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METHODOLOGY FOR CALCULATING THE PRE-EXPONENTIAL FACTOR USING THE ISOCONVERSIONAL PRINCIPLE FOR THE NUMERICAL SIMULATION OF THE AIR INJECTION PROCESS

METODOLOGÍA DE CÁLCULO DEL FACTOR PRE-EXPONENCIAL EMPLEANDO EL PRINCIPIO ISOCONVERSIONAL PARA LA SIMULACIÓN NUMÉRICA DEL PROCESO DE INYECCIÓN DE AIRE

Padilla-Reyes, Jorge-Mario^{a*}; Trujillo-Portillo, Marta-Liliana^b; Niz-Velasquez, Eider^b

ABSTRACT

The main challenge to predict at field scale the performance of an air injection process is to understand the oil oxidation process and to have a kinetic model of reactions enabling the prediction of process behavior in a reservoir numerical simulator, under different operating conditions. Recently, the *Isoconversional Principle* has been implemented for studying the kinetics of reactions associated with oil oxidation during air injection, based on Ramped Temperature Oxidation tests (RTO). In different published papers, the isoconversional analysis has been used to study the oxidation characteristics of different rock-fluid systems, identify groups of dominant reactions during the crude oil oxidation process, and estimate the effective activation energy for each of the identified reactions. However, in none of them has a procedure been established for estimating the pre-exponential factor, as this is not a direct measure of isoconversional methods. In this article, a mathematical procedure is proposed for estimating the pre-exponential factor based on the application of Friedman's isoconversional method, intended for characterizing the kinetics of the reactions associated with the In Situ Combustion process. This procedure was validated with experimental information and a kinetic model proposed in the literature to model the oxidation behavior of heavy oil.

RESUMEN

El principal desafío para predecir el desempeño en campo de un proceso de inyección de aire es lograr entender el proceso de oxidación del crudo y contar con un modelo cinético de reacciones que permita predecir el comportamiento del proceso en un simulador numérico de yacimientos, bajo diferentes condiciones operacionales. Recientemente, se ha incorporado una técnica denominada *Principio Isoconversional* para el estudio de la cinética de las reacciones asociadas a la oxidación del petróleo durante la inyección de aire, partiendo de la información suministrada por pruebas de oxidación con rampa de temperatura (RTO – Ramped Temperature Oxidation). En diferentes trabajos publicados, se ha utilizado el análisis isoconversional para estudiar las características oxidativas de diferentes sistemas roca – fluido, identificar grupos de reacciones dominantes durante el proceso oxidativo del crudo y estimar la energía de activación efectiva para cada una de las reacciones identificadas. Sin embargo, en ninguno de ellos se ha establecido un procedimiento que permita estimar el factor pre-exponencial, debido a que este no es una medida directa de los métodos isoconversionales. En este artículo se presenta un procedimiento matemático para la aproximación del factor pre-exponencial a partir de la aplicación del método isoconversional de Friedman, orientado a la caracterización de la cinética de las reacciones asociadas al proceso de Combustión In Situ, este procedimiento fue validado con información experimental y un modelo cinético propuesto en la literatura para modelar el comportamiento oxidativo de un crudo pesado.

KEYWORDS / PALABRAS CLAVE

Thermal recovery | In situ combustion | Isoconversional analysis | Kinetic modeling.
Recobro térmico | Combustión in situ | Análisis isoconversional | Modelado cinético.

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1 INTRODUCTION

An efficient mechanism to increase oil production in heavy oil fields is the implementation of the In Situ Combustion (ISC) process. During the process, part of the crude oil present in the reservoir is oxidized, generating heat, which increases the mobility of oil due to the reduction of its viscosity, Sarathi [1]. The increase in oil production is associated with mechanisms of displacement by combustion gases, displacement by steam, reduction of viscosity due to the action of temperature, and thermal expansion of crude oil, among others.

Not all reservoirs are suitable to carry out ISC processes. Therefore, prior to its implementation at the field, feasibility studies at experimental level and numerical simulation of reservoirs are conducted. Experimental studies are performed in order to understand the oxidation characteristics of a given fluid/matrix system; in particular, experimental studies can be classified in two types: quantitative and qualitative. Among the quantitative studies, combustion tube is to be highlighted, which, among other things, allows for determining process performance parameters such as the amount of oil burned and the speed of advance of the combustion front. On the other hand, among the qualitative analysis, the following stand out: *RTO* (ramped temperature oxidation), *TGA* (thermogravimetric analysis), *DSC* (differential scanning calorimetry), *DTA* (differential thermal analyzer) and *ARC* (accelerating rate calorimetry); these types of experiments are cataloged as screening tools that allow to determine if a matrix/fluid system meets the characteristics of a good candidate to ISC, Huang and Sheng [2]. These experiments are also used for the formulation of kinetic models that are subsequently validated by numerical simulation models; kinetic models consist of a reaction scheme and a kinetic triplet (activation energy, reaction frequency or pre-exponential factor and a reaction model).

The interpretation of qualitative experiments aimed at the formulation of kinetic models can be classified basically in two groups; the first one is based on the pre-establishment of a reaction model and the subsequent derivation of the kinetic parameters (activation energy and pre-exponential factor) from the Arrhenius equation, conventional method. The second, isoconversional method, Cinar [3], allows to identify if the oil under study has favorable characteristics to support an air injection process, does not require prior assumption of a reaction model, and allows for direct determination of the effective activation energy.

Isoconversional analysis is a technique that has been widely applied in kinetic studies of varied chemical processes; however, the adaptation of this technique to the study of kinetics in reactions associated with the ISC process was carried out by Cinar et. al [4]. From its first application for the study of kinetics in the reactions involved in the ISC process, there has been no clarity on how the value of the pre-exponential factor could be estimated, considering that it is not a direct measure of the mathematical models used for the analysis of experimental data.

Vyazovkin [5] presented a compendium of some correlations proposed in the literature for the application of the reaction model associated with isoconversional methods. In this paper, a mathematical procedure is proposed that allows for estimating the pre-exponential factor using the correlations presented by Vyazovkin [5]. Furthermore, the oxidation characteristics of a fluid/matrix system corresponding to a Colombian heavy oil field are evaluated. To carry out this research, the experimental results reported by Padilla [6] were used, where the RTO reactor of the air injection laboratory of the Colombian Petroleum Institute (ICP) was used to study the oxidation characteristics of a fluid/matrix system belonging to a Colombian heavy oil field.

2. THEORETICAL FRAMEWORK

The oil oxidation process is usually expressed based on elementary reactions involving pseudo-components, which are grouped based on the fact that the behavior of crude oil phases is controlled by PVT (Pressure, Volume and Temperature) data. In general, it is established that at least two reactions must occur: one in which the fuel is deposited and another in which it is consumed, Chen [7]. Although assuming elementary reactions is an accepted practice for modeling the ISC process, it must be emphasized that the oxidation of crude during the ISC involves hundreds or thousands of reactions in tandem and in parallel, with a high degree of exothermicity.

The reaction rate can be expressed based on Equation 1.

$$R_c = k(T)f(C) \quad (1)$$

Where $k(T)$ represents the rate constant and $f(C)$ the model of reactions. The rate constant depends on temperature and is usually expressed from the Arrhenius equation. On the other hand, the reaction model depends on the concentration of the reagents. As can be observed in Equation 1, the reaction rate is a function of both the temperature and the concentration of reagents. With the aim of simplifying calculations to study reaction kinetics, Friedman [8] proposed that for a same conversion value, the reaction rate is only

a function of temperature. The foregoing implies that for different experiments carried out at different heating rates, the value of $f(C)$ is independent of temperature, as it depends only on the concentration of reagents, which can be expressed as a function of conversion (X) and can be estimated from oxygen consumption. Therefore, Equation 1 can be expressed through Equation 2

$$R_c = k(T)f(X) \quad (2)$$

For a reactor batch, the reaction rate according to Equation 2 would be:

$$-\frac{dX}{dt} = Ae^{-E_X/RT} * f(X) \quad (3)$$

The linear shape of the Equation 3 is:

$$\ln\left(\frac{dX}{dt}\right) = \ln A + \ln f(X) - E_X/RT \quad (4)$$

Equation 4 is the base expression for the application of different isoconversional methods.

To apply the isoconversional analysis, a series of experiments must be performed varying the heating rate, on which basis it is possible to obtain the activation energy by of integral and differential isoconversional methods. The first one determines the activation energy from temperature data and the heating rate for the experiments conducted. On the other hand, the differential methods determine the activation energy starting from the reaction rate and the temperature profiles, using Equation 4. The latter are the most commonly used, due to their simplicity and reliability.

With respect to the Friedman's differential method, the left part of Equation 4 related to the inverse temperature, for each of the tests carried out, obtaining the activation energy from the slope of isoconversion values. The value of the intercept includes the reaction model and the pre-exponential factor. The latter are assumed as constant for the different conversion values and also constant for the various heating rates.

After applying the isoconversional principle on the experimental results of a set of RTO tests performed on a heavy crude oil, an activation energy versus conversion energy graph is obtained, similar to that presented in **Figure 1**. On this one, it would be possible to overlay an intercept graph vs conversion, $\ln A^*f(X)$, expecting a similar behavior to that observed in **Figure 1**. A behavior of this type indicates the consistency of the results [8].

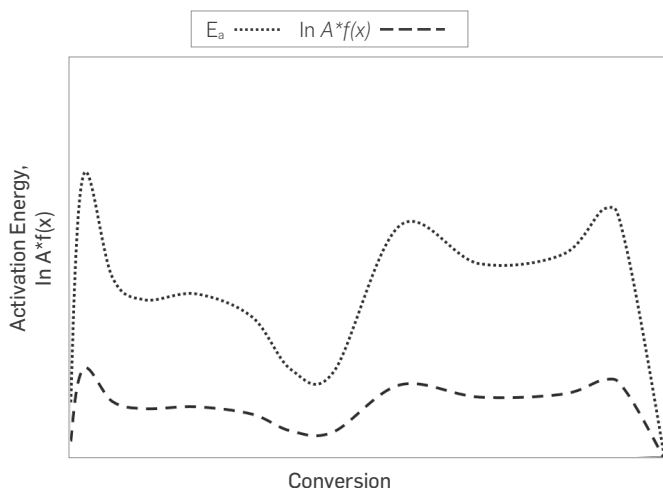


Figure 1. Typical behavior of the Activation energy and against oxygen conversion.

Isoconversional methods do not provide direct information about the pre-exponential factor and the reaction model. However, Friedman [8] presented a mathematical procedure for estimating the pre-exponential factor for elementary reactions, assuming that the reaction model approximates

$$f(X) \cong (1 - X) \quad (5)$$

Whereas the Equation 5 is a good approximation of the reactions model, by multiplying each side of the Equation 5 by A and taking the natural logarithm, you have:

$$\ln[A * f(X)] \cong \ln A + \ln(1 - X) \quad (6)$$

Plotting the intercept of Equation 4 ($\ln A^*f(X)$) vs $\ln(1-X)$, a straight line is obtained which intercept represents the natural logarithm of the pre-exponential factor, according to Equation 6.

In addition to the approach of the reaction model suggested by Friedman, some additional ones have been presented in the literature, Vyazovkin [5] presents a summary of some of them, on which the same procedure presented above could be applied (see **Table 1**). However, it must be taken into account that this procedure must be applied for each group of reactions identified in the analysis of the crude oil fingerprint (graph of activation energy versus conversion), given that the above-described procedure described was proposed for elementary reactions.

Table 1. Correlations for the approximation of the reaction model, Vyazovkin [5].

Operating model	Code	f(X)
Power Law	P4	$4X^{3/4}$
Power Law	P3	$3X^{2/3}$
Power Law	P2	$2X^{1/2}$
Power Law	P2/3	$2/3X^{-1/2}$
Avrami-Erofeev	A4	$4(1-X)[- \ln(1-X)]^{3/4}$
Avrami-Erofeev	A3	$3(1-X)[- \ln(1-X)]^{2/3}$
Avrami-Erofeev	A2	$2(1-X)[- \ln(1-X)]^{1/2}$
Diffusion in one dimension	D1	$1/2X^{-1}$
Diffusion in two dimensions	D2	$[- \ln(1-X)]^{-1}$
Diffusion in three dimensions	D3	$3/2(1-X)^{2/3}[1-(1-X)^{1/3}]^{-1}$

3. EXPERIMENTAL DEVELOPMENT

To perform this work, a series of tests were programmed using the RTO reactor from Colombian Petroleum Institute (ICP). The equipment is fully automated and consists of a gas injection system (nitrogen or air), a tubular reactor where the rock/crude sample is kept, on which a linear increase in temperature is applied, plus a gas measuring system that allows quantifying the amount and concentration of effluent gases (CO, CO₂ and O₂). Moreover, the equipment has security systems that ensure the integrity of people and instrumentation with respect to overpressure, high temperatures and high flows of injection gas. All process variables are monitored and stored in a computer. **Figure 2** is an image of the equipment.

As described in the literature, for the application of isoconversional analysis, at least three RTO tests are required, Glatz [9], at three different heating rates, keeping other operational parameters constant. Based on this, three RTO trials were scheduled with the experimental conditions presented in **Table 2**.

The oil/rock ratio is part of the protocol established by the air injection laboratory of the Colombian Petroleum Institute, considering that the mass of oil loaded in the reactor must be sufficient so that the amount of carbon oxides generated can be measured by the gasometer, not generating deviations from the imposed heating ramp. For conducting the experiments, dead oil from a Colombian heavy oil field was used, which was dehydrated until reaching a water content of 0.8%. **Figure 3** shows the behavior of oxygen consumption versus time and **Figures 4** and **5** show the production of carbon oxides for each of the RTO tests carried out.



Figure 2. Oxidation Reactor with Temperature Ramp, ICP.

Table 2. Experimental conditions for the application of isoconversional analysis.

Sand mass, g	50
Oil mass, g	0.5
Density of oil @ 15 °C, g / cm ³	1.011
Air flow rate, L (S) / h	10
Heating Rate, °C / min	1.5, 1.7, 2.0
Injection gas	Air
Concentration of O ₂ , %	20.94
Operating Pressure, kPa	8234
Maximum temperature (°C)	550
Matrix	Core

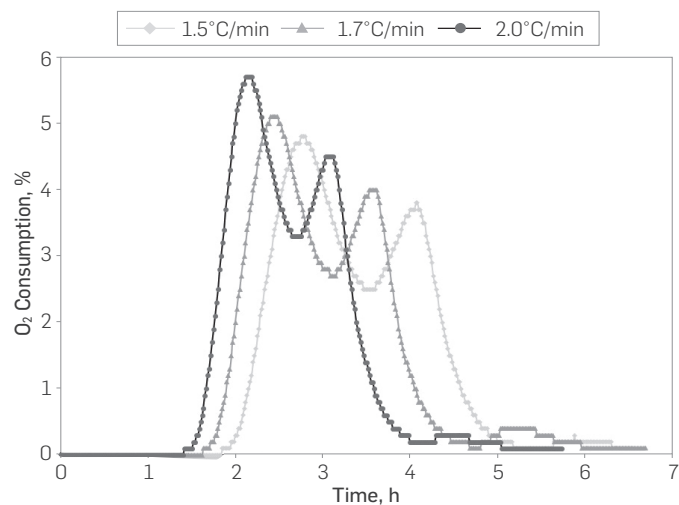


Figure 3. Experimental behavior of oxygen consumption in the different tests.

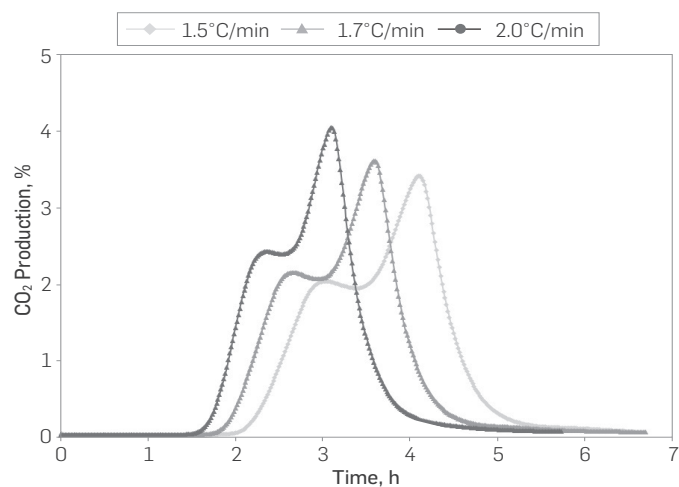


Figure 4. Experimental behavior of production of carbon dioxide in the different tests.

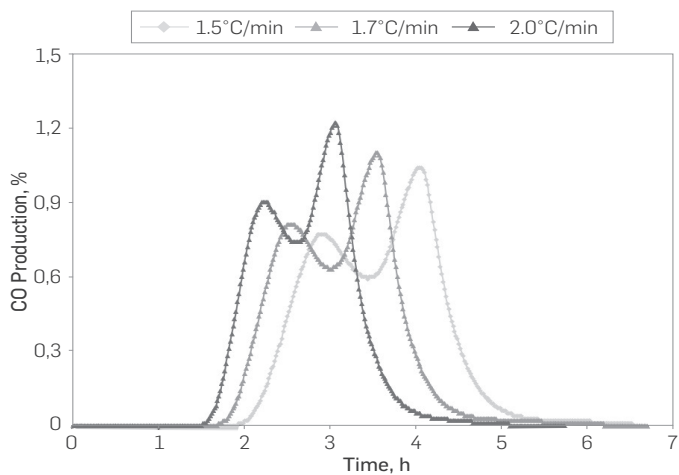


Figure 5. Experimental behavior of production of carbon monoxide in the different tests.

4. RESULTS ANALYSIS

Figure 3 shows three peaks of oxygen consumption for each one of the tests. The first two peaks represent the regions of oxidation at low and high temperatures. In particular for the fluid/matrix system studied it is observed that the first peak of oxygen consumption is greater than the second, contrary to the expected behavior for heavy crude, Gutiérrez et al. [10]. However, this is not observed in the production curves of carbon oxides (Figures 4 and 5), where the first peak is less than the second. Considering that oxygen moles consumed in the production of carbon oxides can be approximated with Equation 7, the oxygen consumed by the addition reactions was determined (Figure 6), finding that the amplitude of the first peak is the product of the occurrence of this type of reactions, evidence of the high reactivity of the fluid analyzed. It also suggests the possibility of achieving spontaneous ignition during process field implementation, Padilla [6] and Padilla et al. [11].

$$O_{2\text{Consumed}} = CO_2 + 0.5 CO \quad (7)$$

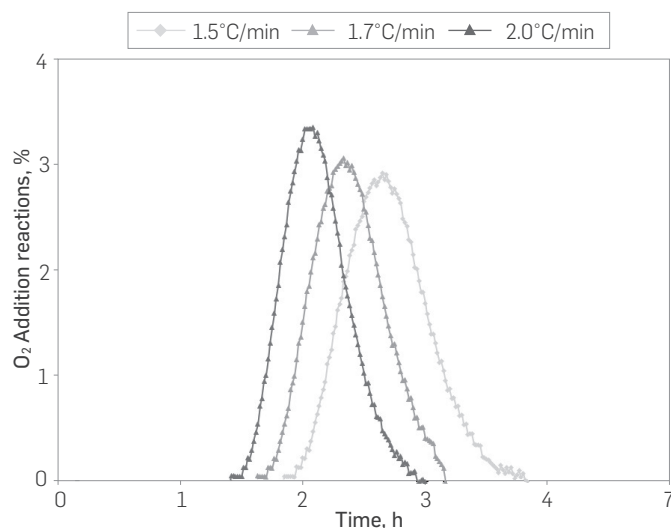


Figure 6. Oxygen consumed by the addition reactions.

Regarding the third peak, a previous investigation carried out on the fluid/matrix system studied, Trujillo [12] determined that this is the product of the reaction between oxygen and some component of the rock matrix at temperatures close to 550 °C, discarding that the peak was caused by a reaction with organic matter in the matrix, as there was no third peak in the production curve of carbon oxides (CO₂ and CO). Figure 4 shows the production of CO₂ for each of the tests, thus ratifying what was stated by Trujillo [12].

CALCULATION OF THE PRE-EXPONENTIAL FACTOR

The Friedman differential method was used to obtain the activation energy graph as a function of the conversion (Crude fingerprint). Figure 7 shows the estimated activation energy for the set of RTO tests performed, as well as the intercept as a function of oxygen conversion. It is to be noted that both the Fingerprint and the intercept follow the same trend, which indicates, as mentioned

by Friedman [8], the consistency in the kinetics of the reactions associated with the process.

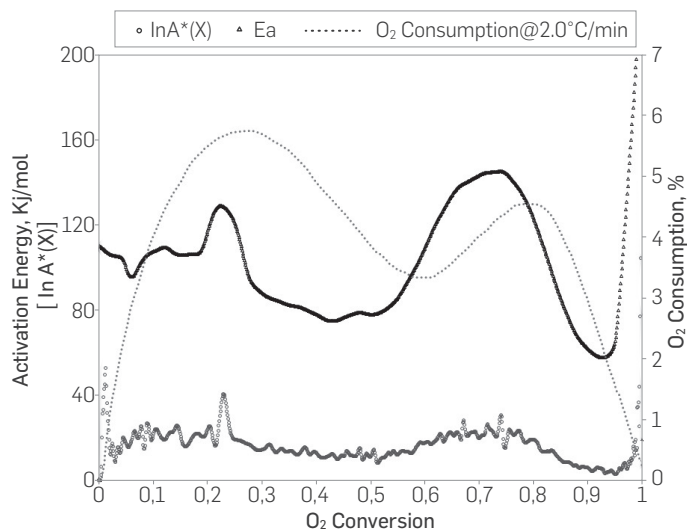


Figure 7. Crude fingerprint, Intercept of the Friedman method and Consumption of oxygen against conversion.

Cinar [3] recommended overlaying the oxygen consumption curve on the fingerprint, to identify groups of dominant reactions and associate them with each of the three-oxidation regions established for ISC studies. The regions or regimes of oxidation are: oxidation at low temperatures (LTO - Low Temperature Oxidation), oxidation at intermediate temperatures (MTO - Middle Temperature Oxidation) and oxidation at high temperatures (HTO - High Temperature Oxidation). Figure 8 shows the activation energy vs the average temperature in the three RTO tests, this graph allows to associate a range of temperatures to each oxidation region and / or group of reactions.

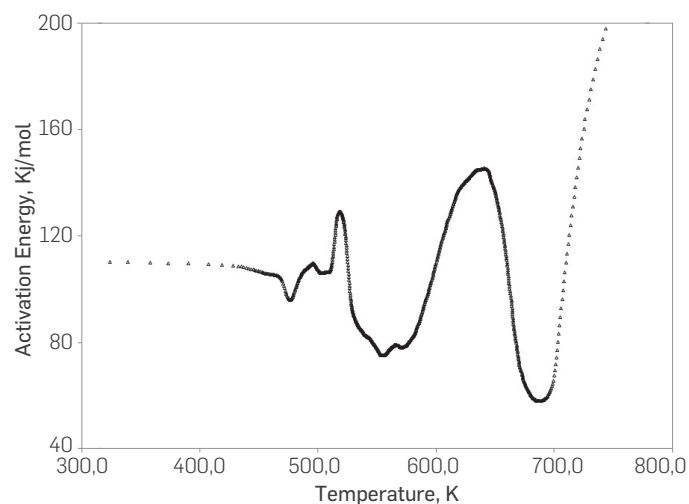


Figure 8. Fingerprint of the crude under study, Activation energy vs Average temperature in the three RTO tests.

Observing the crude fingerprint, it was found that the process is clearly dominated by three reactions, Padilla et al. [11]. The first one occurs in the LTO region between conversion values of 0.08 - 0.27 and temperatures of 170 - 260 °C, with an activation energy

Table 3. Results of approximation of the pre-exponential factor, Friedman method and power laws.

Operating model	Code	Pre-exponential factor, min ⁻¹		
		LTO Region	MTO Region	HTO Region
Friedman	-	8.45E+10	5.45E+07	9.97E+12
Power Law	P4	3.65E+10	9.70E+07	1.26E+16
Power Law	P3	3.13E+10	6.63E+07	2.98E+16
Power Law	P2	2.55E+10	4.00E+07	4.54E+14
Power Law	P2/3	6.14E+10	1.86E+07	6.08E+11

of approximately 114 kJ / mol. The second reaction corresponds to an endothermic reaction associated with the MTO region and it occurs between conversion values of 0.4 - 0.5 and temperatures of 280 - 300 °C with an activation energy of approximately 80 kJ / mol. Finally, a reaction was identified in the HTO regime with an activation energy of approximately 140 kJ / mol, and it occurs with conversion between 0.7 - 0.8 and temperatures of 350 - 400 °C. After determining the average activation energy for each of the reaction groups, we proceeded to implement the method proposed by Friedman and the power law correlations presented by Vyazovkin [5] to approximate the pre-exponential factor for each reaction. Table 3 shows the results obtained after applying the proposed procedure, where it is observed that:

- The pre-exponential factor calculated for the LTO and MTO regions is within the same order of magnitude range, regardless of the model used to approximate the reaction model (see Figures A1 - A10).
- In the HTO region, it can be observed that the model used has a significant effect on the pre-exponential factor, with variations of up to four orders of magnitude. The main reason for this behavior is that the graph $\ln A^*f(X)$ vs $\ln f(X)$ of the Friedman reaction models and Power Law (P2/3) do not present the same degree of linearity as the others (see Figures A11 - A15); therefore, the slope and the intercept for these two graphs are different, resulting in significant differences in the estimated values for the pre-exponential factor.

Padilla *et al.* [13], proposed a kinetic model, based on the experiments used in this work, achieving an acceptable reproduction of the

oxidation behavior of the matrix fluid system in a reservoir numerical simulator. In their work they considered the pre-exponential factor as an adjustment parameter, obtaining values of 8.27E + 09, 5.00E + 07 and 3.72E + 10 for the LTO, MTO and HTO regions, respectively. In general, the pre-exponential factor values presented in Table 3 are close to those estimated by Padilla *et al.* [13], show that the procedure used for the approximation of the pre-exponential factor is a very useful tool for the formulation of kinetic models intended to represent the oxidation behavior of heavy crude in commercial numerical simulators.

CONCLUSIONS

The isoconversional analysis technique has been widely used to study the oxidation characteristics of different fluid/matrix systems, candidates for ISC processes, allowing to eliminate the dependence on the reaction model over kinetic studies of reactions, identify groups of dominant reactions during the oxidation of oil, and estimate the effective activation energy for each of these reactions. Nevertheless, a procedure to estimate the pre-exponential factor has not been considered to date. In this regard, this paper shows that although the pre-exponential factor is not a direct measure of isoconversion methods, it is possible to obtain an approximation based on a simple mathematical treatment. This is a useful tool for building kinetic models intended for the representation of the oxidation behavior of fluid systems/matrix, candidates for ISC processes, using commercial numerical reservoir simulator software.

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NOMENCLATURE

R_c :	Reaction rate
$K(T)$:	Reaction rate constant
$f(C)$:	Reaction model depending on the concentration of reagents
dX/dt :	Reaction rate as a function of oxygen conversion.
A :	Arrhenius pre-exponential factor.
E_x :	Activation energy for isoconversional values.
R :	Universal constant of gases.
T :	Absolute temperature.
X :	Conversion of oxygen.
$f(X)$:	Reaction model based on oxygen conversion.
$O_{2\text{Consumed}}$:	Moles of oxygen consumed.
CO_2 :	Moles produced from carbon dioxide.
CO :	Moles produced from carbon monoxide.

APPENDIX A

GRAPHS FOR THE CALCULATION OF THE PRE-EXPONENTIAL FACTOR

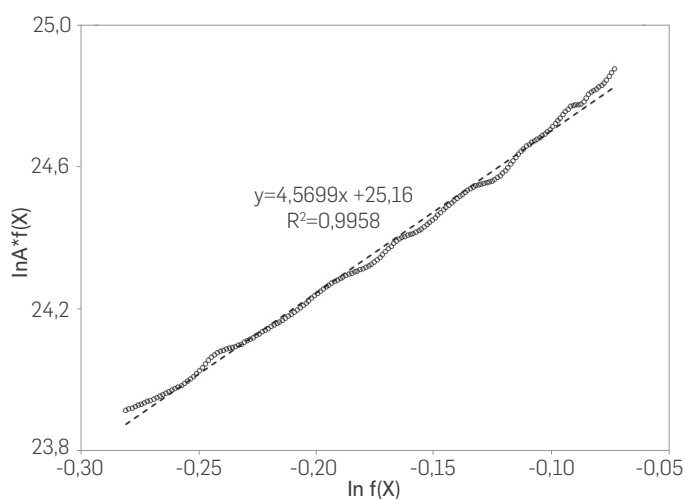


Figure A1. Graph for the approximation of the pre-exponential factor in the LTO region, Friedman (1964).

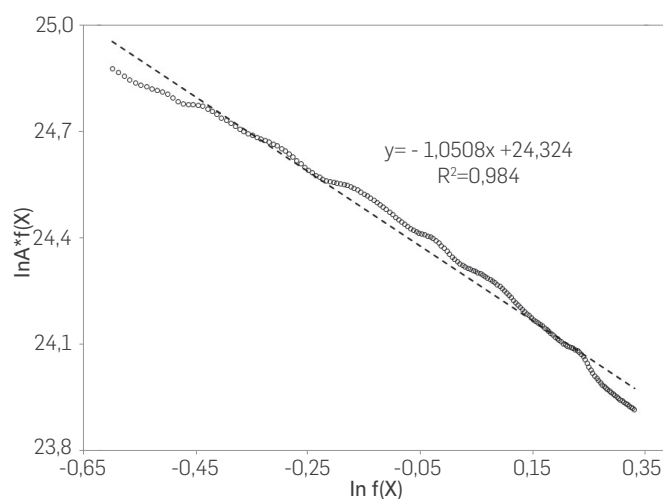


Figure A2. Graph for the approximation of the pre-exponential factor in the LTO region, Power Law, (P4).

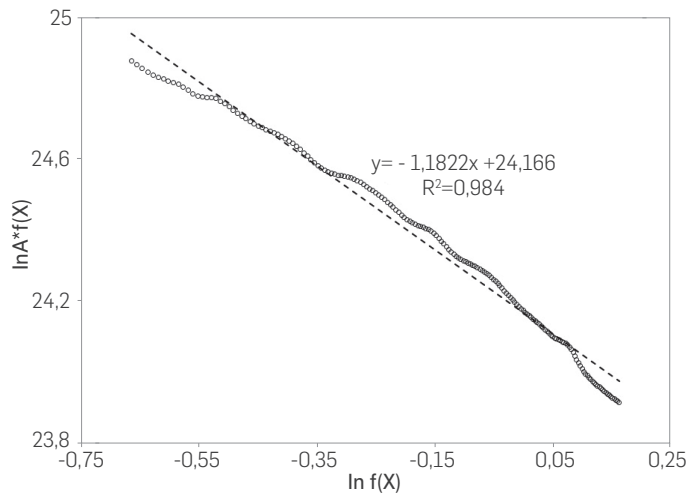


Figure A3. Graph for the approximation of the pre-exponential factor in the LTO region, Power Law (P3).

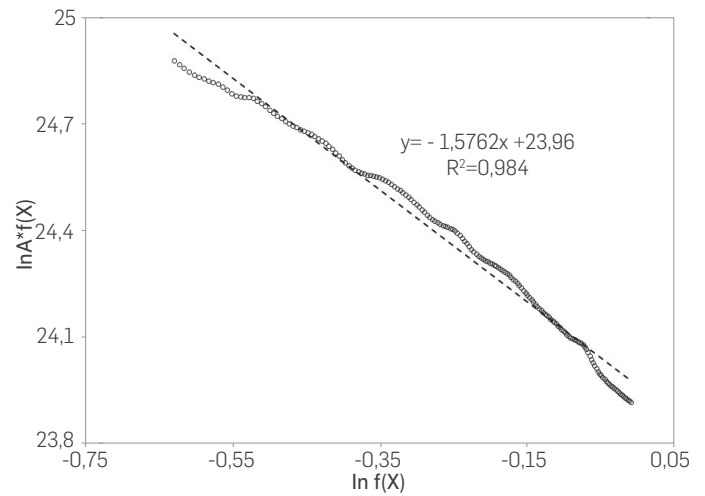


Figure A4. Graph for the approximation of the pre-exponential factor in the LTO region, Power Law (P2).

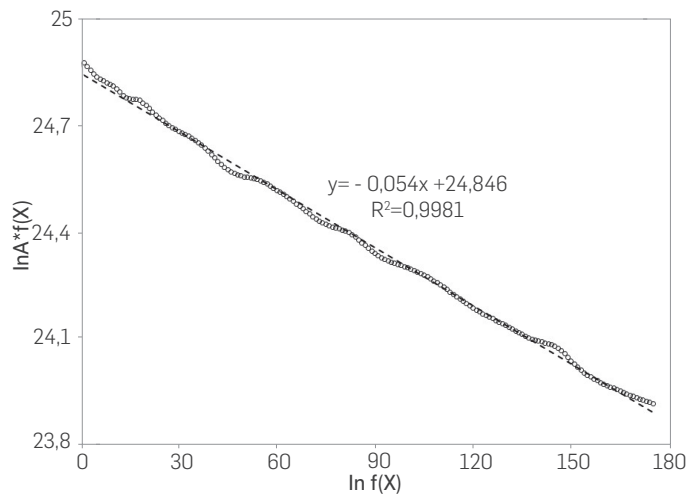


Figure A5. Graph for the approximation of the pre-exponential factor in the LTO region, Power Law (P2/3).

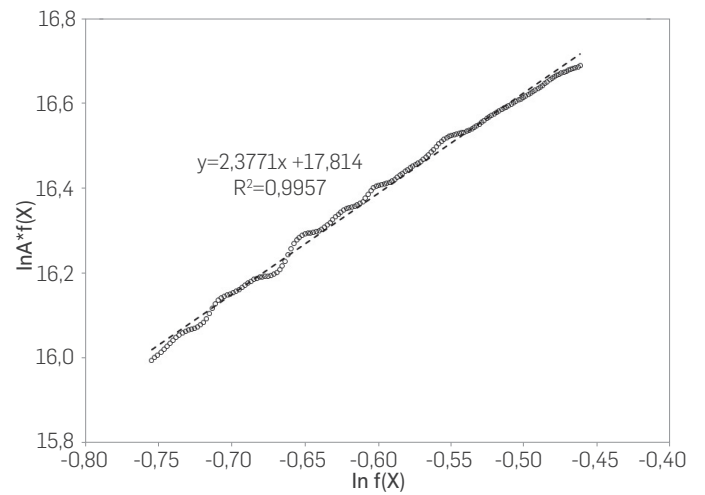


Figure A6. Figure A1. Graph for the approximation of the pre-exponential factor in the MTO region, Friedman (1964).

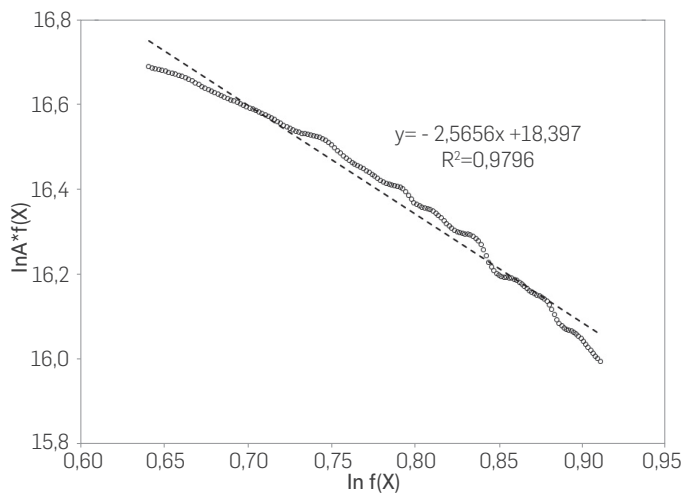


Figure A7. Graph for the approximation of the pre-exponential factor in the MTO region, Power Law (P4).

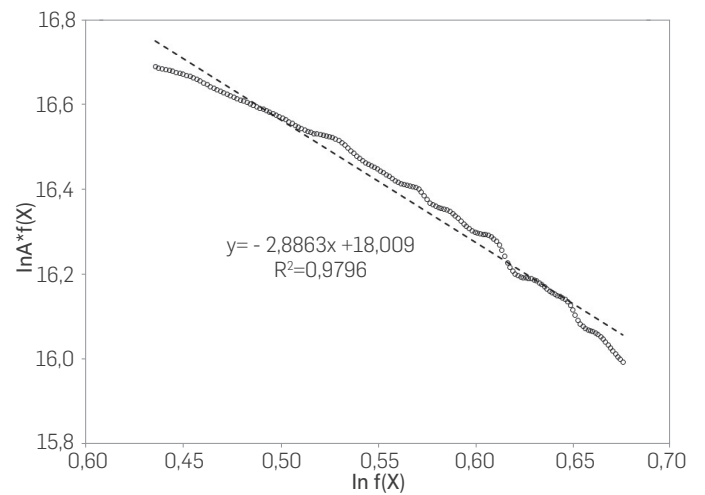


Figure A8. Graph for the approximation of the pre-exponential factor in the MTO region, Power Law (P3).

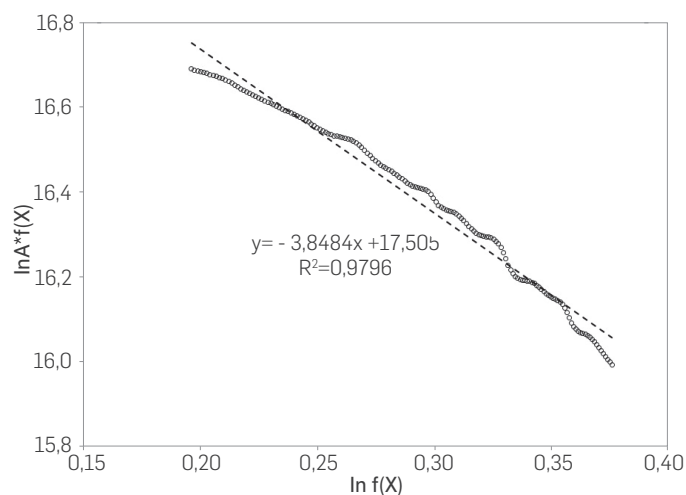


Figure A9. Graph for the approximation of the pre-exponential factor in the MTO region, Power Law (P2).

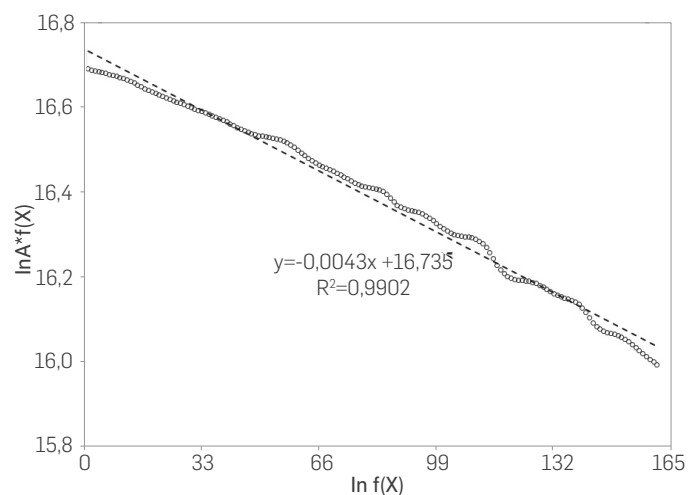


Figure A10. Graph for the approximation of the pre-exponential factor in the MTO region, Power Law (P2/3).

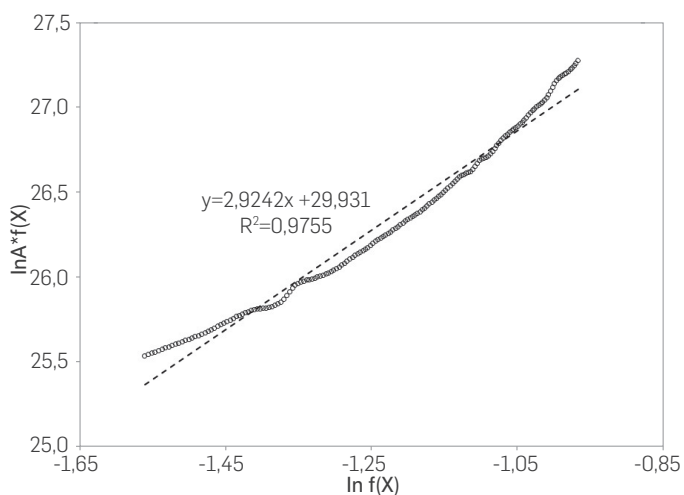


Figure A11. Graph for the approximation of the pre-exponential factor in the HTO region, Friedman (1964).

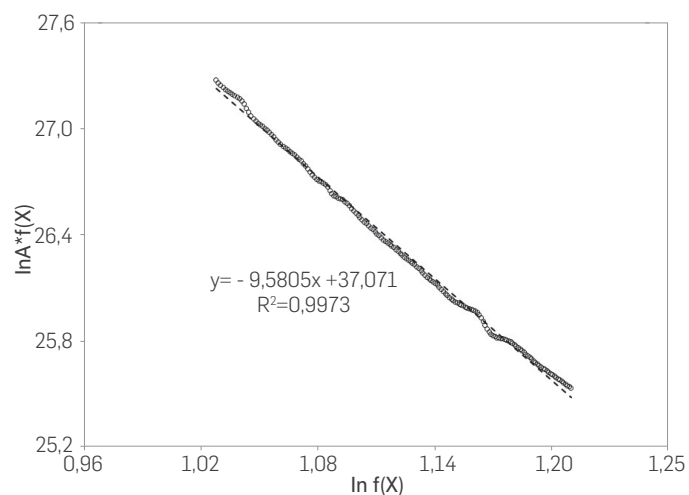


Figure A12. Graph for the approximation of the pre-exponential factor in the HTO region, Power Law (P4).

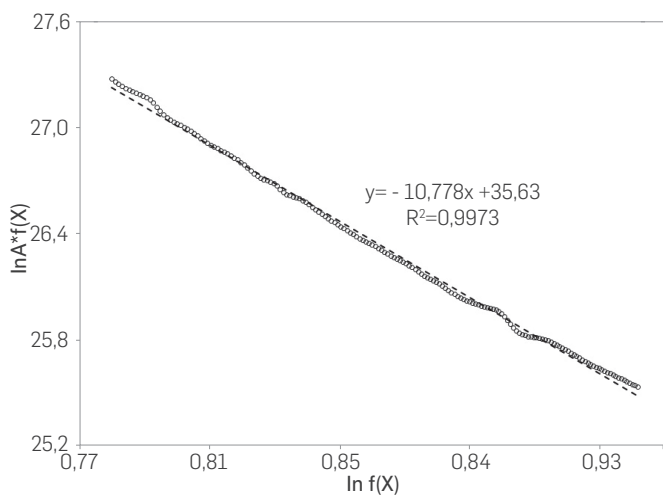


Figure A13. Graph for the approximation of the pre-exponential factor in the HTO region, Power Law (P3).

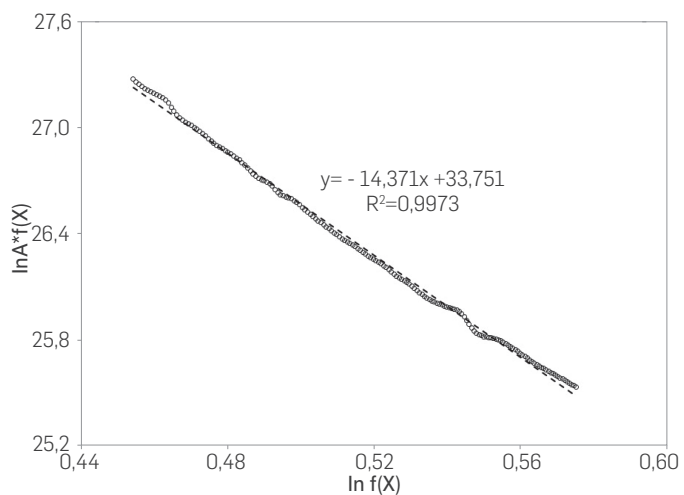


Figure A14. Graph for the approximation of the pre-exponential factor in the HTO region, Power Law (P2).

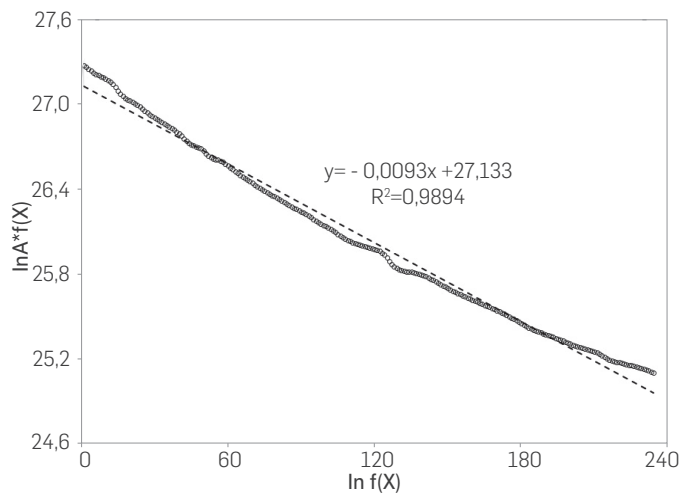


Figure A15. Graph for the approximation of the pre-exponential factor in the HTO region, Power Law (P2/3).



EcoAleshia

(Technological product developed by ICP)

Heavy crude washing tanks to increase the dehydration capacity and reduce the time required for crude oil to meet transport specs. Benefits: Savings of US\$ 122.5 million

EcoAleshia

(Producto tecnológico desarrollado por el ICP)

Tanques de lavado de crudo pesado para aumentar la capacidad de deshidratación y reducir los tiempos necesarios para dejar el crudo en especificaciones de transporte. Beneficios: US\$ 122.5 millones de ahorros