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## **ARRHENIUS LAW MODIFICATION FOR TURBULENT COMBUSTION MODELING**

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ABSTRACT In the Arrhenius law, the effects of turbulent fluctuations on rate of reactions are ignored. The model is generally inaccurate for turbulent flames due to highly non-linear Arrhenius chemical kinetics. Currently great deals of the CFD studies make use of the Eddy-Dissipation Model for investigation of turbulent combustion. Although CFD software such as Fluent allows multi-step reaction mechanisms applying the eddy-dissipation model, they will likely produce incorrect solutions. The reason is that multi-step chemical mechanisms are based on Arrhenius rates, which differ for each reaction. In this paper, a turbulent-chemistry interaction model is developed for numerical investigation of turbulent combustion according to Arrhenius law, which can be generalized to a wide range of complexity, from a single global reaction to a detailed reaction mechanism involving perhaps hundreds of species. It is achieved by introducing a correction turbulent factor (based on turbulent viscosity) added to temperature terms in Arrhenius law. Then, the modified Arrhenius law by this way is applied to simulate an air-methane diffusion flame. The results show that the modified Arrhenius model is in better agreement with the probability density distribution function (PDF) results than eddy-dissipation.

Keywords Turbulent combustion, rate of reaction, molecular collision, Arrhenius law, viscosity

## **INTRODUCTION**

It is well known that, apart from generating heat and power, combustion produces pollutants such as oxides of nitrogen, soot and unburnt hydrocarbons (HC). Furthermore, inevitable emissions of CO2 are known to be a reason of global warming. These emissions will be reduced by improving the efficiency of the combustion process, thereby increasing fuel economy [Poinsot 2001]. In technical processes, combustion nearly always takes place within a turbulent flow field rather than a laminar one [Poinsot 2001]. Therefore, investigating turbulent combustion processes is an important issue to improve practical systems (i.e. to increase efficiency and to reduce fuel consumption and pollutant formation). Since combustion processes are hardly amenable to analytical techniques, numerical methods for turbulent flames become a fast growing field. However numerical simulations of turbulent reacting flows remain a challenging task due to some drawbacks, namely [Fluent Inc. 2006, Wang 2008]:

Combustion, even without turbulence, is an intrinsically complex process involving a large range of chemical time and length scales. The full description of chemical mechanism in combustion flame may require hundreds of species and thousands of reactions leading to considerable numerical complexity.



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Turbulence itself is probably the most complex phenomenon in non-reacting fluid mechanics. Turbulent combustion results from the two way interaction of chemistry and turbulence.

The main effect of turbulence on combustion is to increase the combustion rate [Fluent Inc. 2006]. The way it works in turbulent combustion depends on the turbulent mixing process. Elementary reactions are initiated by molecular collisions in the gas phase. Many aspects of these collisions determine the magnitude of the rate constant, including the energy distributions of the collision partners, bond strengths, and internal barriers to reaction [Kee 2003].

There exists a large body of scientific literature on the chemical mechanisms of combustion modeling in turbulent flames [Wang 2008, Poinsot 2001]. The simplest model based on the molecular collisions of fuel and oxidizer to calculate the rate of reaction, is laminar finite rate model. Laminar finite rate model computes the rate constants and chemical source terms using Arrhenius law. Arrhenius law can be applied to different levels of complexity, from a single global reaction to a detailed reaction mechanism involving perhaps hundreds of species [Kee 2003]. In the Arrhenius law, the effects of turbulent fluctuations on rate of reactions are ignored. The model is generally inaccurate for turbulent flames due to highly non-linear Arrhenius chemical kinetics [Glassman 1987]. This model may, however, be acceptable for combustion with relatively slow chemistry and small turbulent fluctuations, such as supersonic flames [Glassman 1987]. An early attempt to provide a closure for the chemical source term in turbulent reacting flow is due to Spalding [1970]. In model of Spalding (Eddy Break Up model) turbulent mean reaction rate was expressed as a function of variance of the product mass fraction. This model has been modified by Magnussen and Hjertager [1976] who replaced variance of the product mass fraction simply by the mean mass fraction of deficient species calling it the Eddy Dissipation Model [Magnussen 1976].

The direct numerical simulation (DNS), large-eddy simulation (LES), probability density distribution function (PDF) transport equation model and the conditional moment closure (CMC) model, developed in recent years, can well simulate the interaction of turbulence with detailed chemistry, these refined models need rather large computer memory and computation time. They can be used only in simulating very simple flows for fundamental studies to understand the effect of turbulence structure on combustion.

Nevertheless, most computational fluid dynamics (CFD)-based engineering studies make use of much simpler models which are usually considered out-of-date or physically nonsense. On the other hand, direct numerical simulation will not be an engineering tool for design of boilers, furnaces and turbines in future [Brink 2000]. In practice, great deals of the CFD studies make use of the Eddy Break-Up (EBU) Model of Magnussen and Hjertager. Although CFD software such as Fluent allows multi-step reaction mechanisms with the eddy-dissipation model, they will likely produce incorrect solutions [Glassman 1987]. The reason is that multi-step chemical mechanisms are based on Arrhenius rates, which differ for each reaction. In the eddy-dissipation model, all reactions have the same turbulent rate, and therefore the model should be used only for one-step (reactant  $\rightarrow$  product), or two-step (reactant  $\rightarrow$  intermediate, intermediate  $\rightarrow$  product) global reactions. The model cannot predict kinetically controlled species such as radicals.





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In this paper, Arrhenius rate for turbulent reaction is corrected based on turbulent viscosity definition. A correction factor, based on turbulent viscosity is introduced which added to temperature terms in Arrhenius reaction rate law for turbulent combustion flame simulation.

### LAMINAR FINITE RATE MODEL

All chemical reaction, whether of the hydrolysis, acid-base or combustion type, take place at a definite rate and depend on the conditions of the system. A stoichiometric relation describing chemical reactions of arbitrary complexity can be represented by the equation [Poinsot 2001]:

$$\sum_{i=1}^{N} v'_{i,r} M_i \rightleftharpoons \sum_{i=1}^{N} v''_{i,r} M_i$$
(1)

Where  $v'_j$  and  $v''_j$  are the stoichiometric coefficients for species *i*th in the reaction *r*th as reactant and product,  $M_i$  the molar concentration of the *i*th species. The finite-rate model assumes that the reaction rate  $(\hat{R}_{i,r})$  depends on the collision of molecules and is thus proportional to the concentration of the reactants. For a non-reversible reaction the molar rate of creation /destruction of species *i* in reaction *r* is given by:

$$\hat{R}_{i,r} = (v_{i,r}'' - v_{i,r})RR_r = (v_{i,r}'' - v_{i,r})Z_{AB} \exp(-E/RT)$$
(2)

Where  $Z_{AB}$  is the gas kinetic collision frequency and exp(-E/RT) is the Boltzmann factor [Poinsot 2001]. Kinetic theory shows the Boltzmann factor gives the fraction of all collisions that have energy greater than *E*. Considering Eq. (3) and referring to E as activation energy, attention is focused on the collision rate  $Z_{AB}$ .

#### COLLISION FREQUENCY IN ARRHENIUS LAW

The kinetic theory of gases assumes that molecules in a gas consist of rigid, hard spheres of mass m and diameter d in continuous, randomly directed translational motion. Collisions between molecules are instantaneous, and the molecules travel in straight-line trajectories between collisions until randomly encountering another collision partner. The frequency of molecular collisions is an important factor involving gas-phase reaction rates. The kinetic energy associated with the velocity of one molecule relative to another is important in understanding molecular collisions. In particular, the distribution of relative velocities obeys the Maxwell-Boltzmann distribution and the average relative speed is obtained as [Kee 2003]:

$$v_{\rm rel} = \sqrt{\frac{8k_BT}{\pi m_{12}}} \tag{3}$$



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transport properties. The average distance traveled between collisions (called the mean-freepath) is given by:

$$L = \frac{1}{\sqrt{2\pi}d^2(N/V)} \tag{4}$$

The density of molecules is written in terms of the number of molecules, N, in a given volume V. Finally, for a container of molecules (without dominant flow field), the number of molecules colliding with unit area of a wall per unit time is:

$$Z_w = \frac{V_{rel}}{4} \left(\frac{N}{V}\right) = \left(\frac{k_B T}{2\pi m}\right)^{1/2} \left(\frac{N}{V}\right) \tag{5}$$

Based on molecules colliding number, the collision rate in Arrhenius law (for one order reaction) is written as [Poinsot 2001, Kee 2003]:

$$Z_{AB}_{tur} = [C_{1,r}][C_{2,r}]d^2 \left(\frac{8\pi k_B T_{tur}}{m}\right)^{1/2}$$
(6)

It is noticed that this equation calculates the collision frequency in the container of molecules with random motion due to the gas temperature. In turbulent flow, random molecular motions (with Gaussian distribution) are extremely increased resulting in higher collision rate. Fluctuations of velocity in a turbulent swirl burner are shown in Fig. (1) [Al-Abdeli 2006]. Therefore, turbulent parameters must be used to calculate the collision rate. Consideration of transport phenomena shows that turbulent viscosity can be used to correct the collision rate in turbulent flow.



Figure 1. Turbulent velocity in a swirl burner [Al-Abdeli 2006]



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#### TRANSPORT COEFFICIENTS/PHENOMENA

Collisions between molecules in the gas phase can transfer momentum and energy between the collision partners, or lead to net transport of mass from one part of the system to another. In laminar flows, velocity and scalars have well-defined values. In contrast, turbulent flows are characterized by continuous fluctuations of velocity, which can lead to variations in scalars such as density, temperature, and mixture composition [Warnatz 2006]. The Reynolds-averaged conservation equations that are derived from Navier-Stokes equations, allow the simulation of turbulent reacting flows [Warnatz 2006, Hejazi 2007]:

$$\frac{\partial \bar{\rho}}{\partial t} + \operatorname{div}(\bar{\rho}\tilde{\vec{v}}) = 0 \tag{7}$$

$$\frac{\partial \bar{\rho} \widetilde{\mathbf{w}_{1}}}{\partial t} + \operatorname{div}(\bar{\rho} \vec{\tilde{\mathbf{v}}} \ \widetilde{\mathbf{w}_{1}}) + \operatorname{div}(-\bar{\rho} D_{1} \operatorname{grad} \mathbf{w}_{1} + \bar{\rho} \vec{\tilde{\mathbf{v}}}^{"} \mathbf{w}_{1}^{"}) = \overline{M_{1} \omega_{1}}$$

$$\tag{8}$$

$$\frac{\partial \bar{\rho} \vec{v}_{l}}{\partial t_{i}} + \operatorname{div}(\bar{\rho} \vec{\tilde{v}} \otimes \vec{\tilde{v}}_{l}) + \operatorname{div}(\bar{\bar{p}} - \overline{\rho \mu_{l}} \operatorname{grad} \vec{\tilde{v}} + \overline{\rho} \vec{v}^{"} \otimes \vec{v}_{l}^{"}) = \bar{\rho} \ \bar{\vec{g}}$$

$$\tag{9}$$

$$\frac{\partial \bar{\rho} h}{\partial t} - \frac{\partial \bar{p}}{\partial t} + \operatorname{div}(\bar{\rho} \vec{\tilde{v}} \ \tilde{h}) + \operatorname{div}(-\overline{\lambda_{l} \operatorname{grad} T}) + \overline{\rho \mathbf{v}^{"} \mathbf{h}^{"}} = \overline{q_{r}}$$
(10)

In order to solve the closure problem, models are proposed, describing the Reynolds stresses  $\overline{\rho v'' q''}$  in terms of the dependent variables. Two equation turbulent models interpret  $\overline{\rho v'' q''}$  as turbulent transport and model it in analogy to in the same manner as the laminar case, using a gradient-transport assumption which states that the term is proportional to the gradient of the mean value of the property,

$$\rho v'' q'' = -\bar{\rho} v_T grad \,\tilde{q}_i \tag{11}$$

Where  $v_T$  is called the turbulent exchange coefficient [Lipatnikov 2005]. Defining effective exchange coefficient ( $v_{eff}$ ) and substituting Eq. (12) into Eq. (9-11), gives:

$$\frac{\partial \bar{\rho}}{\partial t} + \operatorname{div}(\bar{\rho}\tilde{\vec{v}}) = 0 \tag{12}$$

$$\frac{\partial \bar{\rho} \widetilde{w}_{i}}{\partial t} + \operatorname{div}(\bar{\rho} \widetilde{\vec{v}} \ \widetilde{w}_{i}) - \operatorname{div}(\bar{\rho} \ D_{\text{eff}} \ \text{grad} \ \widetilde{w}_{i}) = \overline{M_{i} \omega_{i}}$$
<sup>(13)</sup>

$$\frac{\partial \bar{\rho} \vec{\tilde{v}}}{\partial t} + \operatorname{div}(\bar{\rho} \vec{\tilde{v}} \otimes \vec{\tilde{v}}) - \operatorname{div}(\bar{\rho} \,\mu_{\text{eff}} \,\text{grad}\, \tilde{v}) = \overline{\rho \vec{g}}$$
<sup>(14)</sup>

$$\frac{\partial \bar{\rho}\tilde{h}}{\partial t} - \frac{\partial \bar{p}}{\partial t} + \operatorname{div}(\bar{\rho}\tilde{\vec{v}} \ \tilde{h}) - \operatorname{div}(\bar{\rho} \ k_{\text{eff}} \operatorname{grad} \tilde{\vec{h}}) = \bar{q}_{\text{r}}$$
<sup>(15)</sup>

Transport coefficients appear as parameters in the macroscopic conservation equations for momentum, energy, and mass. In laminar flow, the viscosity coefficient ( $\mu_l$ ), thermal conductivity ( $k_l$ ) and diffusion coefficient ( $D_l$ ) are defined according to molecular collisions [Kee 2003, Magnussen 1976]. It is usually considered that turbulent transport is much faster than the laminar transport process [Warnatz 2006]. The analysis of momentum transfer in the gas was used to obtain an expression for the viscosity coefficient [Kee 2003, Kee 2004]:





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(16)

$$\mu = \frac{5}{16} \frac{\sqrt{\pi m_k k_B T}}{\pi \sigma^2 \Omega_{kk}^{(2,2)*}}$$

where  $\sigma$ , the low-velocity collision cross-section for the species of interest in Angstroms; W, the molecular weight; T, the temperature in Kelvin; and  $\Omega^{(2,2)*}$ , the reduced collision integral, a function of reduced temperature  $T^*$  ( $T^* = T/(\varepsilon/k)$ ) where ( $\varepsilon/k$ ) is the potential parameter for the species of interest).

Furthermore effective viscosity in turbulent flow is defined by sum of laminar and turbulent viscosity. Turbulent viscosity is modeled by using turbulent models. In k- $\epsilon$  turbulent model, turbulent viscosity is calculated by Eq. (18) [Bray 2006]:

$$\mu_{\rm t} = C_{\mu} \frac{\tilde{k}^2}{\tilde{\epsilon}} \tag{17}$$

Comparing Eq. (17) and Eq. (18), we can define the turbulent temperature parameter ( $T_{tur}$ ), in which the collision rate of gas molecules in a container equals the collision rate in turbulent flow of viscosity  $\mu_r$ :

$$\mu_{t} = \frac{5}{16} \frac{\sqrt{\pi m_{k} k_{B} T_{tur}}}{\pi \sigma^{2} \Omega_{kk}^{(2,2)*}} \implies (18)$$

$$T_{tur} = \frac{\left(16 * \mu_{t} * \pi \sigma^{2} \Omega_{kk}^{(2,2)*} / 5\right)^{2}}{\pi m_{k} k_{B}}$$
(19)

The reduced collision integral represents an averaging of the collision cross-section over all orientations and relative kinetic energies of colliding molecules. The values of this integral are tabulated for various reduced temperature by Camac and Feinberg [Kee 2004]. The values can be approximated to within 2% (for  $T^* > 2.7$ ) by the expression:

$$\Omega_{\rm kk}^{(2,2)*} = 1.2516({\rm T}^*)^{-0.1756} \tag{20}$$

Evaluating the Eq. (18) at  $T_{ref}$  and using this as a reference point results in the following expression for the viscosity:

$$\mu = \mu_{ref} (T/T_{ref})^{0.6756} \tag{21}$$

Accordingly, Eq. (20) is simplified to:

$$T_{tur} = T(\mu_t/\mu_l)^{1/0.6756}$$
(22)

where  $\mu_l$  is gas viscosity at temperature T. By substituting of turbulent temperature in Arrhenius expression (Eq. (8)), we can use of Eq. (7) for calculating rate of reaction in turbulent flow.



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CASE STUDY

In order to assess the capability of the modified model for investigation of axial methane-air burner, two cases are studied here: a) one step, b) two step reaction mechanism of  $CH_4$ / Air. The temperature and CO mass fraction distributions on the axial and radial positions of the burners are compared to the modified Arrhenius, Eddy dissipation and PDF models.

The burner is modeled as turbulent 2D axisymmetric non-premixed CH<sub>4</sub>/air ones with the inlet fuel and air flow rates of 14.5 kg/hr and 336 kg/hr, respectively. In the ref. [Hejazi 2007] the test case is documented in greater detail. Steady state simulations are performed on a cylindrical coordinate with a grid space of 0.3 mm in the radial direction and 0.6 mm in the axial direction and an expansion ratio of 1.02. Showing a small variation in results (< 1% in temperature values) on finer mesh simulations, proved it to be grid-independent.

A comparison of the temperature profile at axis and outlet of furnace between modified Arrhenius, eddy dissipation model and the PDF model is presented in Fig. 2, 3. It is noteworthy that the results of PDF model have a very good agreement by experimental result [Bray 2006]. It is seen that the difference between the results of the modified Arrhenius and eddy-dissipation model for one step reaction is very low.

In Fig. 4, turbulent viscosity in furnace is shown. In high turbulence viscosity region, collision rate of gas molecules are increased.

Fig. 5 demonstrates the collision frequency and Boltzmann factor in the modified model in compare with the Arrhenius model. It is seen that the Boltzmann factor in turbulent flow is higher than laminar flow with a sharp increase at the fuel inlet. Collision frequency also shows the same tendency (up to 50 times higher than the laminar flow).



Figure 2. The comparison of Temperature distribution on axis of furnace (one step reaction mechanism)



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Figure 3. The comparison of Temperature distribution at outlet (one step reaction mechanism)

1.37E-03	7.13E-04	1.81E-03		3.34E-03		4.00E-03
	4.44E-03		4.00E-03		4.00E-03	3.78E-03
1.37E-03	4.94E-04	2.03E-03		3.34E-03		4.00E-03

Fig. 4. Turbulent viscosity contour in axial burner



Figure 5. Comparison between collision frequency (ZAB) and the Boltzmann factor, e(-E/RT), in the turbulent-to-laminar Arrhenius expression





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The two-step chemistry takes into account the formation of CO as an intermediate in CO2 formation and allows a better prevision of the temperature. At case (b), CH4/Air flame is simulated by two step global reaction. Fig.6 presents the calculated mass fractions of CO at different position of furnace. Comparison between predicted mass fraction of CO by different models show that the predictions using modified Arrhenius are in better agreement with prediction using the PDF model compared with those obtained with eddy-dissipation model.



Figure 6. Comparison of CO mass fraction at two position of furnace

# CONCLUSIONS

In this paper, the modification of Arrhenius model which is based on the molecular collision theory is discussed. In contrary to the eddy dissipation model, which yields incorrect results for multi-step reaction mechanisms, it is considered as a detailed reaction mechanism model in laminar flow investigations. Therefore, any improvement in this method for turbulent flow can have far reaching implications.

Based on this work, the following conclusions can be drawn:

- Collision rate in turbulent flow can be expressed in term of turbulent viscosity.
- Due to dependency of momentum transfer on the turbulent viscosity and molecular collision, turbulent viscosity is used to modify the collision frequency and the Boltzmann factor.
- Having revised the Arrhenius model, the results for one-step reaction mechanisms approach to the eddy-dissipation results.
- In multi-step reaction mechanisms, modified Arrhenius model is preferred to Magnussen model.

The modified model of Arrhenius is better than eddy-dissipation model to model turbulent reacting model by detailed reaction mechanisms.



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