



Assessing Metacognitive Awareness during Problem-Solving in a Kinetics and Homogeneous Reactor Design Course

Dr. Nelly Ramirez-Corona, Universidad de las Americas Puebla

Dr. Nelly Ramírez-Corona is currently a full-time professor of Chemical Engineering at Chemical, Environmental and Food Engineering Department, Universidad de las Américas at Puebla, México. Her teaching experience is in the area of Process Dynamics and Control, Kinetics, Catalysis and Reactor Design. She did her undergraduate studies in Chemical Engineering at the Universidad Autónoma de Tlaxcala, México, and his Master and Doctoral studies at the Instituto Tecnológico de Celaya, México. Her research interests are in the field of Process Systems Engineering, and include the analysis and design of thermally coupled and alternative distillation configurations, the design of nonideal distillation systems and the synthesis, optimization and control of chemical process with recycles streams.

Prof. Ramirez Apud Lopez Zaira, Universidad de las Americas Puebla

Zaira Ramírez is Science, Engineering, and Technology Education Ph.D. Student at Universidad de las Americas Puebla in Mexico. She teaches ethics and development complex thinking skills related courses. Her research interests include faculty development, outcomes assessment, and creating effective learning environments.

Prof. Aurelio Lopez-Malo, Universidad de las Americas Puebla

Dr. Enrique Palou, Universidad de las Americas Puebla

Professor Dr. Palou is director of the Center for Science, Engineering, and Technology Education in the Department of Chemical, Food, and Environmental Engineering at Universidad de las Americas Puebla in Mexico. He teaches engineering, food science, and education related courses. His research interests include emerging technologies for food processing, creating effective learning environments, using tablet PCs and associated technologies to enhance the development of 21st century expertise in engineering students, and building rigorous research capacity in science, engineering and technology education.

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Abstract

Since practicing engineers are hired, retained, and rewarded for solving problems, engineering students should learn how to solve workplace problems^{1,2}. Therefore, we designed and implemented several problem-solving learning environments (PSLEs) for the junior course entitled Kinetics and Homogeneous Reactor Design at *Universidad de las Américas Puebla*³.

Metacognition has been shown to be important for the solution of more open-ended and well-structured problems². Flavell^{4,5} distinguished two characteristics of metacognition: *knowledge of cognition* (KC) and *regulation of cognition* (RC). In order to support student metacognitive processing while learning to solve kinetics and homogeneous reactor design problems, the instructor created a supportive social environment in the course and inserted a series of question prompts during PSLEs, as a form of coaching where the problem to be solved was represented as a case, and cases were used in various ways (worked examples, case studies, structural analogues, prior experiences, alternative perspectives, and simulations) as instructional supports.

The Metacognitive Awareness Inventory (MAI) designed by Schraw and Dennison⁶ was utilized as a pre- (first day of classes) post- (last day of classes) test. MAI is a 52-item inventory to measure adults' metacognitive awareness. Items are classified into eight subcomponents subsumed under two broader categories, KC and RC. Furthermore, in order to assess metacognitive awareness during problem-solving activities, students had to answer the corresponding problem as well as approximately 2-3 embedded problem-solving prompts (from Jonassen²) and 4-6 embedded metacognitive prompts (from MAI).

Results for the pre-post MAI exhibited a significant ($p < 0.05$) increase in student metacognitive awareness. This increase was also noticed by means of the embedded MAI prompts while solving different kinds of problems (such as story problems, decision-making problems, troubleshooting, and design problems) throughout the course, in which students also improved the quality of their embedded problem-solving answers and corresponding grades.

Promoting metacognitive awareness and skills could be a valuable method for improving learning and student performance during kinetics and homogeneous reactor design problem-solving, as has been previously reported for professional educators⁷ and dental hygiene students⁸.

Introduction

Practicing engineers are hired, retained, and rewarded for solving problems. Usually workplace engineering problems are substantively different from the kinds of problems that engineering students most often solve in the classroom; therefore, learning to solve classroom problems does not necessarily prepare engineering students to solve workplace problems^{1,2}. Therefore, the primary purpose of engineering education should be to engage and support learning to solve problems¹⁻³. Hence, we designed and implemented several problem-solving learning environments (PSLEs), a term that represents problem-solving instruction in a more open-ended way than problem-based learning².

Problem solving is a schema-based activity^{1-3,9}. That is, in order to solve problems, learners must construct schemas for problems. Constructing models of problems greatly facilitates schema development. Having constructed a robust schema for different kinds of problems, learners are better able to transfer their problem-solving skills. Learning to solve problems requires practice in solving problems, not learning about problem solving². PSLEs assume that learners must engage with problems and attempt to construct schemas of problems, learn about their complexity, and mentally wrestle with alternative solutions^{2,9}. Hence, we built PSLEs to engage and support students in learning how to solve problems by practicing solving problems³.

PSLEs were developed by following the design activities proposed by Jonassen²: 1) First we interacted with the teacher of the studied course to identify and articulate problems relevant to the discipline; 2) We analyzed problems, first by creating a causal model of the problem space; 3) Then we conducted an activity theory analysis to identify the historical, cultural, experiential factors that affect problem solving on the context chosen⁶; 4) Determined what kind of problems were each one of them; 5) Constructed case supports and cognitive scaffolds for each problem type; 6) To then construct each PSLE that included some combination of case components and cognitive strategies; 7) Finally implemented and assessed the effects of the developed PSLEs. Preliminary results are discussed elsewhere³.

Problems vary in different ways, so different kinds of problems call on different conceptions and skills; consequently learning methods should also vary^{1-2,9}. Based on those differences among problems, different kinds of reaction engineering problems were developed, such as story problems, troubleshooting/diagnosis problems, decision-making problems, and design problems.

Making decisions to manage process operation conditions is the most common problem that engineers have to face in real life, and those decisions have to be based on proper knowledge and prediction of the process performance. In chemical engineering, reactor design is considered as a fundamental knowledge since reactor conditions define the operation settings for most of the other units of the process¹⁰⁻¹². At *Universidad de las Américas Puebla*, chemical engineering

students develop the basic knowledge and skills to design and operate chemical reactors in the junior course Kinetics and Homogeneous Reactor Design course (IQ-407), which is the first one of a two course sequence. Learning outcomes for IQ-407 include that students will be able to: 1) determine reaction rate expressions from experimental data; 2) use basic concepts of kinetic, mass and energy balances, as well as principles from thermodynamics to design ideal homogeneous reactors; and 3) assess and propose reactor operation conditions to achieve a specific objective³.

In a preliminary study regarding the implementation and assessment of PSLEs for IQ-407, which was exploratory and intended to provide formative evaluation along the course, our findings³ indicated that students developed several metacognitive skills along the problem solving process, which we considered as important as finding the “right” solution, especially for open-ended (ill-structured) problems. Therefore the main goal of this work was to further study the development of IQ-407 students’ metacognitive skills along their problem solving process.

Flavell^{4,5} distinguished two characteristics of metacognition: *knowledge of cognition* (KC) and *regulation of cognition* (RC). KC comprises three sub-processes that facilitate the reflective aspect of metacognition: declarative knowledge (knowledge about self and about strategies), procedural knowledge (knowledge about how to use strategies), and conditional knowledge (knowledge about when and why to use strategies). KC includes knowledge of task, strategy, and personal variables. RC covers five areas: planning (goal setting), information management (organizing), monitoring (assessment of one’s learning and strategy), debugging (strategies used to correct errors) and evaluation (analysis of performance and strategy effectiveness after a learning episode). RC includes the ability to monitor one’s comprehension and to control one’s learning activities. The self-regulation factor of metacognition describes activities that regulate and oversee learning such as planning (predicting outcomes, scheduling strategies) and problem-monitoring activities (monitoring, testing, revising and rescheduling during learning). Self-regulation also involves evaluation. That is, metacognitive knowledge includes knowledge of the skills required by different tasks, strategic knowledge (knowledge of alternative learning strategies and when to use them) and self-knowledge (knowledge of one’s abilities and the abilities of others)⁴⁻⁸.

Methodology

Along the fall 2012 semester we implemented a series of PSLEs with the aim of developing specific skills and/or to improve the understanding of key concepts related to chemical reactor engineering. Finally, a design (open-ended) problem was used to assess students’ transfer of expected course learning outcomes and problem solving skills (this transfer of learning is not part of this research and will not be presented here).

In order to assess students' metacognition awareness, the Metacognitive Awareness Inventory (MAI) designed by Schraw and Dennison⁶ was utilized as a pre- (first day of classes) post- (last day of classes) test. MAI is a 52-item inventory to measure adults' metacognitive awareness (see appendix A). Items are classified into eight subcomponents subsumed under two broader categories, KC and RC. Furthermore, in order to assess metacognitive awareness during problem-solving activities, students had to answer the corresponding problem as well as approximately 2-3 embedded problem-solving prompts (from Jonassen²) and 6-10 embedded metacognitive prompts (from MAI). No additional instruction on metacognition was given. IQ-407 class population was integrated by eight students (two women), thus monitoring their individual progress along the course was relatively easy.

Instructional materials were available on the course website and students and instructor were using Tablet PC's with selected instructional platforms for PSLEs implementation, which allowed strong real-time interaction among students and instructor during classes. In order to support student metacognitive processing while learning to solve kinetics and homogeneous reactor design problems, the instructor created a supportive social environment in the course and inserted a series of question prompts during PSLEs, as a form of coaching where the problem to be solved was represented as a case, and cases were used in various ways² (worked examples, case studies, structural analogues, prior experiences, alternative perspectives, and simulations) as instructional supports.

Examples of PSLEs implementation in IQ-407

Story problems are commonly used for enhancing variable recognition and the use of algorithms. This kind of problems was utilized to support students' learning to describe stoichiometric relationships as mathematical models. If the degrees of freedom are specified, obtained models can be used to determine unknown variables. An example (that was assessed for this study during the course) is described in Figure 1.

<p>Problem 1</p> <p>The following reactions are taking place simultaneously in gas phase.</p> $A \Leftrightarrow B + C$ $B \Leftrightarrow D + C$ <p>The equilibrium constant for each one is 6 and 4, respectively. The total pressure is 2 bar and it remains constant along the process. Determine the partial pressure for each component at equilibrium, if reactor is fed exclusively with A and it is operated isothermally.</p>

Figure 1. Story problem example (adapted from Tiscareño¹²).

Students were asked to describe shortly the procedure they utilized to calculate the required parameters. In the same activity they answered 7 items from MAI (items numbered 10, 12, 16, 17, 20, 32, and 46 on Appendix A) related to *knowledge of cognition*, particularly associated with declarative knowledge.

An example (that was assessed for this study during the course) of a troubleshooting/diagnosis problem is described in Figure 2. In this case students were asked to manipulate a process variable to achieve a specified conversion. Once more, students had to describe the methodology they employed and 9 MAI items (they had to answer items numbered 3, 14, 15, 18, 26, 27, 29, 33, and 35 on Appendix A) related to *knowledge of cognition*, several of them particularly associated with procedural and conditional knowledge, were used as a form of coaching.

Problem 2

Mixed flow reactor (CSTR) is used for carrying out the next first order reversible reaction:

$$A \rightleftharpoons B$$

The reactant A is fed with a composition of 1 M. The equilibrium conversion is 66.7% and the actual conversion is 33.3%. We are looking for raising the actual conversion to 50%. You are asked to determine how we must adjust the feed flowrate to achieve that goal.

Figure 2. Troubleshooting/diagnosis problem example (adapted from Levenspiel¹¹).

Problem 3

There are two reactors available for installation, the first one a CSTR with a 5 m³ volume and the second one a PFR with 2 m³ volume to process 80 L/min containing 0.5 M of A and 0.1 M of B. The desired product C may continue reacting to a side product with no commercial value. The important reactions are:

$$A + \frac{1}{2}B \rightarrow C$$

$$C + \frac{1}{2}B \rightarrow D$$

The kinetic expression for each reaction, which are referred to component B, are:

$$(-r_B)_1 = k_1 C_A C_B^{0.5} = 0.0068 \left[\frac{L^{0.5}}{\text{min mole}^{0.5}} \right] C_A C_B^{0.5}$$

$$(-r_B)_2 = k_2 C_B C_C = 0.0745 \left[\frac{L}{\text{min mole}} \right] C_B C_C$$

Determine the proper order to install both reactors. Justify your decision.

Figure 3. Decision-making problem example (adapted from Tiscareño¹²).

An example (that was assessed for this study during the course) of a decision-making problem is described in Figure 3. In this case students had to decide (and justify their decision) the order for placing both reactors, based on their reactor and kinetic knowledge, as well as 10 MAI items (they had to answer items numbered 2, 6, 8, 11, 21, 22, 23, 34, 41, and 42 on Appendix A) related to *regulation of cognition*, most of them particularly associated with planning and monitoring, were used as a form of coaching.

Additionally, a design problem (Appendix B) was implemented as final project, which was assigned for teamwork (groups of two students) on the last week of the semester and students had a period of one week to develop their proposal, which they presented as their final exam. The same chemical process was used for all teams, but a particular study case was assigned for each one. Students were asked to carry out a presentation of their problem solution methodology, the obtained results and their final conclusions. The presentation was videotaped to be further examined. Analysis of these presentations will allow us to identify students' abilities to solve complex problems, as well as their argumentative skills. Since the problem is open-ended, a number of alternative solutions can be generated, for this reason students had to define a methodology to constrain the number of scenarios to be evaluated. Along students' presentations, the instructor conducted some prompts to encourage students' argumentation for supporting their selections. As stated before, this final project will be utilized to assess students' transfer of expected course learning outcomes and problem solving skills. However, this transfer of learning study is not part of this research and will not be presented here).

Results and discussion

Pre-post MAI total mean scores are presented in Figure 4. The blue bars represent *knowledge of cognition* while the red bars display the *regulation of cognition* results. Global MAI results are summarized by the green bars. It is clear that significant progress ($p < 0.05$) in students' global metacognitive awareness, as well as specifically *knowledge of cognition*, and *regulation of cognition* were achieved. At Florida State University the MAI was used as a measuring tool in a research that examined the effects of teaching metacognitive strategies to 60 students in a photography class. The results of the MAI exhibited an increase in the total mean score, from 65 to 68 out of 100. The MAI was answered before and after assignments with instructions and practice in reflection, planning and evaluation¹⁵.

MAI has also been used at the faculty of Odontology in Malmö University in Sweden, for a project focusing on students' proficiency to learn in a problem-based curriculum. Students took part in different workshops; they watched a tutorial that was followed by discussions and worked in small groups designing cases. After the workshops the MAI-data from students taking part in the project was compared to data from other students, displaying significantly higher ($p < 0.10$) metacognitive awareness amongst students taking part in the project⁸. In a following project,

using a modified model of the PBL-tutorial, Malmö students increased on the MAI mean total score from 62.1 to 68.6 out of 100⁸.

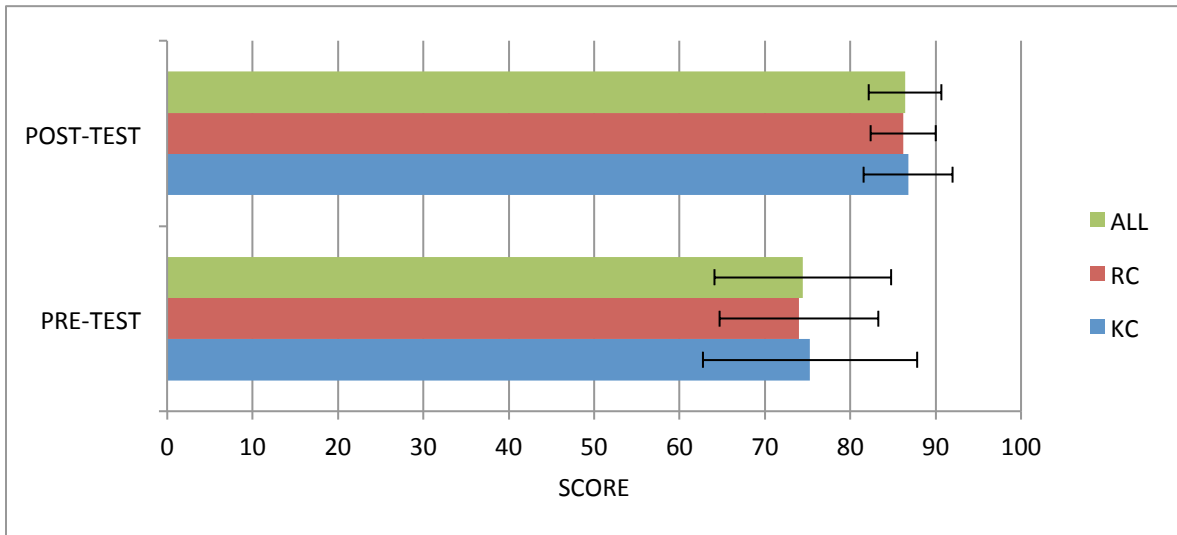


Figure 4. Pre-post (first-last day of classes) students' global *Metacognitive Awareness Inventory* (ALL), *knowledge of cognition* (KC), and *regulation of cognition* (RC) results (n = 8).

Furthermore, students' individual mean scores were also analyzed. Figure 5 displays every student pre-post metacognitive awareness (in which his/her *knowledge of cognition* and *regulation of cognition* are included).

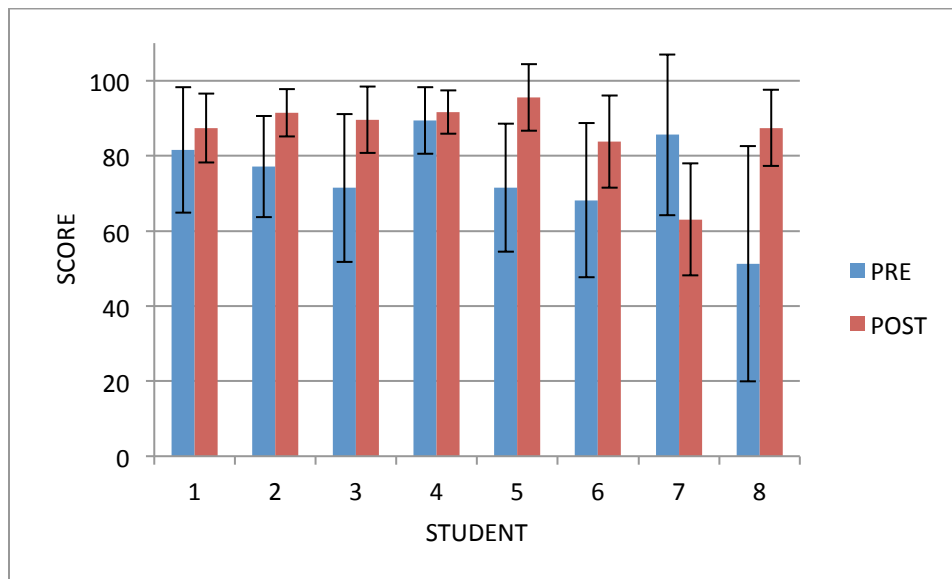


Figure 5. Pre-post (first-last day of classes) individual student *Metacognitive Awareness Inventory* results.

It can be observed in Figure 5 that the studied approach helped almost every student, regardless of its gender or academic strength. Students (numbered 1 and 4) that achieved high scores in the pre-test obtained minor gains in metacognitive awareness scores in their post-tests while students (numbered 2, 3, 5, 6, and 8) that achieved lower scores in the pre-test obtained larger gains in metacognitive awareness scores in their post-tests. In general, higher progresses were observed for lower pre-test MAI scores. Student numbered 7 is the only one that decreased its metacognitive awareness score, we think that he over-assessed its metacognitive awareness in the pre-test and after a whole semester of practicing, recognized its limitations regarding his metacognition skills.

Along the semester, several MAI items were included within the problem-solving activities. In order to analyze the development of student's metacognitive awareness, the obtained results were compared to those obtained in the MAI pre-test corresponding items. According to our findings, a significant progress was observed through each activity. A summary of results is presented in Table 1. As can be seen, the *knowledge of cognition* of students steadily and significantly increased from pre-test to problems 1 ($p < 0.05$) and 2 ($p < 0.01$), being more significant as course (and metacognitive awareness of students) progressed. Furthermore, *regulation of cognition* of students significantly ($p < 0.10$) increased from pre-test to problem 3, which was applied close to the end of the course.

Table 1. Comparisons of students' *Metacognitive Awareness Inventory* (MAI) mean scores regarding MAI prompts' scores (KC: *knowledge of cognition* and RC: *regulation of cognition*) to MAI pre-test corresponding items' scores for each studied problem (1: story problem, 2: troubleshooting/diagnosis problem, 3: decision-making problem).

		Mean	Standard deviation	Significant* difference at $p <$
KC	<i>Pre-test</i>	75.27	12.54	
	Problem 1	80.44	20.31	0.0203
	Problem 2	87.92	9.47	0.0001
RC	<i>Pre-test</i>	74.00	9.29	
	Problem 3	80.78	18.73	0.0699

*Significant results by using Mann-Whitney Test

As stated before, the final design problem was implemented to assess students' chemical reactor problem-solving skills, as well as their metacognitive awareness to improve students' learning. The analysis of the proposed solution allowed instructor to identify students' ability to solve workplace problems. Students were able to organize and recognize useful information, get the missing data, develop a mathematical model to represent the problem, evaluate different scenarios to achieve the specified goal, choose operation conditions and suggest alternative solutions. Since different study cases were assigned for each team, students had the opportunity

to discuss different design and operation conditions, even some comparison between study cases were conducted by them. Furthermore, every report included a detailed description of their problem solving process. As can be noted in Appendix B, the course instructor did not require that description. This is an indication about how students enhanced their metacognitive awareness along the course, and how these skills, along with the acquired chemical reactor design knowledge, were used by students to improve their learning processes. As part of the design project presentations, discussion was encouraged regarding the problem solving strategies used by a particular team as well as on the problem solution itself. Therefore, opportunity was provided through the given format for students to not only assess their own problem solving ability, but to also be exposed to the problem solving strategies employed by other groups. This exposure also benefits the students.

Final remarks

Results for the pre-post MAI show a significant ($p < 0.05$) increase in student metacognitive awareness. This increase was also noticed by means of the embedded MAI prompts while solving different kinds of problems (such as a story problem, a troubleshooting/diagnosis problem, a decision-making problem, and a design problem) throughout the course, in which students also improved the quality of their embedded problem-solving answers and corresponding grades. It is important to note that with respect to the students, no resistance to this approach was noticed.

Furthermore, instructor reflection about the implemented PSLEs allowed her to be aware of these metacognitive processes, their impact on her students' learning, and its potential in order to incorporate more of such activities in several senior courses in chemical engineering. She realized that instructional activities implemented along each problem enhanced students' conceptual and procedural knowledge, promoting students' metacognitive awareness. Based on the assessed problems and especially on the final project, it was noted by her that encouraging these skills is valuable to improve learning and student problem-solving performance.

Acknowledgments

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APPENDIX A: Metacognitive Awareness Inventory (MAI)*

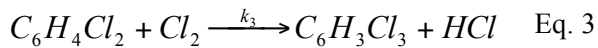
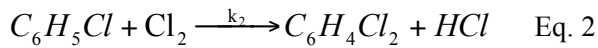
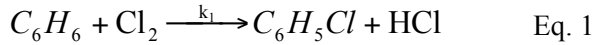
1. I ask myself periodically if I am meeting my goals. (M)
2. I consider several alternatives to a problem before I answer. (M)
3. I try to use strategies that have worked in the past. (PK)
4. I pace myself while learning in order to have enough time. (P)
5. I understand my intellectual strengths and weaknesses. (DK)
6. I think about what I really need to learn before I begin a task. (P)
7. I know how well I did once I finish a test. (E)
8. I set specific goals before I begin a task. (P)
9. I slow down when I encounter important information. (IMS)
10. I know what kind of information is most important to learn. (DK)
11. I ask myself if I have considered all options when solving a problem. (M)
12. I am good at organizing information. (DK)
13. I consciously focus my attention on important information. (IMS)
14. I have a specific purpose for each strategy I use. (PK)
15. I learn best when I know something about the topic. (CK)
16. I know what the teacher expects me to learn. (DK)
17. I am good at remembering information. (DK)
18. I use different learning strategies depending on the situation. (CK)
19. I ask myself if there was an easier way to do things after I finish a task. (E)
20. I have control over how well I learn. (DK)
21. I periodically review to help me understand important relationships. (M)
22. I ask myself questions about the material before I begin. (P)
23. I think of several ways to solve a problem and choose the best one. (P)
24. I summarize what I've learned after I finish. (E)
25. I ask others for help when I don't understand something. (DS)
26. I can motivate myself to learn when I need to. (CK)
27. I am aware of what strategies I use when I study. (PK)
28. I find myself analyzing the usefulness of strategies while I study. (M)
29. I use my intellectual strengths to compensate for my weaknesses. (CK)
30. I focus on the meaning and significance of new information. (IMS)
31. I create my own examples to make information more meaningful. (IMS)
32. I am a good judge of how well I understand something. (DK)
33. I find myself using helpful learning strategies automatically. (PK)
34. I find myself pausing regularly to check my comprehension. (M)
35. I know when each strategy I use will be most effective. (CK)
36. I ask myself how well I accomplish my goals once I'm finished. (E)
37. I draw pictures or diagrams to help me understand while learning. (IMS)
38. I ask myself if I have considered all options after I solve a problem. (E)
39. I try to translate new information into my own words. (IMS)
40. I change strategies when I fail to understand. (DS)
41. I use the organizational structure of the text to help me learn. (IMS)
42. I read instructions carefully before I begin a task. (P)
43. I ask myself if what I'm reading is related to what I already know. (IMS)
44. I reevaluate my assumptions when I get confused. (DS)
45. I organize my time to best accomplish my goals. (P)
46. I learn more when I am interested in the topic. (DK)
47. I try to break studying down into smaller steps. (IMS)
48. I focus on overall meaning rather than specifics. (IMS)
49. I ask myself questions about how well I am doing while I am learning something new. (M)
50. I ask myself if I learned as much as I could have once I finish a task. (E)
51. I stop and go back over new information that is not clear. (DS)
52. I stop and reread when I get confused. (DS)

Knowledge of cognition (KC): declarative knowledge (DK), procedural knowledge (PK), and conditional knowledge (CK). *Regulation of cognition* (RC): planning (P), information management strategies (IMS), monitoring (M), debugging strategies (DS), and evaluation (E).

* Schraw, G. and Dennison, R. S. 1994. Assessing metacognitive awareness. *Contemporary Educational Psychology*, 19: 460-475.

APPENDIX B: FINAL PROJECT

Chlorobenzene is obtained at industrial scale from the reaction between liquid benzene and gaseous chlorine (for design purposes system can be modeled as liquid homogenous reaction), catalyzed at moderate conditions of pressure and temperature. The reactor yield (conversion and selectivity) depends on reactants feed ratio, due to further chlorination reactions can take place and other byproducts can be generated. The referred reactions are given as:



For this work, the third reaction can be neglected. Table A displays the kinetic parameters for the reaction rate expressions.

Table A. Kinetic parameters for reaction rate expressions (Bodman, 1968)¹³

Reaction	Reaction kinetic (kmol/s*m ³)	Frequency factor A	Activation energy (BTU/lbmol)
$C_6H_6 + Cl_2 \xrightarrow{k_1} C_6H_5Cl + HCl$	$r_1 = A_1 C_{C_6H_6} C_{Cl_2} e^{-E/RT}$	5.9833×10^{12}	19600
$C_6H_5Cl + Cl_2 \xrightarrow{k_2} C_6H_4Cl_2 + HCl$	$r_2 = A_2 C_{C_6H_5Cl} C_{Cl_2} e^{-E/RT}$	3.6085×10^{20}	32600

Process contains a reaction section, followed by a separation train where the unreacted benzene is recovered and sent back to the first reactor. Kokossis y Floudas (1990)¹⁴ developed a synthesis problem to determine the optimal design for this process. They considered different structural alternatives for reaction and separation stages as well as different objective function (minimization of total cost, maximization of selectivity and reactor yield, maximization of profit, etc.). According to the original article, a recycle stream between reaction and separation sections is considered for all cases. Fresh benzene and a recycle benzene stream are fed to the first reactor, which operates in isothermal and isobaric fashion. The effluent from the first reactor is fed to the second reactor, which operates at the same conditions. The second reactor effluent is fed to a flash unit to remove all chlorine and hydrochloric acid; the liquid stream is then fed to the first distillation column, where the recycled benzene goes overhead, and a mixture of Chlorobenzene and Dichlorobenzene (bottoms product) are fed to the second column in which Chlorobenzene is recovered as a distillate product; Chlorine has to be fed as additional

stream in each reactor, unfortunately authors did not report those streams flowrates. Operating conditions of temperature and pressure are described as “moderate”, but no value was reported. We are planning to install a plant to produce Chlorobenzene; the class is the engineering group on charge. The chief engineer asked you to evaluate four of the optimal structures reported by Floudas and Kokossis (1990)¹⁴, to define the best option as well as proper operation conditions and preliminary costs.

Additional Data

Prices for reactants and products (USD): Chlorine \$19.88/kmol; Benzene \$27.98/kmol;

Monochlorobenzene \$92.67/kmol

Cost of capital investment (USD): reactor installed cost (\$) = $222.142 * D^{1.066} * H^{0.802}$

where the diameter (D) and length (H) are in ft.

Develop the proper mathematical model involving the mass and mole balances; the reaction rates expressions, the stoichiometric relationships, etc., to model system performance. Solve that model by using Excel and Polymath™ software, analyze different scenarios to define the effect of following variables on benzene conversion and its selectivity to the main product: feed reactants ratio and operating temperature.

Analyze the obtained results. In order to support your conclusions, analyze the behavior of every design variable (reactor volume, volumetric flow, residence time, spatial time, concentration of each component along the reactor, etc.). Based on such analysis propose a reactor design, justify the selected operation conditions.

Study cases (referred to the original source, Kokossis and Floudas, 1990¹⁴):

- Team 1: Student 1 and Student 2 (Case 1)
- Team 2: Student 3 and Student 4 (Case 4a)
- Team 3: Student 5 and Student 6 (Case 4b)
- Team 4: Student 7 and Student 8 (Case 4d)

References

- Bodman, S. W. 1968. *The Industrial Practice of Chemical Process Engineering*. MIT Press: Cambridge, MA.
- Kokossis, A. C. and Floudas, C. A. 1990, Synthesis of Isothermal Reactor- Separator- Recycle Systems. *Chemical Engineering Science*, 46: 1361-1383.