

# Foundations of Chemical Kinetic Modeling, Reaction Models and Reactor Scale-Up

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## Abstract

The majority of manuscripts dealing with the subject of chemical reactions, detailed chemical reaction models, and chemical kinetics present the material with strong mathematical foundations and emphasis. This is the reason for the conceptual approach taken in this manuscript where the result of the application of theory in the form of mathematical models is not the major consideration but the understanding of the fundamental concepts behind the complex mathematical theories.

**Keywords:** Chemical kinetics; Quantum chemistry; Pollution; Heat

## The Pillars of Detailed Chemical Kinetic Modeling (DCKM)

Even today, unfortunately, the industrial professional performing the design of a chemical reactor may not have the practical experience in Mathematics as well as Quantum Mechanics or Physical Chemistry to fully understand the implications of the mathematical formalism. This is especially true of the mathematical methods used in Quantum Chemistry as well as quantum mechanics applications to the estimate of rates of chemical reactions.

Another important consideration that may be difficult during the composition of reaction models is the generation of elementary chemical reactions to explain the formation of products and most important trace components some of which may be of concern due to pollution of the environment.

Reaction models based on mathematical correlations to predict formation of trace components of interest are not likely to be accurate. Note that trace components may have a significant importance for pollution control in effluent streams sometimes a very expensive proposition. It is necessary to develop complex models based on elementary chemical reactions.

Figure 1 depicts the pillars of engineering/science knowledge to be able to construct a detailed chemical kinetic model (DCKM) consisting of hundreds of elementary chemical reaction.

General Chemistry & Chemical reactions

Chemical Thermodynamic/Statistical Mechanics

Chemical Kinetic Principles and Theories of Rate Coefficients

Quantum Chemistry

This author is aware of only two manuscripts dealing with the subject of DCKM. Both are limited in content as well as lacking a comprehensive and conceptual description of the issues and concepts. This would make it very difficult for the readers to begin the task of constructing a complex reaction model and the associated data that is required such as rate coefficients. The works are highly theoretical and assume an understanding of Chemical Kinetics, General Chemistry, and Quantum Chemistry [1,2].

The predictions of any reactor model are only as good as the chemistry that is assembled to explain the formation of products and important byproducts. This is frequently the most important and

difficult task.

To model a chemical reactor involves developing Heat, Mass, and Component balances requiring heats of reaction as well as chemical kinetics for each reaction participating in the model. The activation energy for newly found reactions can be estimated with the use of Quantum Chemistry methods.

Rate coefficients can be estimated with Transition State theory or the simpler Kinetic theory, better yet is the use of experimental data applicable to the temperature and pressure conditions of the reactor.

## Detailed Chemical Kinetic Models: A Stepwise Procedure

As introduced above, the design of complex chemical reactors including the modeling of the relevant chemistry is a fairly complex undertaking. The design depending on reactor configuration and thermodynamic conditions may involve solving mass, heat, and momentum transport equations. This also requires mathematical tools to solve stiff differential equations involving the chemical reaction rates.

A very good pictorial representation of the above is given in Figure 2. The most important step is always the assembly of the elementary reactions that will participate in the model. These reactions should explain the formation of major products of reaction, but as important the formation of intermediates and trace components. Very often, trace components are considered an environmental hazard or pollutant.

It will be necessary (for Heat and Mass balance purposes during the chemical reactor design) to gather the thermo-chemical data for the reactions such as Heats of Formation. Experimental data can be most important. Also through Quantum chemistry and reaction rate theory

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such as Transition State, the activation energy and rate of reaction can be estimated respectively.

The reaction model is normally validated by the careful selection of an experimental apparatus. The final objective is then to develop a commercial scale chemical reactor and predictive tools based on mathematical models. Figure 3 Westbrook [3] provides a general guide to the choosing of experimental technique based on reaction conditions.

### Chemical Reactor Scale-up: Fundamental Concepts

It is necessary to introduce reactor scale-up in a manuscript dealing with detailed chemical kinetic models because it impacts the development of the model as depicted in Figure 2 during (a) selection of experimental apparatus and (b) model validation. Further analysis of reactor scale-up can be found in the work of Edgeworth and Wooldridge [4]. Other important sources of information are Euzen et al., Zlokarnik and Worsetell [5-7].

The temperature and pressure of operation of the reactor would lead to selecting a plug flow reactor for example or a burner to investigate combustion of a fuel or chemical. Attention to detail will lead to better prediction of the formation of toxic trace components.

The complexity of the Heat, Mass, and Momentum transport equations to be solved in a Computational Fluid Dynamic (CFD) simulation of the reactor may necessitate the use of a reduced mechanism rather than a full blown reaction mechanism consisting of hundreds of reactions. Chemical reaction because of its exponential dependence on temperature results in stiff equations that need special mathematical treatment to be solved and convergence to a solution could be very time consuming.

Figure 4 is an attempt to expand on the above subject as well as to depict the necessary steps to take to insure the proper design of equipment. It is needed that a chemical reactor of commercial size be designed in accordance to the values of similarity parameters identified in the smaller model reactor:

- Geometric
- Kinematic
- Dynamic,
- Thermal
- Chemical similarity

One similarity condition requires all of the above. Thus, Chemical Similarity requires fulfillment of geometric, kinematic, dynamic and thermal similarity. However, one important consideration in simplifying scale-up is the identification of the controlling regime: in a reactor where diffusion is controlling dynamic similarity would be predominant criteria over all others including chemical similarity.

The approach to developing scaling up parameters depends on the level of understanding of the system:

- Where the differential equations that govern the behavior of the system are unknown, but provided that we do know all the variables which would be of importance in the differential equations, it is possible to develop the similarity criteria by means of Dimensional Analysis.
- Where the differential equations of the system are known but cannot be integrated, the similarity criteria can be derived from the

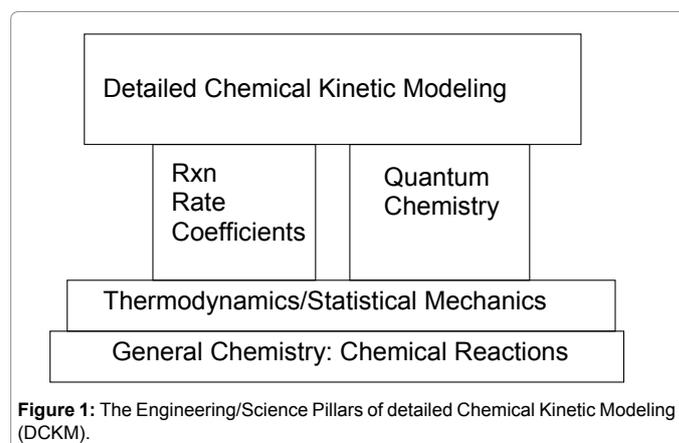


Figure 1: The Engineering/Science Pillars of detailed Chemical Kinetic Modeling (DCKM).

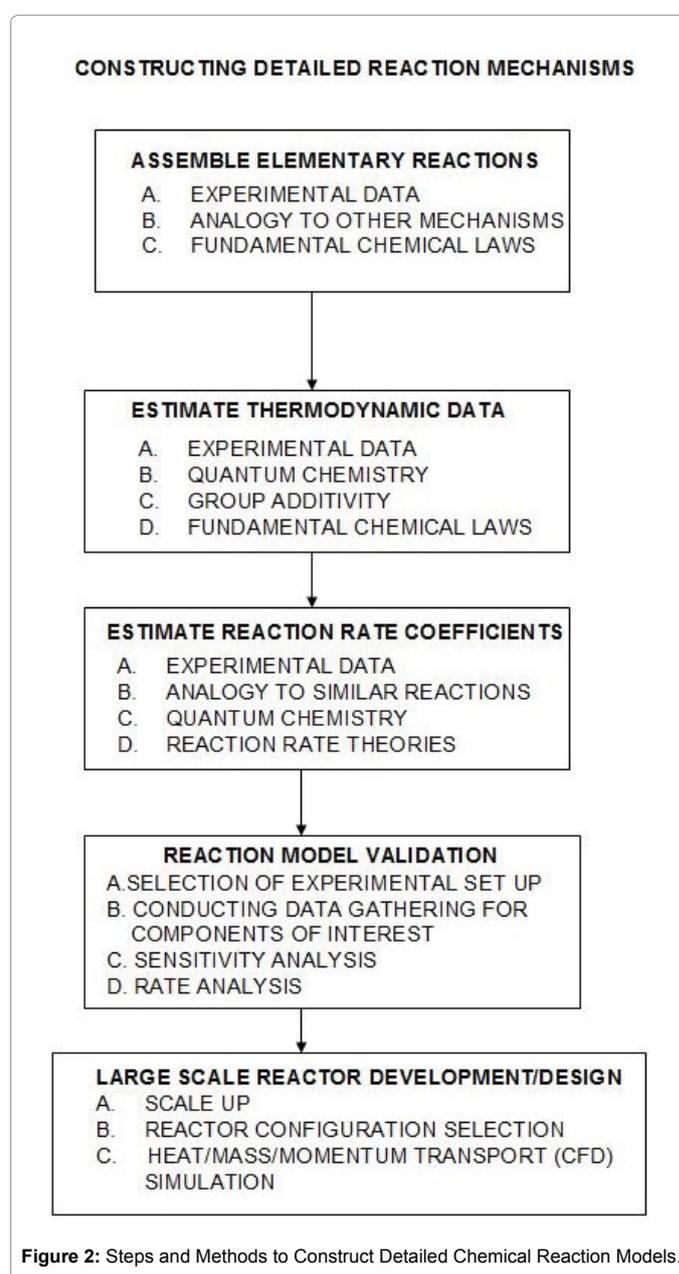


Figure 2: Steps and Methods to Construct Detailed Chemical Reaction Models.

EXPERIMENT	PRESSURE RANGE	TEMPERATURE (K)	DILUTION	MIXING LIMITS	TRANSPORT EFFECTS
SHOCK TUBES	High	<1300	Yes	None	NO
PLUG FLOW REACTOR	Atmospheric	850-1300	Yes	None	NO
FLAMES (Combustion)	Low	800-2500	No	Flammability Limits	Yes

Figure 3: Model validation: Experimental Approach.

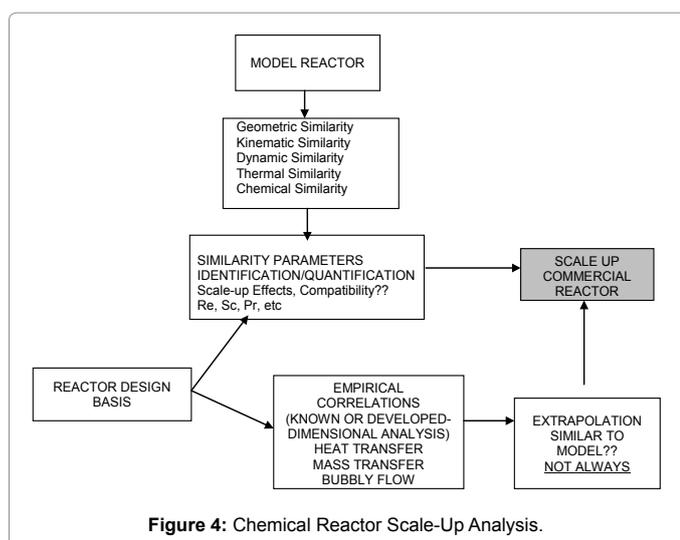


Figure 4: Chemical Reactor Scale-Up Analysis.

differential form.

- Finally, where the differential equations are both known and capable of being integrated and solved, there is no need for either similarity criteria or model experiments, since the behavior of a large-scale system should be directly calculable.

- The approach based on similarity can be very strict and sometimes not possible due to conflicting parameters leading to unpractical results such as very large power consumption in the case of an agitated vessel for example. It requires then selection of the controlling regime to base scale-up on the proper criteria ignoring others.

A more practical and often used approach is to design the scaled-up reactor by “extrapolation” making use of empirical correlations based on dimensionless numbers such as Reynolds, Prandtl, Schmidt etc. founded on mass and heat transfer differential equations to arrive at the proper size of the commercial reactor. In this approach, the model reactor and scaled-up version are necessarily geometrically similar but other similarity criteria are not met.

One example is the heat transfer correlation based on the Nusselt number for the heat transfer coefficient inside tubes of shell and tube exchangers which has the form,

$$Nu = C * Re^x * Pr^y$$

where C is a constant, Re is the Reynolds number with exponent x, and Pr is the Prandtl number with exponent y. Parameters C, x, and y are determined experimentally with the model system.

### Application of Similarity-Bioreactor Scale-Up

Design of a production-scale system on the basis of the performance of a model system [8,9].

### System characteristics

Thermodynamics e.g.  $C_L^*$ ,  $C_p$  (scale independent specific heat)

Kinetics e.g. specific growth rate,  $m$  (scale independent)

Transport-phenomena e.g.  $k_L a$  (scale independent mass transfer coefficient)

One of the fundamental problems is incomplete mixing on scale-up, concentration gradients, cell growth on walls etc.

Similarity between model (M) and prototype (F) in two mixing systems, it is (ideally) important to achieve:

**Geometric similarity:** both systems must be the same shape and linear dimensions in model must be related to the corresponding dimensions in prototype by a constant factor

$$\text{i.e. } \left(\frac{D}{T}\right)_M = \left(\frac{D}{T}\right)_F \text{ and } \left(\frac{H}{T}\right)_M = \left(\frac{H}{T}\right)_F \text{ etc. } D, \text{ diameter } H, \text{ height}$$

**Kinematic similarity:** similar flow patterns in both vessels; velocities at corresponding points must have a constant ratio.

**Dynamic similarity:** all corresponding forces at corresponding points in two geometrically similar systems have a constant ratio, i.e.

$$\left(\frac{P_0}{\rho N^3 D^5}\right)_M = \left(\frac{P_0}{\rho N^3 D^5}\right)_F \text{ or Power number (a function of Reynolds (Re) and Froude (Fr) numbers)}$$

and Froude (Fr) numbers)

$$\left(\frac{\rho N D^2}{\mu}\right)_M = \left(\frac{\rho N D^2}{\mu}\right)_F \text{ or Re number}$$

$$\left(\frac{N^2 D}{g}\right)_M = \left(\frac{N^2 D}{g}\right)_F \text{ or Fr number}$$

Note: P, power and N, Impeller rotational speed

However, it is very difficult to achieve dynamic similarity when more than one dimensionless group is involved in a system as shown below. For this reason, some bioreactor common criteria for scale-up have been developed based on which regime is controlling:

- Constant P/V
- Constant impeller tip speed (pND)
- Constant volumetric mass transfer coefficient ( $k_L a$ )
- Constant impeller Reynolds No
- Constant mixing time ( $t_{mix}$ ) or circulation time ( $t_{circ}$ )
- Common to scale-up in the basis of geometric similarity and at least one of the above

### Power/Impeller Speed of Fermenter

Assuming dynamic similarity as controlling, can you determine the power consumption and impeller speed of a 1000 L fermenter, based on the findings of the optimum conditions from a geometrically similar 1 L vessel? If not, can you scale-up by using a different fluid system?

Solution

$$\left(\frac{V_M}{V_F}\right) = \frac{1}{1000} \Rightarrow \frac{D_M}{D_F} = \left(\frac{1}{1000}\right)^{\frac{1}{3}} = \frac{1}{10} \text{ Geometric Similarity Condition}$$

$$\left(\frac{P}{\rho N^3 D^5}\right)_M = \left(\frac{P}{\rho N^3 D^5}\right)_F \text{ Dynamic Similarity Condition}$$

If using same fluid in both systems ( $\rho_M = \rho_F$ , and  $\mu_M = \mu_F$ ),

$$P_F = P_M \left( \frac{N_F}{N_M} \right)^3 \left( \frac{D_F}{D_M} \right)^5 = 10^5 \left( \frac{N_F}{N_M} \right)^3 P_M$$

Equality of Reynolds Number demands that

$$N_M D_M^2 = N_F D_F^2 \Rightarrow N_F = N_M (D_M / D_F)^2 = 0.01 N_M$$

But, equality of Froude Number demands that

$$N_M^2 D_M = N_F^2 D_F \Rightarrow N_F = N_M \sqrt{D_M / D_F} = N_M \sqrt{1/10} = 0.3162 N_M$$

It is impossible to achieve dynamic similarity using same fluid in both model and full-scale systems.

But, if  $\rho_M \neq \rho_F$ , and  $\mu_M \neq \mu_F$ , then for this system can satisfy dynamic similarity using a model fluid such that

$$\left( \frac{\mu}{\rho} \right)_M = \frac{1}{31.62} \left( \frac{\mu}{\rho} \right)_F$$

i.e. if the full-scale system involves a fluid similar to water, then the chosen model fluid must have a kinematic viscosity of 1/31.62 that of water. IMPOSSIBLE!

Thus as indicated before some limited similarity criteria needs to be chosen, as compromise:

A. Constant P/V – most commonly used criterion for scale-up

$$\left( \frac{P}{D^3} \right)_F = \left( \frac{P}{D^3} \right)_M \text{ and if } \left( \frac{P}{\rho N^3 D^5} \right)_F = \left( \frac{P}{\rho N^3 D^5} \right)_M$$

$$\Rightarrow \left( \frac{N_F}{N_M} \right)^3 = \left( \frac{D_M}{D_F} \right)^2 \Rightarrow N_F = N_M \left( \frac{D_M}{D_F} \right)^{\frac{2}{3}}$$

B. Constant  $k_L a$  – desirable for biological processes recall

$$k_L a = K \left( \frac{P}{V} \right)^{\alpha} (u_{SG})^{\beta}, \text{ and for turbulent flow } N_p = \text{const } t \Rightarrow P \propto N^3 D^5$$

$$\text{and the superficial gas velocity } u_{SG} = \frac{Q_g}{A} \propto \frac{Q_g}{D^2}$$

$$k_L a \propto (N^3 D^2)^{\alpha} \left( \frac{Q}{D^2} \right)^{\beta}$$

$$\text{to maintain } (k_{L_a})_F = (k_{L_a})_M$$

$$(N^3 D^2)_F^{\alpha} \left( \frac{Q}{D^2} \right)_F^{\beta} = (N^3 D^2)_M^{\alpha} \left( \frac{Q}{D^2} \right)_M^{\beta}$$

can keep both  $k_L a$  and P/V constant, but only if  $u_{SG}$  is also constant. If geometric similarity is maintained, then  $D_F$  is fixed, with respect to  $D_M$  and can determine  $N_F$  and  $Q_F$

C. Constant Impeller Tip Speed (pND) – may be appropriate for shear sensitive systems (although shear rate is frequently correlated with impeller rotational speed N):

$$\text{If constant tip speed } (\pi N D)_F = (\pi N D)_M \text{ turbulent flow}$$

$$\left( \frac{P}{\rho N^3 D^5} \right)_F = \left( \frac{P}{\rho N^3 D^5} \right)_M$$

$$\left( \frac{P}{(ND)^3 D^2} \right)_F = \left( \frac{P}{(ND)^3 D^2} \right)_M \Rightarrow \left( \frac{P}{D^2} \right)_F = \left( \frac{P}{D^2} \right)_M$$

$$P_F = P_M \left( \frac{D_F}{D_M} \right)^2$$

If impeller tip speed is kept constant, what about Re?

$$\frac{Re_F}{Re_M} = \frac{\rho N_F D_F^2}{\mu} \frac{\mu}{\rho N_M D_M^2} = \frac{(N_F D_F) D_F}{(N_M D_M) D_M} = \frac{D_F}{D_M}$$

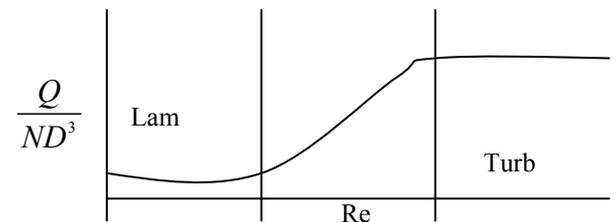
$$Re_F = Re_M \frac{D_F}{D_M}$$

D. Constant Mixing Time/Circulation Time most commonly used in chemical reactors

$$(1) \text{ Circulation Time: } t_{\text{circ}} \approx \frac{V}{Q} \text{ and } t_{\text{mix}} \approx 5 \rightarrow 6 (t_{\text{circ}})$$

In a standard configuration STR

$$N_Q = \frac{Q}{ND^3} \approx 2.8 \text{ for turbulent flow}$$



Constant  $t_{\text{mix}}$ ?

Assuming turbulent flow:

$$\left( \frac{Q}{ND^3} \right)_F = \left( \frac{Q}{ND^3} \right)_M \Rightarrow \left( \frac{Q}{D^3} \right)_F \left( \frac{1}{N} \right)_F = \left( \frac{Q}{D^3} \right)_M \left( \frac{1}{N} \right)_M$$

for constant mixing time,  $N_F = N_M$

Note: Calculating Apparent Viscosity Problem

$$\mu_a = \frac{M(R_o - R_i)}{2\pi R_i h \Omega}$$

The nomenclature relating to this formula

$\mu_a$  = Apparent viscosity

$M$  = Torque

$R_o$  = Radius of outer cylinder

$R_i$  = Radius of inner cylinder

$h$  = height of cylinder

$\Omega$  = Angular velocity

For this case using a rotational speed in RPM

$$\Omega = \frac{2\pi N}{60}$$

Where N is the rotational speed of the cylinder in RPM

Convert N to W using above formula

Calculate  $m_s$  for each of the above points

### Scale-Up by Extrapolation

Extrapolation calls for the model and prototype to be geometrically similar as necessary condition.

An example is the scale up of a heat exchanger which would make use of the well know relation involving the Nusselt number as function of Re and Pr dimensionless numbers to calculate heat transfer coefficients or,

$Nu = C * Re^X * Pr^Y$  Nu or Nusselt no., Re, Reynolds no., and Pr, Prandtl no.

The task would be to determine via pilot plant work for a given system, the value of the constant C, and exponents X, and Y. Note that this dependency arises naturally from the development of the heat transfer differential equations for the system.

The sizing of the commercial heat exchanger makes use of such empirical correlations to be able to meet the expected performance specifications, but it will not necessarily be similar to the small scale model.

The equivalent for mass transfer is

$Nu_m = C * Re^X * Sc^Y$  Nu or Nusselt no. for mass, and Sc, Schmidt no.

Another application arises in the design of packed towers for Gas/Liquid mass transfer, the overall mass transfer coefficient K often takes the form,

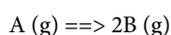
$$K = C*(L)^X*(G)^Y$$

Where L is the liquid flux over the packing and G is the Gas flux flowing up the packing. By means of pilot work, the constant C, and exponents X, and Y are determined directly for the system under consideration.

### Detail Analysis Chemical Reactor Scale-up

The concepts presented in the previous sections can now help elucidate issues on the scale-up of chemical reactors as developed below in more detail. To identify the pertinent dimensionless quantities, the Heat, Mass, and Momentum Conservation equations are considered:

First consider a gas phase reaction of first order or,



and considers the elements in conservation of mass, heat, and momentum.

### Conservation of Mass

$-r$  = rate of accumulation due to chemical reaction

$\frac{\delta}{\delta L}(uC) = u \frac{\delta C}{\delta L} + C \frac{\delta u}{\delta L} = u \frac{\delta C}{\delta L} + rC\Delta V$  = rate of accumulation due to bulk flow, where  $\Delta V$  is volume change Per unit disappearance of A

$$\frac{\delta}{\delta L}(D \frac{\delta C}{\delta L}) = \text{rate of accumulation due to diffusion.}$$

### Conservation of Momentum

$-\rho u \frac{\delta P}{\delta L}$  = force as a result of momentum transferred by bulk flow,  
 $\frac{\delta P}{\delta L}$  = force a result of external pressure (relatively nil),  $\rho u \Delta V$  = force as a result of volume change due to reaction,  $\frac{u}{\rho} \frac{\delta^2 u}{\delta R^2}$  = force as a result of shear viscosity.

### Conservation of Thermal Energy

$-\frac{\delta C}{\delta L}(C_p \rho u \Delta T)$  = accumulation of heat accompanying flow of fluid  
 Where  $\Delta T$  is the temperature in excess of the wall temperature  $rQ$  = heat generated by reaction  $u \frac{\delta P}{\delta L}$  = heat generated by friction (relatively nil)  $k \frac{\delta^2 \Delta T}{\delta R^2}$  = heat transferred by conduction from direction of reactor wall  $4\alpha\epsilon\sigma T^3 \Delta T \sim 4\alpha\epsilon\sigma [(T+\Delta T)^4 - T^4]$  = heat transferred by radiant exchange with walls, where  $\alpha$  is the wall surface per unit volume.

The conservation laws lead to seven dimensionless groups that must be equal for model and prototype in order for complete similitude to exist as identified by Walas [10].

Table 1 depicts the chemical reactor dimensionless groups. Tables 2- 4 examine other reactor parameters pertinent to scale-up. The Tables lead to the following conclusions:

- Groups (a) through (g) for homogeneous reactions or

Homogeneous Reactions		Heterogenous Reactions		Name of Group
(a)	$\frac{rL}{uC}$	(h)	$\frac{r}{SC}$	Damkohler
(b)	$\frac{u}{rL\Delta t}$	(i)	$\frac{S}{r\Delta V}$	
(c)	$\frac{u}{rL\Delta V}$	(j)	$\frac{S}{r\Delta V}$	Reynolds
(d)	$\frac{Ru\rho}{\mu}$	(k)	$\frac{D_p S L_p}{\mu}$	
(e)	$\frac{QrR^2}{k\Delta T}$	(l)	$\frac{C_p S_p R^2}{k + \frac{4}{3} D_p \rho \sigma T^3}$	Peclet
(f)	$\frac{QrR^2}{k\Delta T}$	(m)	$\frac{QrR^2}{(k + \frac{4}{3} D_p \rho \sigma T^3) \Delta T}$	
(g)	$\frac{Qr}{\Delta T(4\alpha\epsilon\sigma T^3 - C_p \rho r \Delta V)}$	(n)	$\frac{Qr}{C_p \rho r \Delta V \Delta T}$	

Table 1: Chemical Reactor Dimensionless Groups.

(h)-(n) for heterogeneous reactions, of Table 1 must be satisfied for complete similitude between model and prototype. On the whole the requirements for complete similitude are impractical.

Conditions	Dimensionless Groups of Table 1 That can be ignored	
	Homogenous	Heterogeneous
Batch or low flow rate	a, c, d, e	h, j, k, l
Small diffusional resistance	b	i
Constant volume	c, g	j, n
Small radiation Heat transfer	g	n
Adiabatic with small heat of reaction	f, g	m, n

Table 2: Special cases resulting in Limited Similitude.

Item	Ratio of Prototype to Model ( $\lambda$ , ratio of radius)		
	Homogeneous	Heterogeneous	
Particle Diameter	-----	$\lambda$	1
Heat-Transfer surface	$\lambda^3$	$\lambda^3$	$\lambda^3$
Volumetric Flow rate	-	$\lambda$	1
Heat Transfer per unit of surface	$\lambda^3$	$\lambda^2$	$\lambda^3$
Heat Transfer per unit of throughput	1	$\lambda$	1

Table 3: Geometrical Relations in Chemical Similitude.

Item	Ratio of Prototype to Model ( $\lambda$ , ratio of radius)		
	Homogeneous	Heterogeneous	
Particle Diameter	-----	$\lambda$	1
Heat-Transfer surface	$\lambda^2$	$\lambda^2$	$\lambda^2$
Volumetric Flow rate	$\lambda^3$	$\lambda^2$	$\lambda^3$
Heat Transfer per unit of surface	1	$\lambda^2$	$\lambda^2$
Heat Transfer per unit of throughput	$\lambda^{-1}$	1	$\lambda^{-1}$

Table 4: Heat-Transfer Relations in Chemical Similitude.

- Whether chemical or dynamic or thermodynamic, or geometric similitude individually or in partial combination is adequate depends on the characteristic of each reaction mixture and the operating conditions.

- Table 2 lists the dimensionless numbers that may be ignored in certain special cases such as for a batch reactor.

- When the controlling condition is either heat transfer or diffusive mass transfer, dynamical similitude is required since the coefficients of both processes depend on reynolds number.

- Chemical Similitude alone is required when the chemical reaction rate controls the operation, equality of the Damkohler group  $rL/uC$  is then sufficient. Chemical similitude is achieved when reaction rates, reaction times, and the initial concentration are the same in both model and prototype.

When chemical similitude is preserved, the geometry and the heat transfer behavior are also fixed, but not necessarily as required by dynamical similitude. Geometric and Heat Transfer relations when chemical similitude is controlling [10].

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