

ROLE OF MULTICOMPONENT DIFFUSION IN PREMIXED AND NONPREMIXED LAMINAR FLAMES

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Introduction

Existing models of microscale mixing in turbulent combustion do not address the issue dealing with the role of multicomponent diffusion (MD) in molecular transport models. Usually, various approximations are applied for diffusion tensor D_{ij} : either diffusion coefficients are taken equal for all species or, at most, the binary diffusion coefficients of each species in a diluent are used. With direct numerical simulation (DNS), it has been possible to analyze different interactions between turbulence and chemical kinetics; however, none of available studies addressed the issue, evidently due to current restrictions in CPU power.

It has been commonly adopted that reduced and overall kinetics with several stable species are sufficient to adequately represent thermodynamics and heat release in premixed and nonpremixed flames. However, for correct predicting pollutant formation and autoignition, it is often necessary to involve extended reaction mechanisms including atoms, radicals, and stable molecules. In such applications, MD will inevitably manifest itself because combustible mixtures contain species with highly diverse molecular weights. The objective of this presentation is to show some applications where MD can play the important role.

Premixed Flames

Laminar hydrogen–oxygen premixed flame is a simple example illustrating importance of MD [1]. The results of detailed calculations of premixed stoichiometric flame structure by using various approximations for diffusion tensor D_{ij} of 8 participating species are shown in Table 1.

Table 1 Results of detailed calculations of premixed, stoichiometric, laminar hydrogen–oxygen flame structure by using various approximations for diffusion tensor D_{ij} of 8 participating species [1]

Variant	Flame speed, m/s	Error, %	Material misbalance, %
Experiment [2]	8.70	—	—
Multicomponent diffusion [1]	8.78	0.9	0
Approximation A [3]	9.17	5.4	0
Approximation B (diagonal elements of tensor D_{ij} = binary diffusion in H_2)	9.44	8.5	2.5
Approximation C (diagonal elements of tensor D_{ij} = binary diffusion in H_2O)	8.41	3.3	0.7
Approximation D_{ij} (independent diffusion in H_2)	8.42	3.2	4.2

It follows from the table that various approximations of D_{ij} result in violation of local material balance $\Delta = |\sum y_i - 1|$ up to about 4%, where y_i is the species mass fraction. This means that MD is important if species with small concentrations (e.g., atoms and radicals) are of interest.

Autoignition

The other example is preflame autoignition in spark-ignition engine or controlled autoignition in Homogeneous Charge Compression Ignition (HCCI) engine. Analysis of chemical mechanisms of low-temperature n -heptane [4, 5] and methane [6] autoignition reveals the important role of alkylperoxide and hydrogen peroxide decomposition reactions at the

initial stages of fuel oxidation leading to cool and blue flame manifestation. Propagation of cool and blue flames is usually not considered in relevant autoignition studies; however, there is no fundamental substantiation of their minor role in the spatial development of low-temperature autoignition. Diffusion of mobile active species formed in autoignition centers can promote chemical activity in neighboring volumes and contribute to the overall evolution of the reactive mixture.

Liquid Drop Flames

Figure 1 shows the results of detailed calculations of *n*-heptane drop vaporization in air [7]. Solid curves correspond to the solution of governing equations with regard for MD in gas phase. Dashed curves correspond to the solution of the same problem based on the independent diffusion approximation for D_{ij} with nitrogen taken as a diluent. Such an approximation results in a significant material misbalance (up to 40%) in the vicinity to drop surface, and considerable errors in predicted species concentrations. With using the assumption on the same diffusion coeffi-

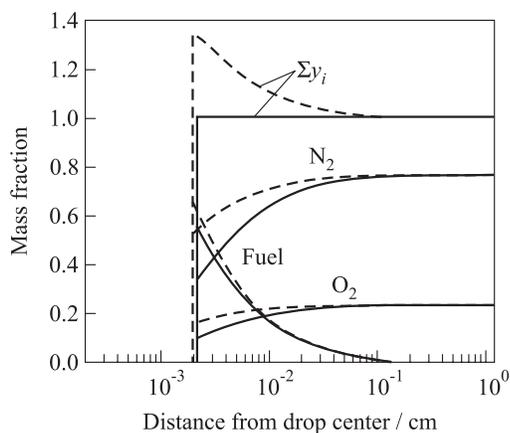


Figure 1 Results of detailed calculations of *n*-heptane drop vaporization in air [7]. Initial drop diameter is 70 μm , initial temperatures of gas and drop are 573.15 and 293.15 K, pressure is 0.1 MPa, time is 24 ms

cients for all species involved, the material misbalance is zero; however, the temporal and spatial dynamics of species concentrations differ considerably from those obtained with regard for MD. Similar results were obtained for burning drops.

Spray Effects

Vaporization and combustion of liquid sprays are usually considered in the approximation of d^2 -law for individual fuel drops. However, this law has been obtained in the classical theory [8] for the isolated drop under far reaching simplifications. Figure 2 shows the results of detailed modeling of n -tetradecane drop vaporization with regard for spray effects in a homogeneous monodisperse drop suspension in air [9]. The spray effects were modeled by using proper boundary conditions at the outer boundary of an elementary cell around a drop. These boundary conditions ensure the “screening” effect of neighboring drops on mass and

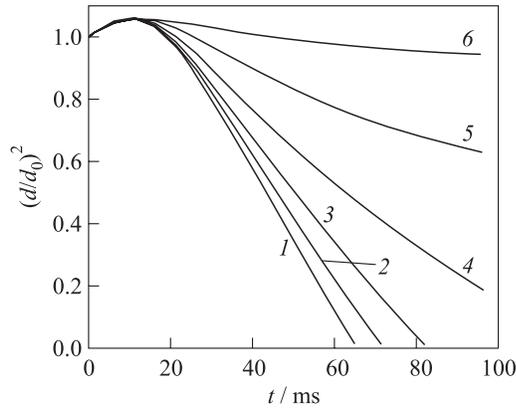


Figure 2 Predicted surface regression rates during vaporization of n -tetradecane drop in air depending on the local equivalence ratio Φ (distance between drops) [9]: 1 — $\Phi = 0$ (∞), 2 — 0.6 (40.0), 3 — 1.2, (31.5), 4 — 2.4 (25), 5 — 4.75 (20), and 6 — $\Phi = 9.5$ (15.7). Initial drop diameter is $70 \mu\text{m}$, initial temperatures of gas and drop are 573.15 and 293.15 K, pressure is 0.1 MPa

energy fluxes and constant pressure in the medium. Three important findings are worth mentioning with respect to Fig. 2.

First, spray effects result in the slower vaporization of liquid drops as compared to the isolated drop vaporization in an unconfined atmosphere. Second, the classical d^2 -law does not hold in general, in particular for heavy hydrocarbon fuels like *n*-tetradecane (usually used for modeling Diesel oil). Third, there exists the equivalence ratio starting from which drops vaporize only partly (curves 5 and 6 in Fig. 2). Note that fuel vapor distribution in the space around the drop in these calculations evolves in time with regard for MD. Similar to the isolated drop considered in the previous section, the simplified modeling of molecular diffusion results in considerable errors in mass balance and/or local species concentrations. These features manifest themselves at spray autoignition and combustion too.

Concluding Remarks

Modeling of combustion phenomena with finite-rate chemistry (lean/rich combustion, pollutant formation, autoignition, etc.) requires careful coupling of molecular mixing models with reaction mechanisms involving atoms, radicals, and stable molecules. As these species possess highly diverse molecular weights, the neglect of possible effects of MD becomes the issue. Several examples demonstrating the importance of this issue in laminar premixed and nonpremixed flames are presented in the paper. In view of it, and taking into account that many available DNS studies of turbulent combustion apply a single progress variable and assume a constant molecular weight, it is reasonable to pose a question on the range of validity of such studies. This is particularly important as DNS tends to become a main tool for validating various mixing models.

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