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PBL with the Application of Multiple and Nonlinear Linear Regression in Chemical Kinetics and Catalysis

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Abstract In the resolution of problems in chemical kinetics and catalysis the mathematical models relate the independent variable that is usually time, with the dependent variable which is normally the concentration of a reactant. They conform to linear models, whose parameters such as the ordering to origin and the slope are kinetic parameters, applying linear regression to the experimental data can be obtained linear models that are the simplest to be found. On other side, multiple models can be found with two or more independent variables, however, there are models that are not feasible to linearize and therefore you can make use of nonlinear regression. This paper is exemplified by the Problem-Based Learning (PBL) scheme and the use of software such as Stat graphics and Polymath to solve the resulting mathematical models. The results obtained are compared with traditional methods as multiple linearized least squares, in this way the learning teaching process is strengthened, since the student raises the kinetic model and then solves it with computer tools. Through the PBL cycles generated in the resolution of real problems in chemical kinetics and catalysis, significant learnings are achieved on difficult comprehension topics such as initial rate for the determination of the reaction order, half-life times, temperature effect and heterogeneous catalysis.

Keywords: *PBL, kinetics, heterogeneous catalysis, multiple regression, Stat graphics, Polymath*

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1. Introduction

Chemical kinetics is a branch of Physical Chemistry that is responsible for the study of the course of reactions as a function of time, as well as the factors that modify it. The rate reaction refers to the number of moles of substance transformed per unit volume per unit of time, so speed is an infinitesimal change, and can therefore be expressed as a derivative. Speed is of interest in kinetic studies as the order of a reaction, as well as other kinetic parameters, can be determined from it. The kinetics of a reaction are greatly affected by temperature, and a small variation in temperature can accelerate or slow it down [1]. Because rate is a function of temperature, equations can be combined with these two variables to perform calculations of interest in chemical kinetics, as well as mathematical models serve to predict, explain, and interpret chemical phenomena. It is usual to resort to the linearization of the resulting models to determine certain parameters, it is feasible to perform simple regressions and even for the resolution of certain problems multiple regressions are proposed as it is in the case of heterogeneous catalysis [2]. During the resolution of some problems, the resulting mathematical expressions are not feasible to be linearized, for this purpose their resolution is raised by a nonlinear multiple regression. The resolution of problems by regressions can be done by traditional methods such as linearized least squares, where it is necessary to perform a summation and propose a system of simultaneous equations, but they can also be summarized with the help of software such as the Stat graphics and Polymath [3].

To improve the learning teaching process, various teaching strategies have been designed, including problem-based learning (PBL) [4]. PBL is learning that results from the process of entertainment or solving a problem [5]. The PBL offers students an obvious answer to the questions: Why do we have to learn this information? [6].

PBL is a teaching method that substantially increases the motivation of students, since by its dynamics it makes students active subjects of the learning teaching process [7]. The student is also at the heart of this process and working with this method requires non-individual student teamwork [8,9].

The PBL consists of the approach to a problem situation, where its construction, analysis and/or solution are the central focus of the experience, and where teaching is to deliberately promote the development of the process of researching and resolving the problem in question. The various modalities adopted today by the PBL are tax on the constructivist theories of learning, which highlight the need for students to inquire or intervene in their environment and build for themselves significant learnings [10].

One of the procedures of the PBL is the generation of questions of study and analysis of the case, since the questions of analysis or discussion around the case are fundamental, because they are those that allow the case to be examined intelligently and in depth, while also leading to the central points of the case. Analysis questions are the teacher's fundamental means of mediating the student's encounter with the study material. It raises four types of questions that it is important to integrate into the case analysis: study questions, discussion questions, facilitators and questions about the product or outcome of the case discussion.

The appropriate "good problems" to work in the classroom through PBL cycles, are those defined as open or unstructured, ambiguous, capable of changing and proposing various solutions [11]. The PBL has been applied to the teaching of the Chemical Sciences [12,13,14,15,16] and to the teaching of Physical Chemistry [5,17,18,19,20,21].

When a kinetics with hypochlorite ions and iodide ions are performed experimentally, hypochlorite ions are reduced by iodide ions to determine reaction order and kinetic constant, so the concentration of any of the reacting ions is monitored as a function of time and kinetic treatment can be performed by the differential method of initial speed at constant temperature [22]. When the temperature is changed it is possible to know the activation energy and frequency factor of a reaction; experiments can be performed at different temperatures and the initial speed can be determined to each of them and by linearization of the Arrhenius equation and by combining with the law of speed you can obtain the order of the reaction, activation energy and frequency factor. Likewise, the data obtained in a heterogeneous differential catalytic reactor can be analyzed to obtain the constants of the kinetic equation, for example, the models obtained in the catalytic decomposition of cumene, where the limiting stage of speed is to adsorption on the surface of the catalyst [23] can be analyzed by nonlinear regression.

With the aim of improving the teaching learning of the method topics of initial speed, effect of temperature on the speed of chemical reactions and heterogeneous catalysis by using multiple linear regression and nonlinear regression, in this work the ABP is exemplified as a didactic strategy [8] for the determination of kinetic parameters in the reaction between the hypochlorite and iodide ions, by saponification in basic medium of methyl acetate and heterogeneous decomposition of cumene, using different types of mathematical regression.

2. Methodology

This proposal was applied in a group of the subject Chemistry Kinetics and Catalysis of the career of Chemical Engineering at FES-Cuautitlán UNAM, with 45 students, 63% male and 37% female gender, with average age of 21 years. Students who first took the subject in the morning shift. The group taught face-to-face classes on the topics of differential methods to determine the order of reaction, effect of temperature and equation of Arrhenius and finally heterogeneous catalysis. Subsequently they formed 5-member work teams to address the central problem.

The topic of the central problem was defined as: *Application of multiple and nonlinear linear regression in chemical kinetics and catalysis.* Three questions were established generating the PBL cycle:

1. What is the order of reaction between iodide and hypochlorite ions?
2. What is the reaction order in the basic hydrolysis of methyl acetate, frequency factor and activation energy?
3. What is the law of rate, of the heterogeneous composition of cumene if the limiting stage is an adsorption?

For each of the generating questions, a theoretical framework was established that students should expand and deepen through research as a teaching principle. It was suggested to help software such as Stat graphics and Polymath, because the use of computational packages strengthens the learning teaching process since the use of TCI causes interest, motivation and generates interaction among students [24].

1. *What is the order of reaction between iodide ions and hypochlorite?*

The reaction between the ions is:

$$I^- + OCl^- \rightarrow OI^- + Cl^-$$

[Scheme 1]

Whose law of rate is expressed:

$$R_A = kC_{I^-}^{\alpha}C_{OCl^-}^{\beta} \quad (1)$$

Applying natural logarithm:

$$LnR_A = Lnk + \alpha LnC_{I^-} + \beta LnC_{OCl^-} \quad (2)$$

Being R_A the rate of the reaction $\left[\frac{mol}{Lmin}\right]$, k is the kinetic constant whose units depend on the overall order of the reaction, α y β are partial orders for each reactant.

The overall order of the reaction is the sum of partial orders, and these can be obtained by the differential method of analysis of the initial speed, which consists in determining the speed at different initial concentrations of the reacting ions, iodide, and hypochlorite. Having initial rapidity values at different concentrations, the kinetic constant $n$and partialorders can be obtained as parameters of a multiple linear regression of equation (2):

$$y = a_1 + a_1x_1 + a_2x_2 \quad (3)$$

The order to origin a_1 corresponds to the $Ln\,k$, the value of the slope a_1 corresponds to the partial order α, while the slope a_2 it is the partial order β.

2. *What is the reaction order in the basic hydrolysis of methyl acetate, frequency factor and activation energy?*

The reaction scheme is as follows:

$$CH_3COOCH_3 + OH^- \rightarrow CH_3COO^- + CH_3OH$$

[Scheme 2]

The law of rate of reaction is represented by *equation 4:*

$$R_A = kC^{\alpha}_{CH_3COOCH_3}C^{\beta}_{OH^-} \quad (4)$$

Applying natural logarithm to both members of the equation:

$$lnR_A = Lnk + \alpha LnC_{CH_3COOCH_3} + \beta LnC_{OH^-} \quad (5)$$

By modifying the temperature of the reacting system, the kinetic constant in the same way is modified based on the Arrhenius equation:

$$k = Ae^{-\frac{E_A}{RT}} \quad (6)$$

In its logarithmic form takes the following form

$$Lnk = LnA - \frac{E_A}{R} * \left(\frac{1}{T}\right) \quad (7)$$

Combining equation [14] with equation [25] results in:

$$LnR_A = LnA - \frac{E_A}{R} * \left(\frac{1}{T}\right) + \alpha LnC_{CH_3COOCH_3} + \beta LnC_{OH^-} \quad (8)$$

Being, the frequency factor whose units depend on the order of reaction, A E_a is the activation energy $\left(\frac{J}{mol}\right)$, R the universal constant of gases, α y β are the partial orders of each reactant.

Equation [25] is a multiple linearization whose schematic model is:

$$y = a_0 + a_1x_1 + a_2x_2 + a_3x_3 \quad (9)$$

Regression parameters are: $a_0 = Lnk$, $a_1 = -\frac{E_A}{R}$, , $a_2 = \alpha$ y $a_3 = \beta$.

3. *What is the law of rate of the heterogeneous composition of cumene if the limiting stage is adsorption?*

The catalytic decomposition reaction is:

$$C_9H_{12} \xrightarrow{clinoptilolite} C_6H_6 + C_3H_6$$

The cumene (C_9H_{12}) decomposes on the surface of the catalyst to give as benzene products (C_6H_6) and propylene (C_3H_6), the mechanism of the heterogeneous process is shown in Figure 1:

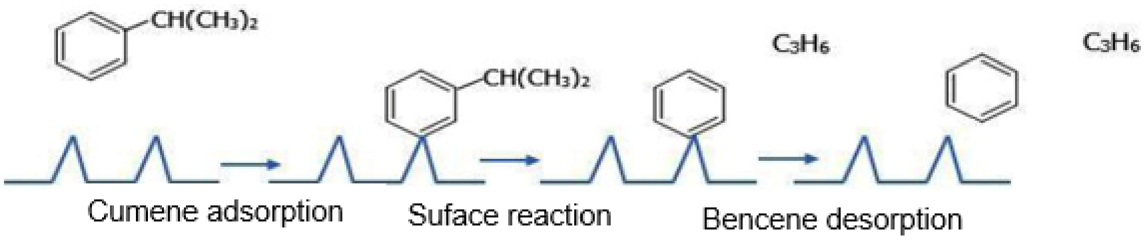


Figure 1. Sequence of steps for catalytic decomposition of the cumene on the surface of a zeolite

Whose individual steps are:

Adsorption of the cumene: $C_9H_{12} + S \rightleftharpoons C_9H_{12} * S$ [Scheme 3]

Surface reaction: $C_9H_{12} * S \rightleftharpoons C_6H_6 * S + C_3H_6$ [Scheme 4]

Benzene desorption: $C_6H_6 * S \rightleftharpoons C_6H_6 + S$ [Scheme 5]

Fogler [25] establishes that the limiting stage of the mechanism is the adsorption of the cumene on the surface of the clinoptilolite and based on it the law of rate is expressed in equation (10):

$$r_{AD} = \frac{k\left(P_C - P_P P_B / K_P\right)}{1 + K_B P_P P_B / K_s + K_B P_B} \quad (10)$$

The resulting model cannot be linearized by some algebraic technique, the data can be processed by a nonlinear regression, which can be performed with the help of a software.

3. Results and Discussion

Rate is of interest in kinetic studies as the order of a reaction, as well as other kinetic parameters, can be determined from it. The kinetics of a reaction are greatly affected by temperature, and a small variation in temperature can accelerate or slow it down [1]. Because speed is a function of temperature, equations can be combined with these two variables to perform calculations of interest in chemical kinetics, as well as mathematical models serve to predict, explain, and interpret chemical phenomena. It is usual to resort to the linearization of the resulting models to determine certain parameters, it is feasible to perform simple regressions and even for the resolution of certain problems multiple regressions are proposed as it is in the case of heterogeneous catalysis [2]. During the resolution of some problems, the resulting mathematical expressions are not feasible to be linearized, for this purpose their resolution is raised by a nonlinear multiple regression. The resolution of problems by regressions can be done by traditional methods for instance linearized least squares, where it is necessary to perform a summation and propose a system of simultaneous equations, but they can also be reasoned with the help of software such as the Stat graphics and Polymath.

From the questioning of the central problem, a series of questions, answers and actions arose from the students. The teacher was providing information on the issue questioned, giving information gradually, so that students could integrate, and process the How? and the What for? So that with their questions and information provided they could answer each of the questions generating the cycle PBL.

Finally, to know the opinion of the students regarding the PBL strategy applied to problem solving, an instrument with a Likert scale was applied, to determine the usefulness and satisfaction of the teaching strategy used. The items of the evaluation instrument were:

1. Very disagreed,
2. Disagreement
3. Indifferent
4. Agreement
5. All right, then.

Students answered it anonymously (Table 1)

During the implementation of the teaching strategy, students presented different questions for the resolution of each of the generating questions. Table 2 presents the most frequently asked questions.

Table 1. Assessment of the degree of usefulness and satisfaction

Point out the option that best represents your opinion on the teaching strategy to study differential methods, temperature effect and heterogeneous catalysis
1. The didactic strategy seems to motivate you to solve problems.
2. The strategy makes it easier to understand the concepts of rapid reaction, temperature effect and heterogeneous catalysis
3. You consider the strategy used for the analysis of kinetic data useful
4. You think it makes teamwork easier
5. You consider yourself satisfied with the strategy used in determining the order of reaction, calculation of activation energy and evaluation of constants in heterogeneous catalysis

Table 2. Types of questions during problem solving.

PBL Cycle	**Questions and actions of the student**	***Information provided by the teacher or taken from the bibliography***
What is the order of reaction between iodide ions and hypochlorite?	Being a reaction of two reagents I need to know the initial speed of the reaction to different concentrations of reagents	*You were provided with the kinetic data in Table 3 for processing*
What is the reaction order in the basic hydrolysis of methyl acetate, frequency factor and activation energy?	What are the initial speed values at different concentrations and temperatures?	*The data in Table 4 were provided*
What is the law of rate of the heterogeneous composition of cumene if the limiting stage is adsorption?	What is the law of rate of cumene decomposition?	*The student was asked of the reaction mechanism and based on this the law of speed was modeled*
	¿What are the values of rate?	*Rapid values were provided at different partial pressures for analysis in Polymath software*

Students solved the problems as follows:

Example 1: Determining order in the reaction of iodide and hypochlorite ions

The reaction occurs based on scheme 1

$$I^- + OCl^- \rightarrow OI^- + Cl^-$$

The results of the initial rapidity at various concentrations of reactants are presented in Table 3.

Table 3. Initial rate at different concentrations of reactants

$C_{I^-}(M)$	**0.001**	**0.002**	**0.003**	**0.004**
$C_{OCl^-}(M)$	0.002	0.003	0.004	0.005
$R_{A_0}(M/s)$	0.000075	0.000225	0.00045	0.00075

To perform multiple linear regression to equation (5) it is necessary to obtain the natural logarithm of the data, Table 4.

Table 4. Logarithmic values of experimental data for the application of multiple linear regression

LnC_{I^-}	**-6.9078**	**-6.2146**	**-5.8091**	**-5.5215**
LnC_{OCl^-}	-6.2146	-5.8091	-5.5215	-5.2983
LnR_{A_0}	-9.4980	-8.3994	-7.7063	-7.1954

Multiple linear regression was performed in the Stat graphics software where the regression parameters in Figure 2 were obtained.

The multiple linear regression equation is:

$$LnR_A = 3.62778 + 0.99861 LnC_{I^-} + 1.00209 LnC_{OCl^-} \quad (11)$$

From equation [15] you can get the kinetic parameters, $Lnk = 3.62778$, $\alpha = 0.99861$ and $\beta = 1.00209$, applying the exponential to the natural logarithm of the kinetic constant is obtained $k = 37.6292\ M^{-1}min^{-1}$ and rounding up partial orders $\alpha = 1$ y $\beta = 1$, therefore the overall order of the reaction is two.

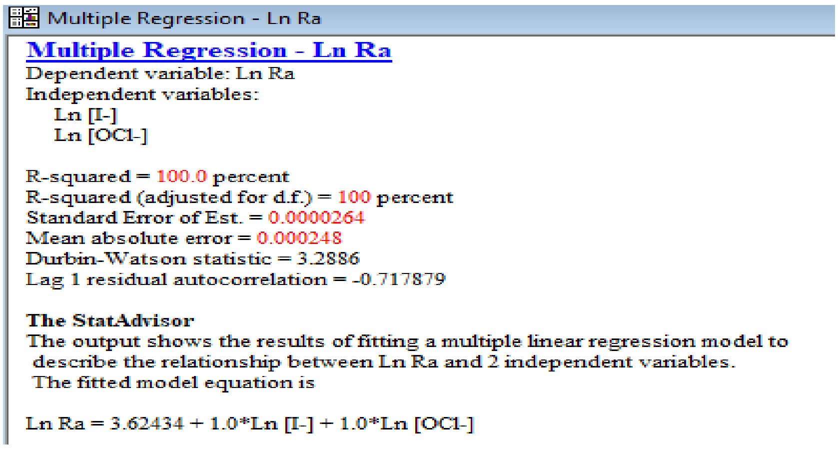


Figure 2. Multiple linear regression parameters for the reaction of iodide and hypochlorite ions

Example 2: Determination of the order of reaction in basic saponification of methyl acetate and evaluation of the frequency factor and activation energy

The reaction occurs in accordance with scheme 2

$$CH_3COOCH_3 + OH^- \rightarrow CH_3COO^- + CH_3OH$$

This example includes the effect of temperature on reaction kinetics, for this purpose experimental data of the rate at different temperatures and initial concentrations of reactants and (Table 5) is available.

Table 5. Experimental data on speed at different initial concentrations of reactants and temperatures

$T\ (C)$	25	30	33	36	40
$C_{CH_3COOCH_3}(M)$	0.05	0.04	0.03	0.02	0.01
C_{OH^-}(M)	0.06	0.05	0.04	0.03	0.01
$R_o\left(\frac{M}{min}\right)$	0.0400	0.0391	0.0293	0.0182	0.0040

The multiple linear models correspond to that presented in equation (8) and for this purpose the inverse of the temperature in Kelvin and natural logarithm must be applied to the initial concentrations and speed, Figure 3 shows the data entered the Stat graphics software as well as the regression parameters obtained with the data:

Multiple linear regression provides the constants that appear in the equation model [25], getting that $LnA = 25.7472,\ \frac{E_A}{R} = 6904.33K,\ \alpha = 0.9998$ y $\beta = 1$, applying the corresponding operations you get that $A = 1.52008x10^{11}$, $E_A = 57402.5996\frac{J}{mol}$, $\alpha = 1$ and $\beta = 1$.

Example 3: Determination of the law of speed in a heterogeneous catalytic process

The mathematical model of heterogeneous decomposition of cumene is expressed according to equation (10):

$$r_{AD} = \frac{k\left(P_C - P_P P_B / K_P\right)}{1 + K_B P_P P_B / K_s + K_B P_B}$$

This model cannot be linearized algebraically, for this a nonlinear regression is performed with the help of the Polymath software to which the experimental data and the mathematical model are entered, for this you must enter an initial value of each constant that appears in the equation for the software to make its iterations until the solution is found.

Based on the mathematical model constants obtained in Figure 3, the law of rate of heterogeneous decomposition of cumene is:

$$r_{AD} = \frac{P_c - \frac{P_P P_B}{10}}{1 + 100 P_P P_B + 9.999 P_B} \qquad (12)$$

The survey to determine the degree of satisfaction and usefulness of the applied teaching strategy, the results presented in a bar chart (Figure 4). It is noted that 94% of students agree or agree very much on the motivation for solving problems. Similar results were obtained by questioning whether the strategy facilitates the compression of the concepts of rapid reaction, temperature effect and heterogeneous catalysis, and whether they consider useful the strategy used for the analysis and compression of kinetic data. In asking whether they consider that the strategy facilitates teamwork 30.7% felt that they were indifferent to it, this is attributed to themselves that students are not accustomed to teamwork in face-to-face theory sessions and therefore do not give it sufficient importance. Finally, 71.4% were pleased with the strategy.

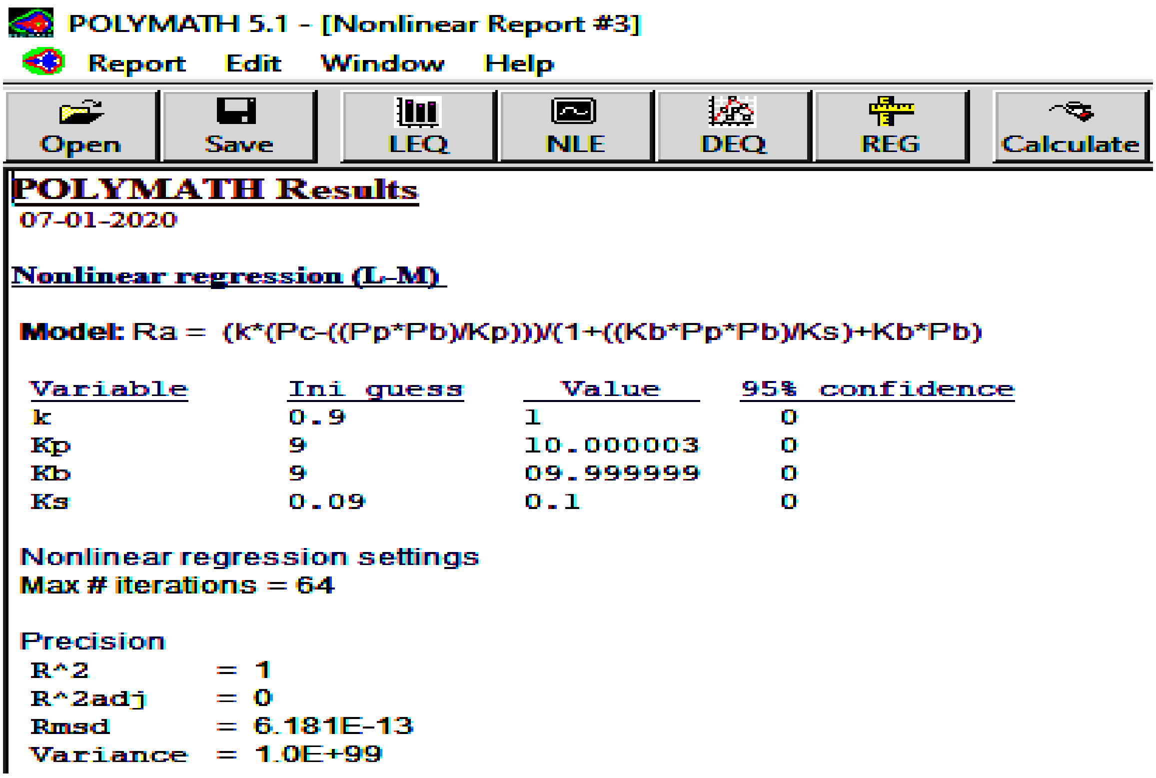
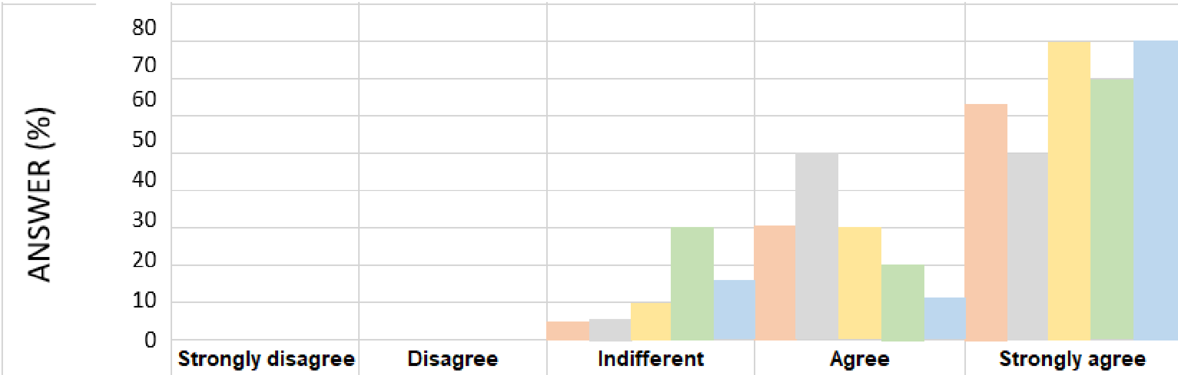


Figure 3. Nonlinear regression for mathematical model of Cumene catalytic decomposition



	Strongly disagree	Disagree	Indifferent	Agree	Strongly agree
1. Motivation	0	0	6	30.2	63.8
2. Compression	0	0	7.1	39.9	53
3. Purpose	0	0	10	30.5	59.5
4. Teamwork	0	0	30.7	20.4	48.9
5. Satisfaction	0	0	15.3	13.5	71.2

Figure 4. Results of the assessment of utility and satisfaction.

By solving the central problem Application of multiple and *nonlinear linear regression in chemical kinetics and catalysis.* Three questions were asked generating the PBL cycle allowing the student to identify the topic, analyze the data and the topic of the PBL cycles by providing significant learning (Table 6).

Table 6. Significant learning on the three exemplified PBL problems

Problem	PBL cycle-generating question	Meaningful learning
1	What is the order of reaction between iodide ions and hypochlorite?	Analysis of kinetic data by the differential method of initial rate
2	What is the reaction order in the basic hydrolysis of methyl acetate, frequency factor and activation energy?	Effect of temperature on kinetics and rapid reaction
3	What is the law of rate of the heterogeneous composition of Cumene if the limiting stage is adsorption?	Mathematical modeling of heterogeneous catalysis and evaluation of the constants of the rapidity equation

4. Conclusions

Examples under PBL scheme [8,26,27,28] as well as those presented in this work, can be applied to improve understanding of fundamental concepts in the courses of chemical kinetics and catalysis. As order and speed of reaction, activation energy, temperature effect, heterogeneous catalysis, etc.

In the approach to problems the student should seek the integration of knowledge about linear regression, and other types of regression.

The student must differentiate how the case of linear regression and other types of multiple regression is presented, when it is linear should consider the use of Stat graphics statistical software as an ICT tool; Also, when presenting the case of a model that is not to be linearized, the student should consider determining the constants of the kinetic model using Polymath software. The proposed strategy is a tool for strengthening teaching learning in kinetics of elementary reactions, using ICT.

Students are considered to have acquired significant learning as they were able to analyze kinetic data by the differential method of initial speed, were able to understand the effect of temperature on the speed of chemical reactions, performed mathematical modeling of heterogeneous catalysis and evaluation of rapidity constants, all using software to solve the mathematical models deduced with satisfactory results.

Based on our teaching experience, its use can be recommended to improve the academic performance of students as they allow the integration of knowledge, as well as collaborative work among students for the problem solving of the area of chemical kinetics and catalysis. A high percentage of students who participated in this strategy were very satisfied with its performance and usefulness.

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