An Example of Falsified Kinetics by Diffusional Limitations in Gas-Solid Catalytic Reactions*

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This experiment explores the area of mass transfer resistances in heterogeneous catalysis using the light-off curves obtained with a catalytic membrane reactor for the evaluation of diffusional limitations. The catalytic reaction carried out over a Pt/γ-Al₂O₃ ceramic membrane is the combustion of methyl-ethyl ketone diluted in air at ppm level. Two different flow configurations for gas–solid contact are studied: flow-through and parallel flow to the membrane, showing different activation energies for the overall reaction rate. Therefore, by means of both single experiences the students are able to understand the kinetics falsification caused by diffusion.

NOMENCLATURE

C_{MEK} — MEK concentration at the reactor outlet, expressed in mol/dm^3

$C_{MEK,o}$ — Initial MEK concentration, expressed in mol/dm^3

E_{obs} — Activation energy for the apparent reaction rate expressed in J/mol

E_{true} — Activation energy for the intrinsic reaction rate expressed in J/mol

P — Permeance expressed in $mol/m^2.s.Pa$

F — Total mass flow rate, expressed in g/s

k_{obs} — Apparent kinetic reaction rate coefficient expressed in s^{-1}

$k_{o,obs}$ — Frequency factor of the kinetic rate coefficient expressed in s^{-1}

L — Membrane thickness expressed in m

M — Molecular weight of permeating species expressed in kg/mol

P_{av} — Average pressure across the membrane expressed in Pa

r — Average pore radius of the catalytic membrane expressed in m

R — Gas ideal constant expressed in J/mol.K

W — Platinum content in the catalytic membrane, expressed in g

X_{MEK} — MEK conversion, dimesionless

α — Knudsen contribution to permeation flux expressed in $mol/m^2.s.Pa$

β — Laminar contribution to permeation flux expressed in $mol/m^2.s.Pa^2$

ε_p — Membrane porosity, dimensionless

μ — Viscosity of permeating species, expressed in Pa.s

τ — Membrane tortuosity, dimensionless

INTRODUCTION

TRANSPORT PROCESSES (heat and mass transfer between the fluid and the solid or inside the porous solid) may influence the overall rate of heterogeneous catalytic reactions so that the conditions over the local reaction site do not correspond to those obtained in the bulk fluid around the catalyst particle [1, 2]. However, the laboratory study of mass transfer resistances in processes involving chemical reactors is seldom attempted at undergraduate level in chemical engineering departments. This is probably due to the time consuming nature of the preparations involved in setting up gas-solid catalytic reaction experiments varying the catalyst particle, gas velocity and reaction temperatures.

Seven steps could be involved in reactions on a solid particle:

1. Transport of reactants from the main stream to the catalyst pellet surface.
2. Transport of reactants in the catalyst pores.
3. Adsorption of reactants on the catalytic site.
4. Surface chemical reaction between adsorbed atoms or molecules.
5. Desorption of the products in the catalyst pores.
6. Transport of desorbed products to the particle surface.
7. Transport of products from the particle surface back to the main fluid stream.

The present work aims to emphasize the importance of the second step on the combustion of methyl-ethyl ketone (MEK) in air at ppm level. In the proposed experiment the reaction is carried out over a Pt/γ-Al₂O₃ catalyst dispersed in a porous membrane. These operational conditions have been selected because diffusional limitations are

especially significant in this system due to the low driving force (concentration gradient) available for diffusion. Pt supported over gamma alumina has been used since is the most widespread catalyst used to control gaseous emissions because of its high activity and selectivity to total combustion products [3]. Previous experiments carried out with the same reaction system show that the reaction rate is pseudo-first order with respect to the organic compound [4–6] and independent of oxygen concentration due to the large excess of air employed. The assumption of first-order kinetics for ketone combustion has been confirmed by other authors [7]. On the other hand, the specific configuration used (Pt catalyst on a porous alumina support) will allow two different flow arrangements (parallel and flow-through operation modes) that will be used to highlight the influence of diffusion resistances.

Catalytic combustion is perhaps the most promising alternative for the removal of Volatile Organic Compounds (VOC's) such as MEK from air streams, and in fact is a well-developed technology by pollution control companies. Therefore, students are faced with a realistic situation of the chemical engineering practice. With two single experiences carried out under forced flow of reactants through the membrane and flow of reactants parallel to the membrane, they will be able to determine experimentally the apparent activation energies for MEK combustion. This will allow them to understand the distortion of activation energies caused by interfering diffusion processes.

Among the pedagogical papers found on chemical reactor engineering for undergraduate students, Hesketh *et al.* [8] propose an automotive catalytic reaction engineering experiment to explore the area of heterogeneous catalysis. The experience is carried out with an automotive catalytic monolith, and is devoted to analyzing the light-off curves, to model the reactor determinate the propane reaction rates by numerical techniques. In a similar way, Saddawi and Schmitz [9] deal with the study of the methane oxidation rate on Pd/alumina cylindrical pellets in a tubular fixed-bed reactor equipped for an adjustable recycle flow in order to estimate the kinetic parameters under gradientless conditions. The utility of recycle reactors of this type for kinetic studies is also presented by Paspek *et al.* [10] who describe the use of such reactor for studying the kinetics of CO oxidation on supported platinum.

On the other hand, the experiments here proposed, will also introduce students to possible applications of inorganic membranes as reactors and to the main transport mechanisms through mesoporous membranes. While the use of inorganic membranes in catalytic reactors is still an emerging field, these are commonly used for micro and ultrafiltration in dairy, food, beverage and pharmaceutical applications. The students will be able to realize that inorganic membranes can also be used as effective contactors for gas-solid

catalytic reactions, in particular for situations when a high effectiveness of pollutant elimination is required. When use in the flow-through mode, the Pt/γ-Al$_2$O$_3$ catalytic membrane can be envisioned as a fixed bed reactor with a total length equal to the thickness of the membrane (around 1.5 mm) and a porous network where the catalyst is dispersed. In this gas-solid contactor, reactants are forced to flow perpendicular and then across the membrane wall; thus, the external film resistance is greatly reduced. With regard to the internal diffusion, the reduction of mass-transfer resistance stems from forced flow under the Knudsen-diffusion regime where molecule-pore wall collisions are predominant. Sometimes laminar flow can appear if the membrane has defects of macropore size, decreasing the contact effectiveness provided by the catalytic membrane. This hypothesis has been validated for VOC removal applications over Pt/γ-Al$_2$O$_3$ catalytic ceramic membranes as is confirmed by the excellent performance achieved in the Knudsen regime for complete combustion of toluene, methyl-ethyl ketone, hexane and their binary mixtures [4–6]. Alternatively, when the membrane is used in the parallel flow mode diffusion resistances are the same encountered in conventional systems (monolith reactors).

The experiment here described illustrates many aspects covered in the Chemical Kinetic and Reactor Design Course, therefore we suggest the experiment will be carried out in the Chemical Reaction Engineering Laboratory. However, the proposed laboratory session will also be valuable to those teaching graduate courses in catalysis and reaction engineering.

In summary, the pedagogical interests of this experience rely on the following engineering aspects:

- the student will be able to witness directly the falsification of the overall reaction rate by mass transfer resistances, a very common situation in gas-solid reaction process carried out at industrial scale;
- the student will familiarize himself with pollution control by means of catalytic combustion, which represents one of the most important technologies for VOC removal;
- the student will obtain a first contact with the field of inorganic membranes reactors, and the alternative gas-solid modes of contact that they can provide.

Additionally, the experiments planned involve tasks such as catalyst preparation, catalytic tests and kinetic parameter estimations, themselves valuable for students training.

EXPERIMENTAL PROCEDURES

Table 1 summarizes all the experimental tasks, time schedule and the deliverables proposed which

Table 1. Different tasks proposed for the two laboratory sessions

	Tasks	Time schedule	Knowledgements acquired	Deliverables
SESSION 1	Membrane preparation	One week	Preparation of the catalytic membrane active in VOC's combustion.	For a given experimental conditions (i.e. 1300 ppmV MEK and WHSV = 1200 s^{-1}) students have to calculate:
	Experimental set-up	30 min	Understanding how to make permeation/reaction experiments	Saturated air flow rate = Pure (dilution) air flow rate =
	Permeation test	60 min	Evaluation of the Knudsen contribution to the total permeation flux.	α (mol N$_2$/m^2.s.Pa) = β (mol N$_2$/m^2.s.Pa^2) =
	Exp. A: Flow-through mode	90 min	Elaboration of a Light-off curve in a catalytic membrane reactor.	$T_{50\%}$ (flow-through) = $T_{99\%}$ (flow-through) =
SESSION 2	Exp. B: Monolith flow	120 min	Comparison between two different contact modes. Evaluation of pore diffusion resistance.	$T_{50\%}$ (parallel flow) = $T_{99\%}$ (parallel flow) =
	Calculations	60 min + HW	Combining experience A and B, students will be able to evaluate the falsification of the activation energy by diffusion.	$k_{o, obs}$ (flow-through) = $E_{act, obs}$ (flow-through) = $k_{o, obs}$ (parallel flow) = $E_{act, obs}$ (parallel flow) =

have been distributed into two laboratory sessions, 3 hours each one. The experiments in the Chemical Reaction Engineering Laboratory Course are held in teams of two or three, depending on the number of students enrolled in the course. Every team will do the complete experiment, (since at the same time this experiment is running other teams are performing different experiments), and will evaluate the falsification of the activation energy.

Starting materials

The preparation of the catalytic membrane is an easy task, however the whole process takes a week, and we suggest that this task should be done by the laboratory technician in the lab. However the students should be given background information on membrane preparation, and invited to witness the key steps (impregnation, reduction) involved. The catalytic membrane was prepared from 10 mm o.d. commercial asymmetric ceramic tubes (US Filter) with a γ-Al$_2$O$_3$ separation layer 3 μm thickness and 5 nm as average pore diameter. The commercial membrane was inmersed in a hexachloroplatinic acid solution with a Pt content of 0.3 g/L prepared from a hexachloroplatinic acid solution (Sigma Aldrich) 8% wt in water. The impregnation during 120 hours results in a 0.02% wt of Pt per total membrane weight. The γ-Al$_2$O$_3$ layer plays the support role for Pt dispersion. After impregnation, the catalyst was activated by reduction in H$_2$ for 2 hours at 400°C in the same experimental device used for combustion experiences. More details on membrane preparation are given in [4–6]. The MEK for the experiments was obtained from Sigma Aldrich ACS chemical with a purity of 99+ % and the air was supplied by a blower, after filtering and regulated to 4 bar.

Experimental system

At the beginning of the class, the experimental set-up, shown in Fig. 1, should be explained and understood by the students. This must be equipped
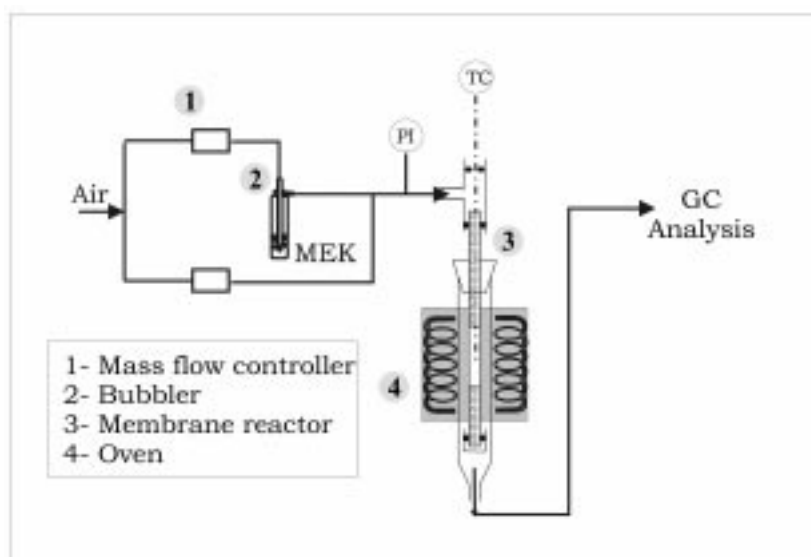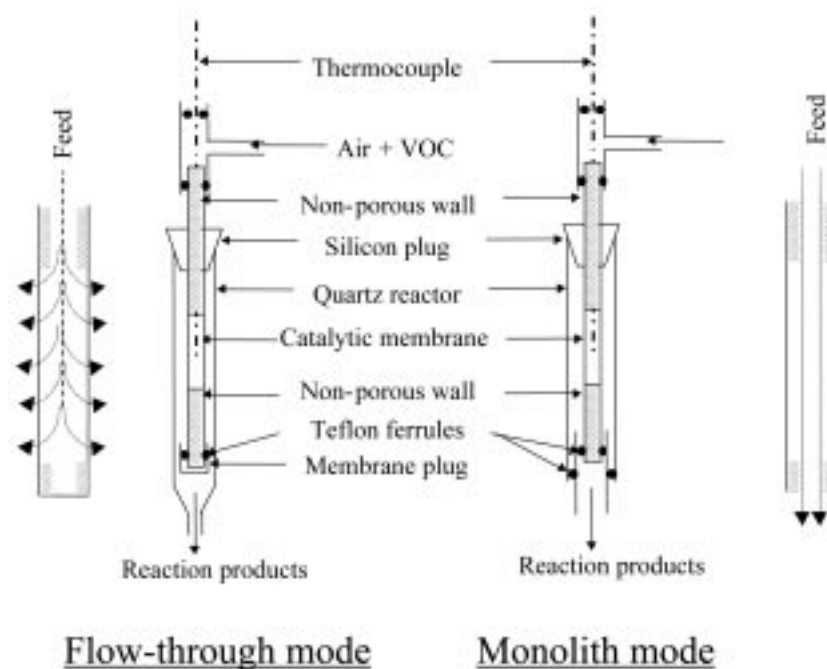

Fig. 1. Experimental set-up.

Fig. 2. Schematic diagram of the membrane reactor operating under different flow configurations.

with mass flow controllers, an electric oven, temperature and pressure transducers in the reaction zone and an on-line analysis system of the gaseous streams.

This reaction system has been described in previous works [4–6], and it mainly consists of:

- a feed section in which a mass flow controlled stream (air) is saturated at controlled temperature with the selected organic compound (MEK) and mixed with a second mass flow controlled air stream to give the desired VOC concentration;
- a reaction section, with a membrane reactor unit containing the catalytic membrane within a quartz shell;
- an analysis section, comprising an on-line gas chromatograph with FID and TCD detectors.

Permeation tests

Given the nature of the work undertaken, it is really important to know the membrane permeation characteristics, which must be obtained in separate permeation experiments. Once the students are familiar with membranes and the experimental set-up, they will conduct a permeation measurement. In order to do that, the membrane module is shown in Fig. 2. A simple reactor design is proposed to facilitate the assembly. This is built in quartz with stainless steel fittings and teflon ferrules are used in order to seal the membrane. For the permeation measurement the flow-through mode is used. The feed, N_2, enters the membrane tube side and then permeates outwards across the membrane wall. The permeated flow is measured by means of a bubble soap flowmeter, and pressure is indicated in the pressure transducer.

The measurements of single gas permeances over a γ-Al_2O_3 asymmetric commercial membrane has been already published [11]. Knudsen diffusion and laminar flow are the two main transport mechanisms for non adsorbable pure gases through mesoporous membranes (pore diameter between 2 and 50 nm) [12]:

$$P = 1.06 \cdot \frac{\varepsilon_p \cdot r}{L \cdot \tau \cdot \sqrt{M \cdot R \cdot T}}$$
$$+ 0.125 \cdot \frac{\varepsilon_p \cdot r^2}{L \cdot \tau \cdot \mu \cdot R \cdot T} \cdot P_{av} = \alpha + \beta \cdot P_{av} \quad (1)$$

where 'P' is the permeance defined as the permeation flux normalized per unit of time, area and pressure difference; and 'P_{av}' is the average pressure across the membrane. The α and β coefficients indicate the Knudsen and laminar contributions respectively to gas permeance, and they are normally evaluated at 1 bar as average pressure. Since in the Knudsen diffusion regime, that is gas permeation independent of average pressure, the probability of collisions between the molecules and the porous structure is maximised, if a catalyst has been dispersed on the pore walls of the inorganic membrane, a highly efficient use of the catalytic material should be possible (see Fig. 3).

In Fig. 4A, nitrogen permeance is plotted as a function of the average pressure across the membrane for two samples with 83.5% and 98.2% of Knudsen contribution, respectively. Operating preponderantly under Knudsen regime, gases permeate as a function of the square root of their molecular weight; that means, as shown in Fig. 4B, the heavier the gas the lower the permeation rate.
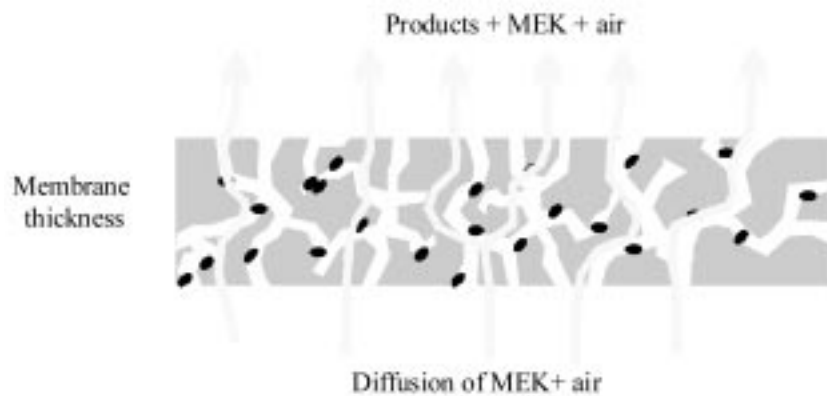
Products + MEK + air

Membrane
thickness

Diffusion of MEK + air

Fig. 3. Schematic drawing for the flow of reactants through the catalytic membrane.

(a)

$Y = 131.1 + 2.3 X$
$\alpha = 9.74 \ 10^{-6} \ mol/(m^2.s.Pa)$
$\beta = 1.71 \ 10^{-12} \ mol/(m^2.s.Pa^2)$ → 98.2% of Knudsen contribution

83.5% of Knudsen contribution ← $Y = 48.1 + 9.5 X$
$\alpha = 3.57 \ 10^{-6} \ mol/(m^2.s.Pa)$
$\beta = 7.06 \ 10^{-12} \ mol/(m^2.s.Pa^2)$

N_2 Permeance $(cm^3/cm^2.min.bar)$

Average Pressure (bar)

(b)

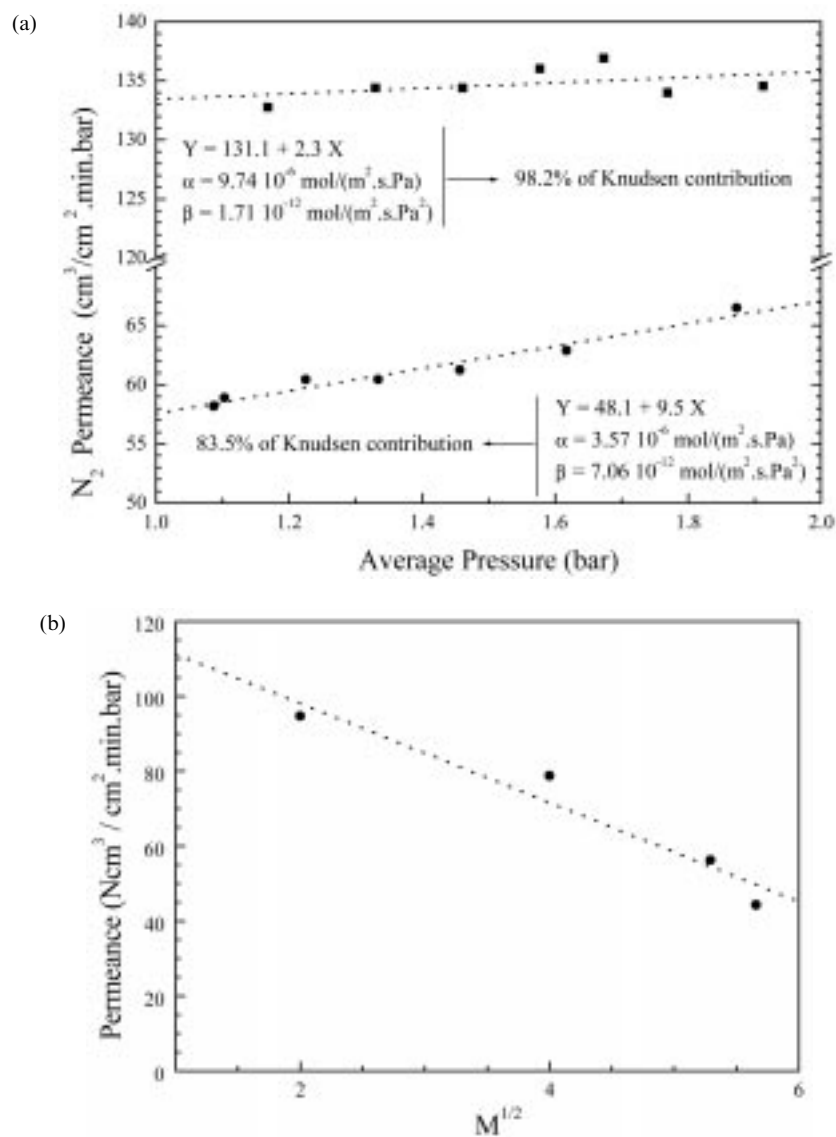Permeance $(Ncm^3 / cm^2.min.bar)$

$M^{1/2}$

Fig. 4. A: N_2 permeance as a function of average pressure across the membrane for two substrates with different percentages of Knudsen contribution; B: permeance as a function of the square root of the permeant molecular weight for a membrane with 99.7 % of Knudsen contribution.

Table 2. Conversion-temperature data for MEK combustion over Pt-γAl$_2$O$_3$ catalytic membrane operating under flow-through mode at different experimental conditions

Initial concentration (ppmV)	Total flow rate (Ncm^3/min)	WHSV(h^{-1})*	T$_{50\%}$ (°C)	T$_{99\%}$ (°C)
1775	485	710	142	181
1775	765	1120	155	199
3000	485	710	146	191
3000	765	1120	151	200

* Weight hourly space velocity calculated as the ratio of total mass flow rate and content of Pt.

REACTION EXPERIMENTS

Next tasks to be carried out are the reaction measurements under two flow configurations. The first exercises that can be proposed to the students are the calculation of saturated air/dilution air ratio necessary to achieve the desired MEK concentration at the reactor inlet and also the total flow rate necessary to operate at a given weight hourly space velocity (defined per platinum content).

Light-off curves

The students will accomplished the task of doing the complete light-off curves, in the experimental set-up previously described. The light-off curve is the conversion-temperature plot of a catalytic reaction. The activity of a catalyst in a reaction is often evaluated through this type of curve since such a measurement is easy to perform. With our experimental system is possible the examination of the MEK light-off curves at different initial concentrations and different flow rates.

To carry out the light-off experiments, the reactor is preheated at 100°C under reaction conditions and stabilized for 30 minutes until feed composition remains constant. After that, the temperature is increased by 1°C/min using a temperature controller, and simultaneously the product stream composition and its volumetric flow rate is measured on line. Considering 10 minutes as a reasonable time for CO$_2$ and MEK G.C. analysis, around ten to fifteen experimental points could be collected per catalytic run.

Experiment A: Catalytic Membrane Reactor Operating under Flow-Through Mode

The first configuration studied in reaction experiments is flow-through mode since the reactor is already prepared from the previous permeation measurements. It could be carried out just after the permeation test, and will last till total conversion of the organic. In the flow-through mode, the feed mixture enters the membrane tube side and then permeates outwards across the membrane wall. Table 2 shows the temperatures required for specific conversion levels at different experimental conditions with a catalytic membrane with an 83.5% of Knudsen contribution (estimated at 1 bar as average pressure), 2.51% of γ-Al$_2$O$_3$ and 0.17% Pt (both expressed per total membrane weight). It can be seen that combustion temperatures are slightly influenced by total flow rate but are less sensitive to initial MEK concentrations.

Experiment B: Catalytic Membrane Reactor Operating as a Monolith

At the beginning of the second lab session the reactant configuration is switched to the parallel flow-mode, see Fig. 2. Monolith operation is simulated by introducing a 4-mm outside diameter quartz cylinder axially in the 7-mm internal diameter membrane tube. In this way, a 1.5-mm wide annular channel is created between the external wall of the quartz cylinder and the internal wall of the membrane.

DATA TREATMENT AND RESULTS

Determination of reaction rate parameters

Once the students have collected all the experimental data for both contact modes, in the second part of this session lab they will also analyze their results. The catalytic membrane reactor can be simulated assuming a plug flow model and isothermal behavior. Considering a first-order kinetics with respect to MEK, the relationship between conversion and temperature is the following:

$$X_{MEK} = 1 - e^{\dfrac{-k_{obx} \cdot W}{F}} \qquad (2)$$

where:

$$X_{MEK} = 1 - \frac{C_{MEK}}{C_{MEK,o}} \qquad (3)$$

This dependence should be obvious if the students have just finished a chemical kinetics course. The results recompiled in Table 2 could be explained using the above equation. The light-off curves shown in Fig. 5 have also been obtained with the same catalytic membrane reactor. Operating under flow-through reactant mode (A type experiment) 99.8% of VOC conversion is achieved at 204°C, whereas in parallel flow (B type experiment) a 90.5% is obtained at 296°C. At 170°C the difference in conversion for both reactors (as read from the light-off curves) is 45 percentage points.

Apparent energy values are evaluated from experimental light-off curves once the apparent reaction rate constants 'k$_{obs}$' at different temperatures have been calculated according to Equation (4):

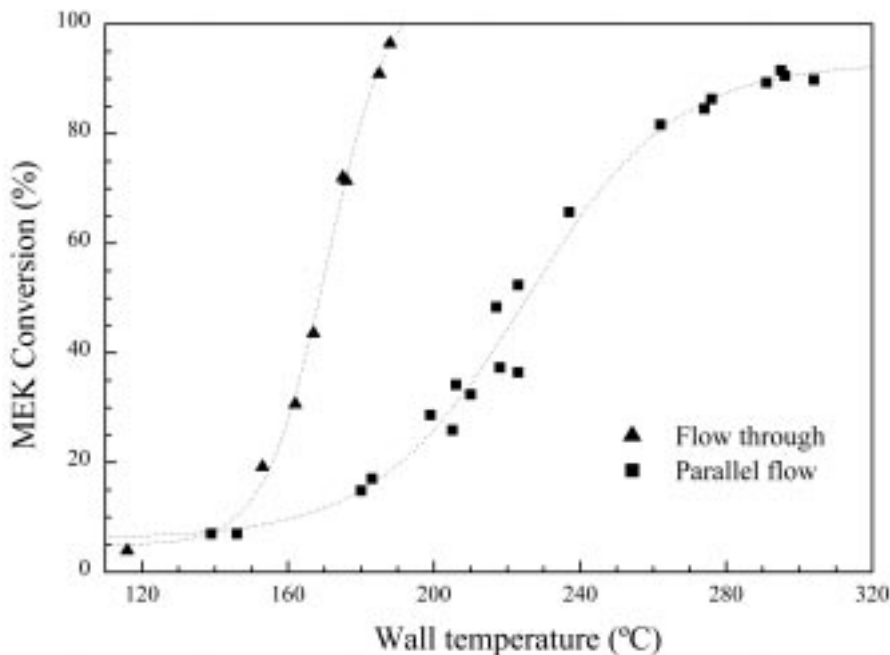$$k_{obx} = -[\ln(1 - X_{MEK})] \cdot \frac{F}{W} \qquad (4)$$

Fig. 5. Light off curves for MEK combustion over Pt-γAl$_2$O$_3$ catalytic membrane for two different flow configurations. Experimental conditions: 1300 ppmV of MEK and WHSV = 1190 g air/(g Pt h).

where:

$$k_{obs} = k_{o,observed} \cdot e^{-E_{obs}/R \cdot T} \qquad (5)$$

From the linear fit of $\ln(k_{obs})$ vs. T^{-1} for both types of configuration (see Fig. 6), the apparent activation energies that have been found appear in Table 3.

Since the activation energy for gas-phase reactions is normally rather high, say $80 \approx 240$ kJ/mol, while that for diffusion is small (about 5 kJ/mol at room temperature or 15 kJ/mol at 1000°C), the observed activation energy for reactions influenced by strong pore resistance is approximately one-half of the true activation energy:

$$E_{obs} \cong \frac{E_{true}}{2} \qquad (6)$$

The MEK combustion reaction rate carried out in parallel flow mode is strongly influenced by internal diffusion through the catalytic pores, as is evidenced by the activation energy observed (46716 J/mol), which is approximately one-half the activation energy obtained from flow-through mode experiments (91080 J/mol). Diffusional
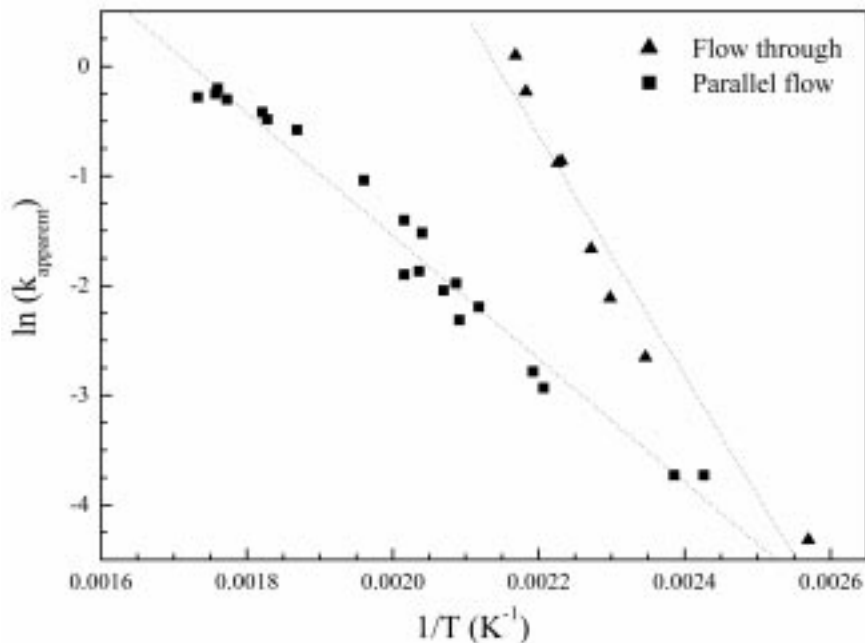

Fig. 6. Arrhenius fitting of the experimental results for MEK combustion under the conditions of Fig. 5.

Table 3. Apparent activation energies for MEK combustion over Pt-γAl$_2$O$_3$ catalytic membrane for different flow configurations

Flow-through	Apparent activation energy $= 91\,080$ J/mol	$R^2 = 0.9744$
Parallel flow	Apparent activation energy $= 46\,716$ J/mol	$R^2 = 0.9874$

limitations arise because reactant flows parallel to the membrane wall and molecules have to diffuse through the external film surrounding the catalyst particles, then into the stagnant gas that fills the pores before reaching the adsorption/reaction sites.

These results indicate that the flow-through reactant configuration notably improves the contact gas-solid phase allowing the operation under a kinetic regime. The value obtained for the activation energy under these conditions (91080 J/mol) must be rather close to the intrinsic activation energy for MEK combustion over Pt/ γ-Al$_2$O$_3$.

Table 4 shows the required temperatures to achieve 50% and 95% of MEK conversion over catalytic membranes with different percentages of Knudsen contribution but under identical experimental conditions. As it can be observed, the higher the α coefficient, the lower the temperature for MEK combustion according to a more efficient gas-solid contact. Additional experiments carried out over a catalytic membrane with a higher percentage of Knudsen contribution (>83%), could be undertaken to complete the laboratory session. These would be very helpful to validate the hypothesis of operation under kinetic regime control and to corroborate the intrinsic activation energy values.

ASSESSMENT

After carrying out the experiences, the students were asked to fill out the following questionnaire about the laboratory experiment. The survey is divided into four sections related to different aspects; suitability of the contents with regards to the theory, difficulty of the experiment, scheduling, and laboratory skills. In this section we also include a brief summary of the answers of a group of 25 undergraduate students.

1. Contents
This first set of questions intends to assess the relation between the content of theory lectures and the laboratory sessions.

Table 4. Light off temperatures for MEK combustion over Pt-γAl$_2$O$_3$ catalytic membranes with different percentages of Knudsen contribution. Experimental conditions: 2500 ppmV of MEK and WHSV = 1190 g air/(g Pt h)

% Knudsen contribution (α)	$T_{50\%}$	$T_{95\%}$
63%	170°C	197°C
83.5%	151°C	177°C

1.1. Do you think that the knowledge previously acquired in the subject 'Reaction kinetics' is enough to carry out and understand this experience in the lab?

　　1: Clearly not enough
　　2: Somewhat lacking
　　3: Barely enough
　　4: Clearly enough
　　5: Much more than enough

1.2. The relevant concepts in the laboratory experiment are also related to other subjects studied during the degree in Chemical Engineering courses in a _____ degree.

　　1: Inadequate
　　2: Low
　　3: Medium
　　4: High
　　5: Very High

1.3. Which laboratory experiment, (in this or in other courses), can you recall that is closely related to this experience?

All the students considered that their previous 'training' is clearly sufficient to carry out and understand this experiment and there is a high correlation between the subjects studied and the laboratory.

None of the students found a similar experiment in their previous laboratory background. This suggests that the proposed experiment is valuable as a complementary overlapping experience.

2. Difficulty
The second set of questions is associated to the difficulty of the experiment.

2.1. Do you think that the use of catalytic membrane reactor adds a _____ difficulty in the comprehension of the experiment?

　　1: Inadequate
　　2: Low
　　3: Medium
　　4: High
　　5: Very High

2.2. Which of the concepts used in this laboratory practice are completely new for you? Do you think that extra time is necessary to explain these concepts in more detail?

35% of the students stated that the membrane reactor concept is not sufficiently explained in theory lectures, however 65% of the students did not have any problem in understanding the membrane reactor concept.

The newest concept for the students was the light-off curve and some of them (30%) were concerned about the flow regimes. A possible solution could be a specific seminar about these concepts.

3. Time Schedule

The third set of questions deals with time, since this experiment uses two days, which it is not usual in other laboratory experiments.

3.1. The time consumed in the laboratory is _____ compared to the amount of concepts involved in the experiment.

1: Inadequate
2: Low
3: Average
4: High
5: Very High

3.2. Do you consider the time periods allowed in table 1 for the different tasks as ' _____ "?

1: Inadequate
2: Short
3: Adequate
4: Long
5: Too long

Most of the students (70%) consider that the time consumed is advantageous, (low), compared to the concepts achieved during the course of the experiment. 20% consider that there is not enough time for the completion of the experiment; 15% estimate that the time for the completion of the light-off curve it is not enough, and the rest consider that scheduling is correct.

4. Skills in the Laboratory

In this section the students evaluate their skills in the lab.

4.1. Do you think that the previously acquired experimental skills are adequate to operate with an experimental set-up like this?

1: Inadequate
2: Low
3: Medium
4: Adequate
5: Very Adequate

4.2. The procedure followed in the laboratory, was difficult to catch on. Mark form 1: Inadequate to 5: Very adequate.

1: Inadequate
2: Low
3: Medium
4: Adequate
5: Very Adequate

4.3. Dou you think that could be interesting for the student the preparation of the catalytic membrane?

4.4. Which one is the most difficult part in the experimental set-up? Why?

20% believe that the already known skills are high, 60% think that their skills are medium and a 20% conclude that are low. 90% of the students consider that it is no necessary to prepare the catalytic membrane, nevertheless 10% affirm that would enjoy the preparation of the membrane. The experimental set-up and procedure are fully understood by the students.

ASSESSMENT RESULTS

The numerical results of the questionnaire from a group of 25 undergraduate students are plotted in Fig. 7. In general, the average value of the answers is higher than 3; therefore we can conclude that students find the laboratory experience highly satisfactory. The exception is for answer number 3.1 evaluated with 2.5, indicating that the time spent in the laboratory is worthy due to the knowledge acquired during the session. Concerning contents, it can be concluded that the paper contributes to the base of knowledge in chemical
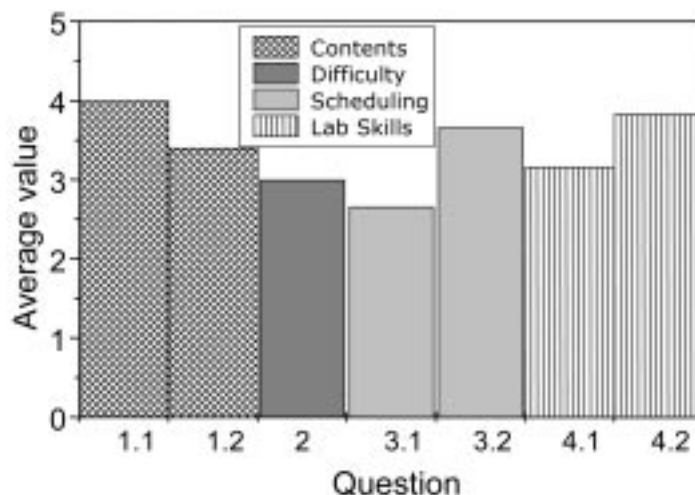
Fig. 7. Average values for the answers of the questionnaire.

reaction engineering experimental methods because none of the students found a similar experiment in previous laboratory sessions. Moreover, the previous training and experimental skills of the 80% of students are clearly sufficient to master adequately the experiment's objectives. Although the experimental procedure here proposed helps to the comprehension of the concepts involved; however, some of them (i.e. light-off curve, Knudsen diffusion and laminar flow through porous media) require an specific seminar for further explanations.

CONCLUSIONS

An experimental reaction system to carry out combustion of methyl-ethyl ketone over a Pt/γ-Al$_2$O$_3$ membrane can be used to emphasize the diffusional distortion to activation energies as a consequence of the controlling resistance to reactant transport from the gas phase inside the catalyst pores to the active centers inside the pores. A simple reactor design built in quartz allows the proposed experiments to be carried out.

Summarizing, the chemical engineering students are able to:

- operate with an inorganic membrane;
- modify the membrane to attain catalytic activity for VOC combustion;
- carry out light-off experiments measuring gas-phase concentrations and flowrates;
- model the reactor and determine the apparent activation energies.

Experimentally they are faced with a fully equipped reaction system which is of interest for their training. The experimental results obtained from two typical combustion curves over the same catalytic membrane reactor but different reactant flow configurations are enough to evidence the falsification of the reaction rate by internal diffusion resistances. From this data, final year undergraduate students, will be able to determine the intrinsic activation energy for MEK combustion over a Pt/γ-Al$_2$O$_3$ membrane (flow-through mode), and also the apparent activation energy under an internal diffusion regime (parallel flow or monolith mode).

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