

# Application of Genetic Algorithm in Kinetic Modeling of Fischer-Tropsch Synthesis

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**ABSTRACT:** Kinetic modeling is an important issue, whose objective is the accurate determination of the rates of various reactions taking place in a reacting system. This issue is a pivotal element in the process design and development particularly for novel processes which are based on reactions taking place between various types of species. The Fischer Tropsch (FT) reactions have been used as the kinetic modeling bench mark. General kinetic models for FT, Water-Gas-Shift (WGS) and overall rates based on Langmuir-Hinshelwood-Hougen-Watson (LHHW) type have been considered and their optimum parameters have been obtained by Genetic Algorithms. The study shows the obtained model outperforms the other alternative models both in generality and accuracy. Due to flexibility and generality of Genetic Algorithms, it seems that GA is a useful technique with lots of potentials in determination of optimum kinetic model corresponding to a set of complex reactions.

**KEY WORDS:** Genetic algorithm, Kinetic model, Optimization, Fischer-Tropsch (FT), Water Gas Shift (WGS).

## INTRODUCTION

Research on chemical kinetics of heterogeneous catalytic reactions often requires rigorous kinetics because of reaction mechanisms are complicated and adsorption rate limiting steps occur by reactants, intermediates and products. By mechanism illustration, these rate equations are derived as non-separable and non-linear functions of

concentration and temperature [1-3].

Mathematical modeling of these complex chemical kinetics leads to non-linear parameter estimation problems in which the experimental data frequently contain more than one optimum. The optimization algorithms, which are based on traditional gradient based

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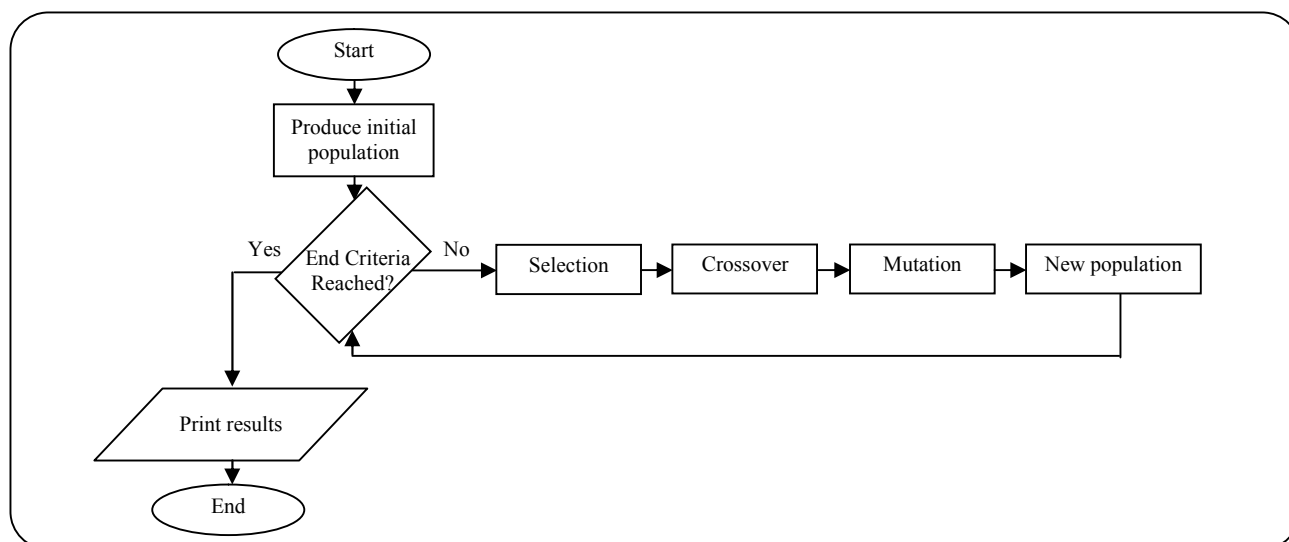


Fig. 1: The Flow Chart of Genetic Algorithm.

methods or direct search methods have the limitation of getting trapped in local optima. In these methods finding the appropriate initial estimates which lead to convergence to the global optimum can be difficult [4].

To overcome these limitations various approaches based on evolutionary algorithms have been recently used for optimization purposes. One of these algorithms is genetic algorithm which is based on the evolutionary process encountered in nature, and can be used as a novel optimization algorithm. The continuing price/ performance improvements of computational systems have made them attractive for some types of optimization.

In particular, genetic algorithms work very well on mixed (continuous and discrete), combinatorial problems. They are less susceptible to getting trapped at local optima than gradient search methods. But they tend to be computationally expensive [5-7]. The application of GA in various scientific and engineering disciplines including chemistry has recently increased [8].

There are a few articles published about kinetic modeling using GA, [8,9], but most of them are based on implementation of GA in prediction of good initial estimates for further application in iterative gradient based methods [10]. A few of the published papers implemented a hybrid GA method instead of pure GA for determination of kinetic parameters [11].

The aim of this work is the application of GA as a robust method for kinetic modeling of complex reaction mechanisms. In this research a GA is used to

find the global optimum without using any other auxiliary optimization method. This work focuses on kinetic modeling of Fischer-Tropsch and Water-Gas-Shift.

## GENETIC ALGORITHM

In Genetic algorithms, the solution procedure starts with an initial set of random candidate solutions called population. Each individual in the population is called a chromosome, representing a potential solution of the problem. A chromosome can be represented by a binary string; each part of this binary string represents one of the decision variables.

The chromosomes evolve through successive iterations, called *generation*. During each *generation*, the chromosomes are compared against each other according to a measure called fitness. To create the next *generation*, new chromosomes called offsprings, are formed through the following procedures:

- a) Merging two chromosomes from current *generation* using a *crossover* operator.
- b) Modifying a chromosome using a *mutation* operator.

In the new *generation* chromosomes with poor fitness are replaced by the obtained offsprings. In this procedure those chromosomes with better fitness have priority to participate in the creation of offsprings [12]. The general flowchart of the GA is presented in Fig. 1. Various stages of GA are elaborated in the following sections:

## INITIALIZATION

To create initial population, a set of chromosomes is randomly generated. Each chromosome is a binary string in which each gene can take a value of zero or one. Each decision variable in the original optimization problem is mapped to a set of genes whose length depends on the feasible range and the precision of the decision variable. As an example, for a decision variable like  $x_i$  whose value is between  $a_i$  and  $b_i$  and its precision is  $p_i$ , the number of required genes can be calculated through the following equation:

$$n_i = \text{int} \left( \log \frac{b_i - a_i}{p_i} / \log 2 \right) + 1 \quad (1)$$

Then the total number of genes in a chromosome ( $n_t$ ) is calculated by following equation:

$$n_t = \sum n_i \quad (2)$$

To convert the encoded parameters (genotypes) to the real parameters (phenotypes) a decoding mechanism by which the set of genes are converted to its corresponding variables is used [13].

## PARENT SELECTION

The chromosomes for the next generation are obtained by mating various pairs of chromosomes from current generation. These chromosomes are called parent and the results of mating are called offsprings. In order to keep the diversity of the offsprings, the parents are randomly selected from a set of chromosomes of current generation called *mating pool*. However, in order to let the chromosomes with higher fitness have more offsprings than those chromosomes with lower fitness, the chromosomes with higher fitness have more samples in mating pool than the other ones. Implementation of this approach is necessary for the enforcement of "Survival of Fittest" principle, which is the main objective of the Genetic Algorithms.

## CROSSOVER

*Crossover* is one of the main genetic operators, in which two chromosomes are selected as parents and at least one randomly selected block of genes is switched between these two chromosomes. The performance of genetic algorithms depends, to a great extent, on the performance of the *crossover* operator used in GA.

**Table 1: Singlepoint Crossover.**

Chromosome 1	1001 010000011110
Chromosome 2	1100 011110000111
Offspring 1	1001  <b>011110000111</b>
Offspring 2	1100  <b>010000011110</b>

**Table 2: Doublepoint Crossover.**

Chromosome 1	1001 01000 0011110
Chromosome 2	1100 01111 0000111
Offspring 1	1001  <b>01111</b>  0011110
Offspring 2	1100  <b>01000</b>  0000111

**Table 3: Multipoint Crossover.**

Chromosome 1	1001 010 00 001 1110
Chromosome 2	1100 011 11 000 0111
Offspring 1	1001  <b>011</b>  00  <b>000</b>  1110
Offspring 2	1100  <b>010</b>  11  <b>001</b>  0111

Various types of *Crossover* operator have been used in GA's, some of these types are as follows [14]:

- 1- Singlepoint.
- 2- Doublepoint.
- 3- Multipoint.

Tables 1-3 illustrates different examples of *crossover* operators. The crossover rate ( $P_c$ ) is defined as the ratio of the number of offsprings produced in each generation to the population size ( $N$ ). This ratio controls the expected number of chromosomes ( $P_c \times N$ ) undergoing the crossover operation. A higher crossover rate increases the exploitation of solution space.

## MUTATION

*Mutation* is a background operator which produces spontaneous random changes in various chromosomes. A simple way to achieve *mutation* would be to alter the value of one or more genes. *Mutation* serves the crucial role of exploration of search space and generation of sufficient variety in the chromosomes being handled in GA.

The *mutation rate* ( $P_m$ ) is defined as the percentage of the total number of genes at each generation whose values are flipped. Smaller the mutation rate, less variety in the candidate solution exists and lower amount of exploration will occur [15].

### FITNESS EVALUATION

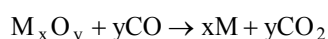
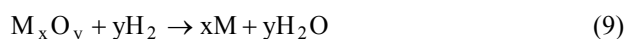
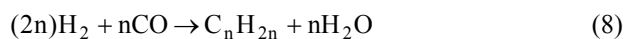
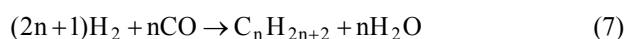
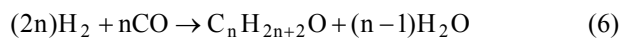
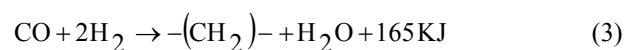
In this step each chromosome is first decoded to the corresponding decision variables, and then its fitness which can be considered as the degree of suitability of each chromosome is calculated. In a maximization problem, the fitness can be assumed to be the value of the objective function, whereas in a minimization problem it can be used as the opposite sign of objective function [16].

### KINETIC MODELS

Fischer-Tropsch synthesis (FTS), as an alternate process, can convert the synthesis gas (H<sub>2</sub>/CO) derived from carbon sources such as coal, peat, biomass and natural gas, into hydrocarbons and oxygenates. In consideration of the limited reserves of crude oil, today, it continuously attracts renewed interests as an option for the production of clean transportation fuels and chemical feedstocks [1-3].

It is of significant interest in both the process and the mechanism of FTS from the practical and theoretical viewpoints. The FTS produce a considerable variety of products that are mainly hydrocarbons and oxygenated compounds. The operation condition has significant influence upon the product distribution; therefore it is critically important to control the selectivity of the product. This is closely related to the kinetics and mechanism of the FTS. In the light of the potential economic and environmental importance of FTS, a detailed understanding of the process is highly desirable [1-3].

The Fischer-Tropsch synthesis is a complex network of parallel and series reactions involving different extents and determining altogether the overall catalyst performance. The whole synthesis reaction can be simplified as the combination of the FTS reactions and the water-gas shift (WGS) reaction [1-3].



The experimental data are derived at constant temperature 503 K and pressure range of 0.8 MPa to 4.0 MPa in a differential reactor with different concentrations of reactants [1-3].

A general kinetic model is selected by using reasonable reaction mechanisms found in literature [1-3]. These models are based on Langmuir-Hinshelwood-Hougen-Watson model which can cover most type of FT, WGS and overall syngas rate equations. Most of these equations are presented in tables 4-6.

According to tables 4-6 three general models with at least eight unknown parameters was selected as equations 12-14. These models can be used instead of different types of kinetic models observed in the literature.

The unknown powers and parameters (decision variables) should be calculated and optimized using GA.

$$r_{\text{FT}} = \frac{K P_{\text{CO}}^{\alpha} P_{\text{H}_2}^{\beta}}{(1 + a P_{\text{CO}}^{\chi} / P_{\text{H}_2}^{\delta} + b P_{\text{H}_2\text{O}})^{\epsilon}} \quad (12)$$

$$r_{\text{WGS}} = \frac{K(P_{\text{CO}} P_{\text{H}_2\text{O}} / P_{\text{H}_2}^{\alpha} - P_{\text{CO}_2} P_{\text{H}_2}^{\beta} / K_p)}{(\chi + a P_{\text{H}_2\text{O}} / P_{\text{H}_2}^{\delta} + b P_{\text{CO}}^{\epsilon} + c P_{\text{CO}_2}^{\phi})^{\theta}} \quad (13)$$

$$r_{\text{Ovl}} = \frac{K P_{\text{H}_2}^{\alpha} P_{\text{CO}}^{\beta}}{(\chi + a P_{\text{CO}}^{\delta} P_{\text{H}_2}^{\epsilon} + b P_{\text{H}_2\text{O}}^{\phi} + c P_{\text{H}_2}^{\theta} + d P_{\text{CO}_2}^{\gamma})^{\eta}} \quad (14)$$

Decision variables used in this study with their corresponding valid intervals are shown in tables 7-9.

A population size of fifty chromosomes (N=50) was selected in this study. Because of uncertainty in model parameters, large intervals are selected for frequency factors and energetic parameters.

Table 4: Kinetic models of Fischer-Tropsch.

Model	Kinetic Equation [1-3]
FT-I	$\frac{kP_{CO_2}^{1/2} P_{H_2O}^{1/2}}{(1 + aP_{CO}^{1/2} + bP_{H_2O})^2}$
FT-II	$\frac{kP_{CO_2}^{1/2} P_{H_2O}^{3/4}}{(1 + aP_{CO}^{1/2} P_{H_2}^{3/4} + bP_{H_2O})^2}$
FT-III	$\frac{kP_{CO}^{1/2} P_{H_2}}{1 + aP_{CO}^{1/2} + bP_{H_2O}}$
FT-IV	$\frac{kP_{CO} P_{H_2}^{1/2}}{(1 + aP_{CO} + bP_{H_2O})^2}$
FT-V	$\frac{kP_{CO} P_{H_2}}{(1 + aP_{CO} + bP_{H_2O})^2}$
FT-VI	$\frac{kP_{CO} P_{H_2}}{1 + aP_{CO} + bP_{H_2O}}$
FT-VII	$\frac{kP_{CO} P_{H_2}^2}{1 + aP_{CO} + bP_{H_2O}}$

Table 5: Kinetic models of Water-Gas-Shift.

Model	Kinetic Equation [1-3]
WGS-I	$kP_{CO}$
WGS-II	$k(P_{CO}P_{H_2O} - P_{CO_2}P_{H_2} / K_p)$
WGS-III	$\frac{k(P_{CO}P_{H_2O} - P_{CO_2}P_{H_2}^{1/2} / K_p)}{(1 + aP_{H_2O} / P_{H_2}^{1/2})^2}$
WGS-IV	$\frac{k(P_{CO}P_{H_2O} - P_{CO_2}P_{H_2} / K_p)}{P_{CO}P_{H_2} + aP_{H_2O}}$
WGS-V	$\frac{k(P_{CO}P_{H_2O} - P_{CO_2}P_{H_2} / K_p)}{P_{CO} + aP_{H_2O} + bP_{CO_2}}$

Table 6: Kinetic models of Overall syngas consumption.

Model	Kinetic Equation [1-3]
OVL-I	$r_{Overall} = KP_{H_2}^\alpha$
OVL-II	$r_{Overall} = KP_{H_2}^\alpha P_{CO}^\beta$
OVL-III	$r_{Overall} = \frac{KP_{H_2} P_{CO}}{P_{CO} + aP_{H_2O}}$
OVL-IV	$r_{Overall} = \frac{KP_{H_2}^2 P_{CO}}{P_{CO}P_{H_2} + aP_{H_2O}}$
OVL-V	$r_{Overall} = \frac{KP_{H_2}^2 P_{CO}}{1 + aP_{CO} P_{H_2}^2}$
OVL-VI	$r_{Overall} = \frac{KP_{H_2} P_{CO}}{P_{CO} + aP_{CO_2}}$
OVL-VII	$r_{Overall} = \frac{KP_{H_2} P_{CO}}{P_{CO} + aP_{H_2O} + bP_{CO_2}^\phi}$
OVL-VIII	$r_{Overall} = \frac{KP_{H_2}^{1/2} P_{CO}^{1/2}}{(1 + aP_{CO}^{1/2} + bP_{H_2}^{1/2})^2}$
OVL-IX	$r_{Overall} = \frac{KP_{H_2}^{1/2} P_{CO}}{(1 + aP_{CO} + bP_{H_2}^{1/2})^2}$
OVL-X	$r_{Overall} = \frac{KP_{H_2} P_{CO}}{(1 + aP_{CO})^2}$

In mechanisms proposed for FT and WGS reactions, the powers of the species concentration are usually integer numbers, although sometimes some of them may be one half if there is possibility of dissociation of molecule and atomic adsorption on the catalytic site. So the interval and resolution of discrete parameters are selected as shown in tables 7-9 to be able to adapt to the mechanism of the reaction.

The fitness function of this program is defined as the negative value of average absolute relative deviations (AARD) of the calculated reaction rates with respect to the experimental rates by the following equation:

$$AARD = \frac{1}{m} \sum_{i=1}^{i=m} \left| \frac{r_{exp,i} - r_{cal,i}}{r_{exp,i}} \right| \quad (15)$$

The convergence criterion used in the GA is to get to 95 % similarity value; suggested GA parameters are listed in Table 10 [17].

**Table 7: Unknown parameters of the model with boundaries and resolution for Eq. (12).**

Parameter	Lower limit	Upper limit	Precision or acceptable values
K	0.0001	5	0.0001
$\alpha$	0.5	1	0.5
$\beta$	0.5	2	0.5, 0.75, 1, 2
a	0	1	0.01
$\chi$	0.5	1	0.5
$\delta$	0	0.25	0.25
b	0	35	0.01
$\varepsilon$	1	2	1

**Table 8: Unknown parameters of the model with boundaries and resolution for Eq. (13).**

Parameter	Lower limit	Upper limit	Precision or acceptable values
K	0	300	0.01
$\alpha$	0	0.5	0.5
$\beta$	0.5	1	0.5
$K_p$	0	100	0.1
$\chi$	0	1	1
a	0	200	0.01
$\delta$	0	0.5	0.5
b	0	100	0.01
$\varepsilon$	0	1	1
c	0	100	0.01
$\Phi$	0	1	1
$\varphi$	0	2	1

**Table 9: Unknown parameters of the model with boundaries and resolution for Eq. (14).**

Parameter	Lower limit	Upper limit	Precision or acceptable values
K	0	5	0.01
$\alpha$	0.5	2	0.5, 1, 2
$\beta$	0	1	0.5
$\chi$	0	1	1
a	0	5	0.01
$\delta$	0.5	1	0.5
$\varepsilon$	0	2	1
b	0	200	0.01
$\Phi$	0	1	1
c	0	100	0.01
$\varphi$	0	2	0, 0.5, 1, 2
d	0	200	0.01
$\gamma$	0	2	1

Table 11: Results of this study and comparison with other results.

Model	Average Percent Relative Error	Model	Average Percent Relative Error	Model	Average Percent Relative Error
16	27.78186	17	27.61686	18	26.60664
FT-III2	42.89747	WGS-I5	86.82425		
FT-IV2	54.49429	WGS-II6	84.88095		
FT-III3	54.93696				

Table 10: Selected GA parameters.

Population size	50
Crossover rate	1
Mutation rate	0.15
Crossover type	Double point.

## RESULTS AND DISCUSSION

### Kinetic Model Validation

With respect to the different possible FT and WGS kinetic mechanisms a general model was selected with at least eight unknown parameters presented in equations (12)-(14). Equations (16)-(18) represent the optimum models. table 11 illustrates the performance of this research.

$$r_{FT} = \frac{0.55P_{CO}P_{H_2}^{1/2}}{1 + 0.65P_{CO} + 0.6P_{H_2O}} \quad (16)$$

$$r_{WGS} = \frac{209.218(P_{CO}P_{H_2O}/P_{H_2}^{1/2} - \dots)}{\left(1 + 121.02P_{H_2O}/P_{H_2}^{1/2} + \dots - P_{CO_2}P_{H_2}/66.59\right)^2} \quad (17)$$

$$r_{Overall} = \frac{1.92P_{H_2}^{1/2}P_{CO}^{1/2}}{1 + 2.47P_{CO} + 173.48P_{H_2O} + 88.65P_{H_2}^{1/2} + 164.78P_{CO_2}} \quad (18)$$

Application of proposed algorithm to find the optimum kinetic parameters based on the experimental data resulted in reliable kinetic model for Fischer-Tropsch and Water-Gas-Shift reactions.

Comparison of the calculated and experimental rates of reactions are shown in Figs. 2 to 9. Furthermore,

table 11 shows the comparison of the absolute relative deviations of the obtained models and their corresponding best alternatives that have been proposed in the literature until now.

## CONCLUSIONS

Unlike traditional gradient based methods one of the most important characteristic of Evolutionary Algorithms such as GA is their effectiveness and robustness when they are used for problems uncertainty, insufficient information and noise. In this approach minimum human effort and little insight into the details of the chemical mechanism is required to generate the optimum value for the reaction rate coefficients. Despite the flexibility and robustness of GA, its efficiency heavily depends on the type of genetic operators used in the algorithm and their corresponding parameters. In this study the appropriate type of genetic operators and their parameters have been studied and obtained.

Due to flexibility and generality of Genetic Algorithms, it seems that GA is a useful technique with lots of potentials in determination of optimum kinetic model corresponding to a set of complex reactions.

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## Nomenclatures

a	Reaction rate Constant
b	Reaction rate Constant
a <sub>i</sub>	Lower boundary of domain of a specific variable
b <sub>i</sub>	Upper boundary of domain of a specific variable
c	Reaction rate Constant
d	Reaction rate Constant
FT	Fischer-Tropsch
K	Reaction rate Constant

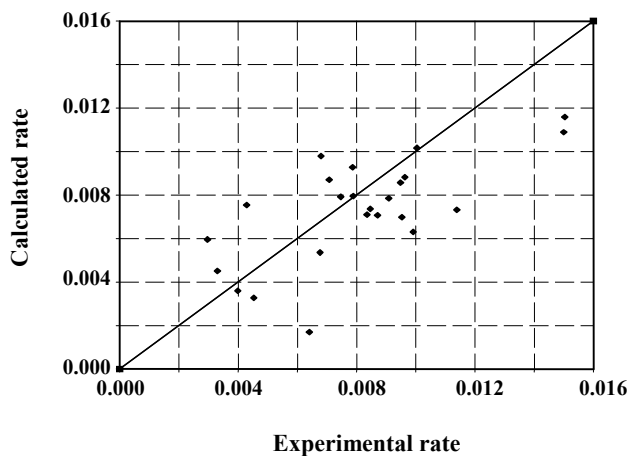


Fig. 2: Experimental rate of FT reaction vs. calculated rate, model no. (18).

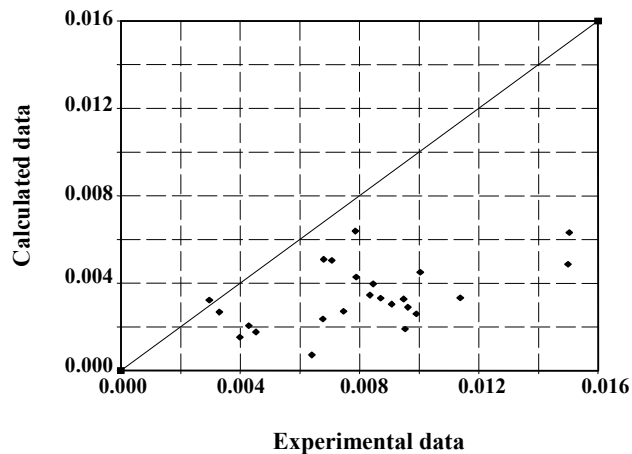


Fig. 5: Experimental rate of FT reaction vs. calculated rate, model no. (FT-1113).

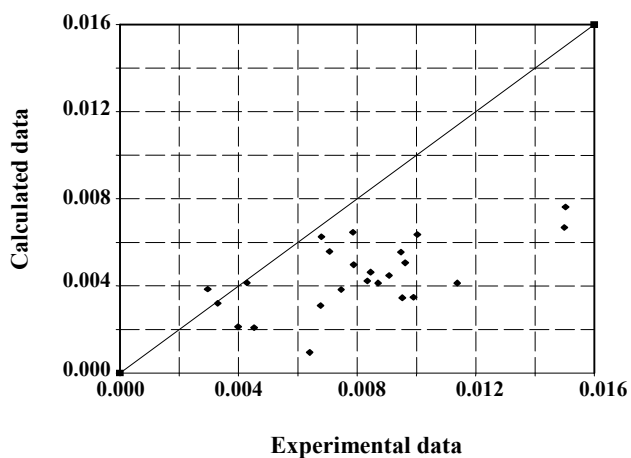


Fig. 3: Experimental rate of FT reaction vs. calculated rate, model no. (FT-1112).

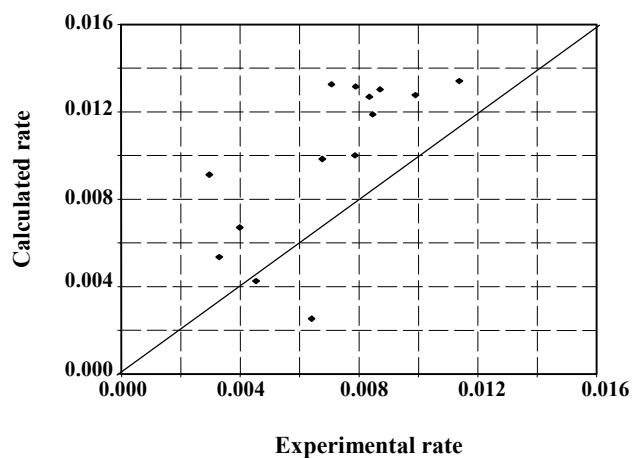


Fig. 6: Experimental rate of WGS reaction vs. calculated rate, model no. (17).

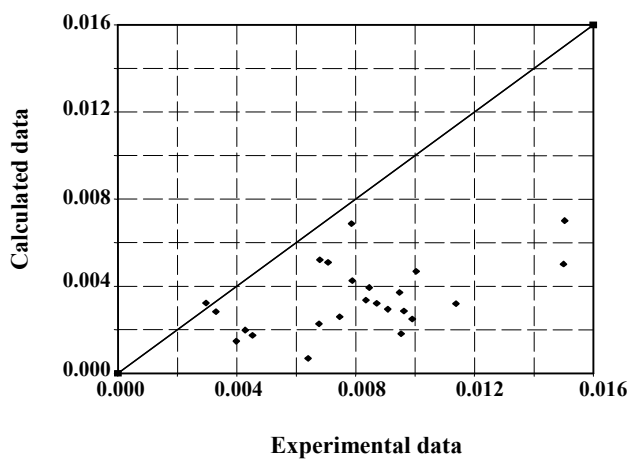


Fig. 4: Experimental rate of FT reaction vs. calculated rate, model no. (FT-IV2).

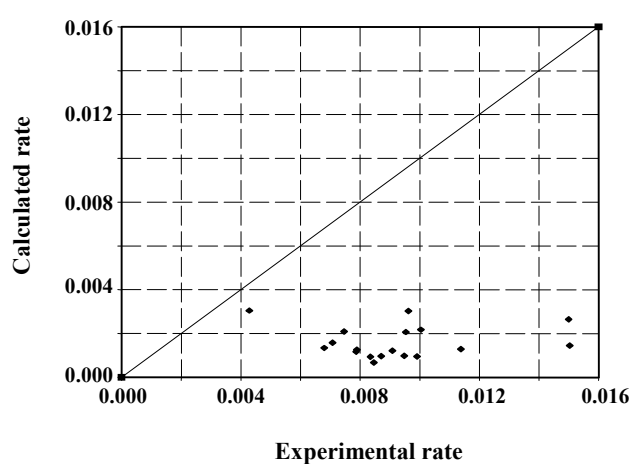


Fig. 7: Experimental rate of WGS reaction vs. calculated rate, model no. (WGS-15).



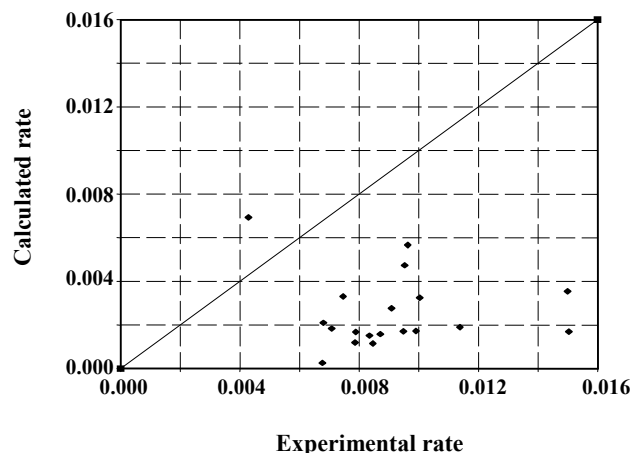


Fig. 8: Experimental Rate of WGS Reaction vs. Calculated Rate, Model No. (WGS-II6).

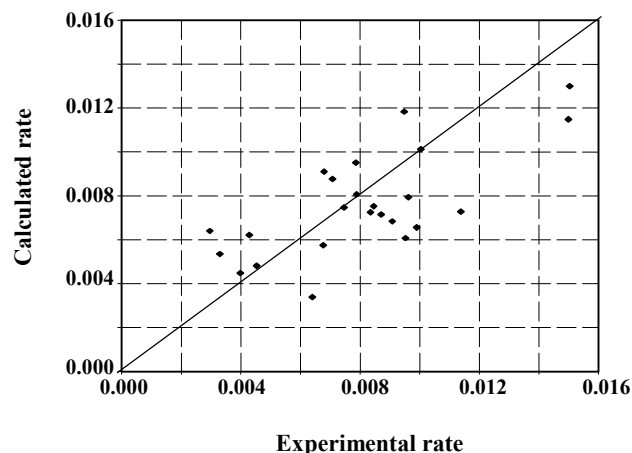


Fig. 9: Experimental Rate of Overall Syngas Consumption vs. Calculated Rate, Model No. (18).

$K_p$	Equilibrium constant water gas shift
$m$	Total number of experimental data
$N$	Population size
$n_i$	Number of bits in a specific variable
$ovl$	Overall synthesis gas consumption
$P$	Pressure (MPa)
$P_c$	Crossover rate
$P_i$	Precision of a specific variable
$P_m$	Mutation rate
$r$	Reaction rate $\left(\text{moleKg}^{-1}\text{catalyst}^{-1}\text{s}^{-1}\right)$
WGS	Water-Gas-Shift reaction
$\alpha$	Reaction rate Constant
$\beta$	Reaction rate Constant
$\gamma$	Reaction rate Constant
$\chi$	Reaction rate Constant
$\delta$	Reaction rate Constant
$\varepsilon$	Reaction rate Constant
$\phi$	Reaction rate Constant
$\varphi$	Reaction rate Constant

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