are required to achieve higher conversion. The mathematical model constants used for this calculation are summarized in Table 1.



Fig: 3: Dependence of reaction mixture viscosity on the solid substrate concentration, circles indicate experimental data used from [9], the line indicates mathematical model data.



Fig. 4: Dependence of batch processing costs on the desired conversion.

Table 1: Parameter values used for the simulation calculation of batch processing costs.

Parameter	Value	Unit
K_E	6	[Kč/kWh]
K_P	600	[Kč/GJ]
k	0,034	$[\min^{-1}]$
$a_{S.N}$	2,5	[% w/w]
$a_{I.N}$	4,5	[% w/w]
$ au_{ind}$	0	[min]
V_R	5	$[m^3]$
chr	1,4	[1]
cmr	0,4	[1]
Re _M	3000	[1]
Ро	0,35	[1]
ρ	1000	[kg.m ³]
Typ of stirrer	propeller	

 τ_{ind} – induction period, V_R – reactor volume, chr – level reactor constant, cmr – stirrer diameter constant, ρ – density of reaction mixture, K_E the price of electricity, K_P -the price of steam

2.4 Mathematical Model of Subsequent Processes of the Technology

As can be seen from the technological process diagram (Figure 1), the hydrolysis is followed by filtration of the reaction mixture and concentration of the filtrate. To calculate the total operating costs (1) (2), knowledge of the evaporated water amount is crucial. Furthermore, to calculate the specific operating costs, which are the most important from the practical point of view, it is necessary to quantify the amount of products produced - either the filter cake in the "refining" variant or the amount of filtrate in the "extraction" variant. All the above information can be obtained from the mass balance of both processes.

It is possible to estimate with a good accuracy from performed experimental work the output dry matter of the filter cake (a_{DM}, F_K) . It consists of an inert portion of feedstock which is not involved in the reaction and is still present in the solid phase and a part of the hydrolysable substrate which has not reacted under the reaction conditions. Filtrate, the dry matter of which contains the hydrolyzate, is not included in the dry matter of the filter cake. Thus, the filtrate retention is neglected. In practice, the filter cake is usually washed, so that the assumption mentioned above of the dry matter of the filter cake consisting only of the inert and unreacted portion is fulfilled. Accordingly, the mathematical model proposed below suggests transferring all of the hydrolyzate to the filtrate. From the mass balance of the process, including the above assumptions for the weight of the filter cake (m_{FK}) , follows:

$$m_{FK} = \frac{m_{RS} \cdot [a_{S,N}(1-x) + a_{I,N}]}{a_{DM,FK}}$$
(14)

The filtrate weight is then equal to:

$$m_F = m_{RS} \cdot \left(1 - \frac{[a_{S,N}(1-x) + a_{I,N}]}{a_{DM,FK}} \right)$$
(15)

In addition to the weights of both streams, the concentration of hydrolyzate in the filtrate is essential. Based on the process mass balance, the following equation can be derived for the hydrolysate concentration:

$$a_{H,F} = \frac{a_{S,N} \cdot x}{1 - \frac{[a_{S,N}(1-x) + a_{I,N}]}{a_{DM,FK}}}$$
(16)

By evaporating the water from the filtrate, the resulting product is obtained - thickened hydrolyzate. It depends on the dry matter concentration requirement in the final product. This requirement, of course, has a very significant impact on overall production costs, but it is also due to market requirements or shelf-life product requirements. It should be noted that the dry matter of the resulting product consists solely of the hydrolyzed fraction, the inert fraction and the unreacted fraction are completely separated in the previous process, filtration. For the calculation of the final product weight after evaporation of the required amount of water, based on the mass balance of the filtrate thickening process, the following relation arises:

$$m_P = \frac{m_{RS} \cdot a_{S,N} \cdot x}{a_{H,P}} \tag{17}$$

The amount of evaporated water is equal to:

$$m_{BP} = m_{RS} \cdot \left(1 - \frac{[a_{S,N}(1-x) + a_{I,N}]}{a_{DM,FK}} \right) \cdot \left(1 - \frac{a_{H,F}}{a_{H,P}} \right)$$
(18)

2.5 Overall Mathematical Model

The overall mathematical model is obtained by synthesizing the expressions discussed in the previous chapters. After the implementation of the mathematical model in computational and simulation programs (e.g. Matlab, Scilab etc.) it is possible to perform extensive simulation calculations that take into account the critical parameters of the modelled process, i.e. the production node of the hydrolysis unit.

The model allows the adjustment of input parameters based on experimental data; similarly, it is possible to supplement the experimental dependencies of individual physicochemical quantities directly.

3 Simulation Calculations

As was discussed in the previous chapters, it can be expected that the overall process costs have a minimum in a particular area. With total specific costs, the situation is somewhat more complicated because not only less water evaporates when high conversions are achieved, but more product is produced, which reduces overall specific costs. For this reason, the optimal conditions from the total and specific costs point of view may vary quite significantly. From a practical point of view, the specific costs are crucial, i.e. costs related to the amount of product per unit.

The simulation performed with the proposed mathematical model under the conditions summarized in Table 2 shows, that specific costs optimization can significantly improve the economy of the entire production process. Reynolds stirring criterion Re_M was set to 5000. The calculated dependence of the main specific cost on conversion is shown in the figure (Figure 5). Under the given input parameters, it is clear that the optimal conversion is approximately 91%. Note that the reaction rate used in the simulation was evaluated from experimental data and refers to "extraction" hydrolysis of lupine flour.

Of course, in addition to conversion, several other parameters significantly affect the overall production costs. As the discussion suggests in deriving of the mathematical model, sufficient stirring intensity is necessary to provide the required mass transport rate in the heterogeneous reaction system. The costs for batch stirring are significantly dependent on the hydrodynamic mode of the plant.

The mathematical model allows comparing the operating costs for hydrolyzate production at different stirring intensities expressed by the Reynolds criterion of stirring. The three-dimensional graph (Figure 6) shows the results of the simulation calculations; the values of the input data are summarized in Table 2.

Table 2: Parameter	values	used	for	simulati	on
calculation.					

Parameter	Value	Unit
K_E	6	[Kč/kWh]
K_P	600	[Kč/GJ]
k	0,034	$[\min^{-1}]$
$a_{S,N}$	2,5	[% w/w]
$a_{I.N}$	4,5	[% w/w]
$ au_{ind}$	60	[min]
V_R	5	$[m^3]$
chr	1,4	[1]
cmr	0,4	[1]
Ро	0,35	[1]
ρ	1000	[kg.m ³]
$a_{DM.FK}$	30	[% w/w]
$a_{H.P}$	95	[% w/w]
$\Delta H_{_{vyp}}$	$2,26 \cdot 10^{6}$	$[J \cdot kg^{-1}]$
Typ of stirrer	propeller	



Fig. 5: Dependence of the main operating costs on conversion.

It can be seen from the simulation that the optimum conversion at which the lowest specific costs are achieved is dependent on the desired mixing intensity. It is advisable to reach higher conversions with decreasing mixing intensity. On the other hand, the specific reactor performance (amount of product manufactured during unit of time) is decreasing, which has to be compensated by reactor larger volume to ensure a constant production rate; thus the investment costs for the acquisition of the hydrolysis unit are increasing. Hence, the mathematical model enables comparison and optimization of the various operating modes of the industrial equipment.



Fig. 6: Dependence of overall specific costs on conversion and Re_M .

From the viscosity measurements follows that this quantity significantly depends on the concentration of the solid input substrate in the reaction mixture. From the economic point of view, it is desirable to process as much concentrated suspension of a substrate in water as is possible because the specific performance of the hydrolysis unit is increasing while decreasing the cost of thickening the product. Hydrodynamic conditions influence the process rate, so it is desirable to keep these at a sufficiently high level. For this reason, we performed simulation calculations to describe the dependence of total specific conversion costs and initial input substrate. The observed dependence is presented in Figure 7, the table of values of the input parameters is summarized (Table 2), Reynolds stirring criterion Re_M was set to 2000 and mass was calculated from relation fraction a_{SN} $0,35 \cdot (a_{S,N}+a_{I,N})$ specifying initial composition of solid reactant - lupine flour.



Fig. 7: Dependence of overall specific costs on conversion and initial solid phase concentration in the reaction mixture.

The results show that there is an economic minimum cost for a given conversion that corresponds to the specific value of the solid phase concentration of the feedstock in the reaction mixture. Similar dependencies can be compiled for a wide range working modes of the operating unit, allowing setting optimum conditions to ensure hydrolyzate economical production.

Let us add that the substrate input concentration is one of the parameters that can be changed on the already finished production plant and thus directly affect the economy of operation. The dependence of the main operating costs on the Reynolds stirring criterion and the initial solid phase concentration in the suspension also offers an interesting comparison (Figure 8). The constants used in the calculation are summarized in the table (Table 2), the conversion was set to 0.9 and mass fraction a_{SN} was calculated from relation $0,35 \cdot (a_{S,N} + a_{I,N})$. It can be seen from the figure that, also, in this case, an economic optimum can be determined in a wide range of conditions, which corresponds to the specific combination of the stirring criterion Reynolds and the initial composition of the reaction mixture. The main operating costs on the z-axis document that the costs savings during production unit operating in the economic optimum area are significant.



Fig. 8: Dependence of total specific costs on initial reaction mixture composition and Re_M .

It is also necessary to assess the feasibility of achieving an appropriate working mode of the production equipment in terms of both design and power requirements. Knowledge of this information is also essential for estimating and calculating investment costs. Figure 9 shows the dependence of the computed input power of stirring device on the initial composition of the reaction mixture, the table (Table 2) summarizes the values of the input parameters of the mathematical model, conversion was set to 0.8 and mass fraction a_{SN} was calculated from relation $0.35 \cdot (a_{SN} + a_{IN})$. The figure shows the sharp nonlinear dependence of input power on the initial composition of the reaction mixture, which is determined by the viscosity of the suspension to its concentration. The mathematical model thus enables estimate and evaluate the technical and to technological feasibility of achieving given specific operating conditions, which is, of course, necessary for the selection of the optimum working area of the entire production node of the hydrolysis unit. Similarly, the proposed mathematical model can be used to calculate the tangential speed of the stirrer and optimize the design of the stirrer in terms of selecting the appropriate type and its diameter. These data are also crucial for the practical optimization of the plant operating mode and the selection of suitable working conditions.



Fig. 9: Dependence of the input power of stirring device required to reach the desired reactor hydrodynamic mode.

4 Conclusion

The proposed mathematical model of the hydrolysis unit includes the basic processes for the preparation of the resulting hydrolyzate, i.e. the chemical reaction itself, filtration of the reaction mixture and concentration of the filtrate, which is the final product. The mathematical model can be used for description of multiple technology variants, both for the "refining" technology and for "extraction" hydrolysis for which most simulation calculations have been performed.

The proposed mathematical model of the batch hydrolytic reactor, which forms the main part of the overall model, takes into account the reaction kinetics, the basic rheological parameters of the batch and the construction of the apparatus. As a result, it is possible to economically optimize the technology not only to find optimal operating conditions under which the specific costs of preparing the final product are minimal but also to assess their technological and technical availability and practical feasibility. The modelling has shown that in most of tested cases the economic optimum exists and thus the application of the model is valuable for the practical purposes.

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