



# On the application of betweenness centrality in chemical network analysis: Computational diagnostics and model reduction



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## ARTICLE INFO

### Article history:

Received 29 January 2015

Received in revised form 13 May 2015

Accepted 14 May 2015

Available online 2 June 2015

### Keywords:

Shortest path

Betweenness centrality (BC)

Chemical reaction network

Species ranking

Mechanism reduction

Computational diagnostics

## ABSTRACT

In this work, the concept of shortest path and betweenness centrality (BC) is introduced in combustion systems, with its application demonstrated in chemical network analysis and skeletal mechanism generation. After establishing the graphical representation of a chemical network for given pressure, temperature, and species concentrations, a metric BC is defined to rank the shortest paths passed by other nodes between any pair of nodes in the network for computational diagnostics. Consequently, the controlling pathways are retained by collecting the species with larger BC values. Such a notion then indicates that the index of importance of species could then be assigned based on their BC values to further guide skeletal mechanism generation. Unlike existing methods, the betweenness centrality approach takes into account of both the fluxes between species and their relative positioning within the chemical network. To demonstrate its potential utility to combustion studies, the approach was applied to the GRI-3.0, LLNL and USC-Mech II mechanisms to identify the important pathways in the chemical network at each local reaction state, and develop skeletal mechanisms from all reaction state samples in auto-ignition and perfectly stirred reactor (PSR) simulations. The performance of the BC ranking is compared to the methods of directed relation graph (DRG), DRG with error propagations (DRGEP) and sensitivity analysis (SA), and is shown to possess sufficient utility in producing skeletal mechanisms with good accuracy and flexibility for the cases studied.

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## 1. Introduction

Chemical kinetics of fuels oxidation invariably involve a large number of strongly-coupled species and reactions, which not only impose considerable difficulty in identifying the controlling entities, but also require high computational cost to attain the understanding, validation, and prediction of the underlying complex chemical and flame processes [1]. Consequently there exists considerable interest in developing computational tools for the diagnostics of the processes and phenomena of interest, and for the reduction of the size and complexity of the detailed reaction mechanisms describing them. For computational diagnostics, sensitivity analysis (SA) [2], computational singular perturbation (CSP) [3] and its extension to the chemical explosive mode analysis (CEMA) [4] have been developed to identify important species and reactions for global and local parameters. However, there are many situations for which these methods are either not feasible

or capable of providing the information needed. For example, in the direct numerical simulation (DNS) of a turbulent flame, it is not practical to perturb a certain reaction rate constant and redo the simulation to obtain the sensitivity coefficients. CEMA is a useful tool to obtain the explosive mode of the local chemical Jacobian matrix and the corresponding contribution from each species and reaction; however, it is not able to be applied to mixtures without a chemical explosive mode or indicate the important pathway associated with the production of certain species.

Regarding mechanism reduction, methods based on graph theory, notably the directed relation graph (DRG) [5–7], and the subsequent extensions of path flux analysis (PFA) [8] and DRG with error propagation (DRGEP) [9], have been developed, resulting in substantial simplifications in reducing the model size and the subsequent application in combustion modeling [10–14]. These methods use different criteria to rank species based on the sampling of reaction states, locally characterized by temperature, pressure, and species concentrations. The species rankings are subsequently used to guide the elimination of species from a chemical mechanism. A systematic comparison of the DRG-based methods can be found in [15].

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In the present work, the concept of betweenness centrality (BC), which measures the relative connectivity of a node in a network based on the shortest paths [16], is adopted to study the complex chemical networks in combustion systems [17]. Our interest in exploring the potential of this concept in chemical systems is motivated by recognizing that over the past decade this concept has been widely and fruitfully employed in the analysis of equally complex networks that exist in, for example, social science [18], computer science [19], biology [20], transport [21], scientific cooperation [22], and interdisciplinary interactions [23]. In the following we shall first present the formulation of the concept of BC within the framework of the chemical network. We shall then demonstrate its utility in autoignition as well as perfectly stirred reactor (PSR) systems, and compare its performance with those of DRG, DRGEP and SA. It will be shown that the concept of BC, which considers both fluxes and the relative positioning of species within a chemical network, provides useful information of indirect kinetic influence between any pair of species, and further generates a species ranking that both effectively quantifies the relative importance of species and predicts target parameters in combustion simulations with considerable accuracy and flexibility.

## 2. Graphical representation and methodology

Different from graph representation in the DRG based methods, the forward and backward reactions of each elementary reaction are treated separately so as to retain as much kinetic information as possible. A graphical representation of a chemical network is chosen such that each node represents a chemical species and an edge of weight  $L_{AB}$  between nodes A and B is defined as the sum of the reaction rates, with species A being a reactant and B a product:

$$L_{AB} = \sum_{i=1}^N \nu_{A,i} (\omega_{f,i} \delta_{f,i}^{AB} + \omega_{b,i} \delta_{b,i}^{AB})$$

where  $\omega_{f,i}$ ,  $\omega_{b,i}$  are the forward and backward reaction rates, respectively, for the  $i$ th reaction, and  $\nu_{A,i}$  is the stoichiometric coefficient for species A in the  $i$ th reaction.  $\delta^{AB} = 1$  if and only if the forward or backward direction for the  $i$ th reaction includes A as a reactant and B as a product, and  $\delta^{AB} = 0$  otherwise. Such a definition denotes a lumped consumption rate of species A for the formation of species B and the adjacency matrix  $L$  is asymmetric.

Recognizing that a species can participate in many elementary reactions, for an arbitrary reversible reaction  $R_{AB,l}$  with index  $l$  involving both A and B, which can be in the form of either  $\nu_A A + \nu_M M \leftrightarrow \nu_B B + \nu_N N$  or  $\nu_A A + \nu_B B \leftrightarrow \nu_M M + \nu_N N$ , the consumption rate of A for the formation of B through either form of  $R_{AB,l}$  can be expressed as:  $\nu_{A,l} (\omega_{f,l} \delta_{f,l}^{AB} + \omega_{b,l} \delta_{b,l}^{AB})$ .

Therefore, the ratio of the consumption rate of A due to the formation of B through reaction  $R_{AB,l}$  to the total consumption rate of A is:

$$P(R_{AB,l}) = \frac{\nu_{A,l} (\omega_{f,l} \delta_{f,l}^{AB} + \omega_{b,l} \delta_{b,l}^{AB})}{\sum_{i=1}^{N_s} \sum_{j=1}^{N_r} \nu_{A,j} (\omega_{f,j} \delta_{f,j}^{AM_i} + \omega_{b,j} \delta_{b,j}^{AM_i})} \quad (1)$$

where the subscripts  $i$  and  $j$  designate the  $i$ th species and the  $j$ th reaction, respectively. Assuming that each elementary reaction between A and B occurs independently, the normalized rate for the paths from nodes A to B can be measured as:

$$P_{AB} = P(R_{AB,1}) + P(R_{AB,2}) + \dots + P(R_{AB,n}) \\ = \frac{\sum_{j=1}^{N_r} \nu_{A,j} (\omega_{f,j} \delta_{f,j}^{AB} + \omega_{b,j} \delta_{b,j}^{AB})}{\sum_{i=1}^{N_s} \sum_{j=1}^{N_r} \nu_{A,j} (\omega_{f,j} \delta_{f,j}^{AM_i} + \omega_{b,j} \delta_{b,j}^{AM_i})} = \frac{L_{AB}}{\sum_{i=1}^{N_s} L_{AM_i}}$$

Although tracking the flux of element in each elementary reaction would be difficult, the above definition then shows the physical meaning of the fraction of the consumption of species A by the formation of species B. It is noted that there is a major difference in the manner of species B interacting with species A from DRG and its extensions. Specifically, species A must appear on the reactant side and B must appear on the product side in a reaction and hence it carries the information of the direction of flux from A to B. As a result, the normalized rate of such a transfer along any pathway between two nodes (from source  $M_1$  to target  $M_n$ ) can be modeled as:

$$P(M_1 \dots M_n) = \prod_{i=1}^{n-1} P_{M_i M_{i+1}}$$

This quantity then measures the indirect influence of the source node on the target node in a chemical network. It is not unreasonable to expect that the larger the value of  $P$  along a certain path, the more possible is the transfer from the source to the target. Therefore, it would be of interest to investigate paths with large  $P$  values including the most probable path, connecting any two nodes of interest in the reaction network. As will be shown in the following sections, the pathways identified generate useful information of reaction paths especially for nodes without direct connections, which so far cannot be systematically obtained for computational diagnostic purposes.

We now introduce the BC concept in the adjacency matrix  $P$  to describe the effectiveness of a node in connecting any other two nodes as:

$$W_k = \sum_{s \neq t \neq k} \frac{p_{st}(k)}{p_{st}} = \sum_{s \neq t \neq k} \frac{p_{sk} \cdot p_{kt}}{p_{st}}$$

where  $p_{st}$  is the most probable path from nodes  $s$  to  $t$  when traveling through the adjacency matrix  $P$  by any possible path, i.e.,  $p_{st} = \max P(s, \dots, t)$  for all possible paths from  $s$  to  $t$ . The BC is defined for each node  $k$  as the summation of the ratio of the most probable path from nodes  $s$  to  $t$  to the most probable path from nodes  $s$  to  $t$  passing through node  $k$ , which considers how frequently a node serves as a hub between other nodes.

In order to numerically evaluate BC efficiently, by further defining a new matrix  $A$  as  $A_{ij} = -\ln P_{ij}$ , it is readily shown that

$$e^{-\text{path length of } ABC} \equiv e^{-A_{AB} - A_{BC}} = e^{\ln P_{AB} + \ln P_{BC}} = P_{AB} P_{BC} = P_{ABC}$$

which means that the shortest path in  $A$  corresponds to the most probable path in  $P$ . And such a shortest path,  $\sigma$ , can be readily found by using Dijkstra's algorithm [24] with a time complexity of

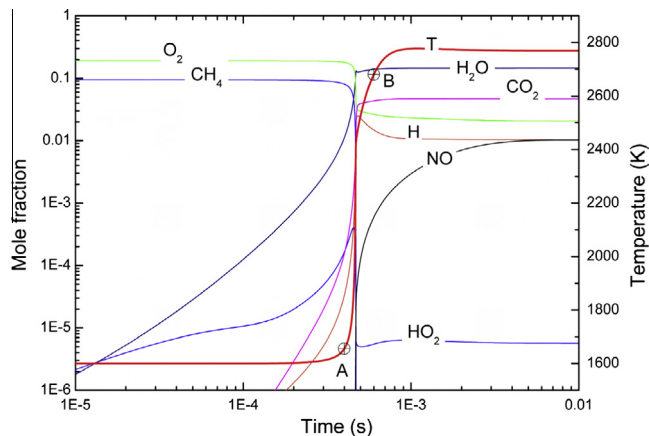


Fig. 1. Temperature and mole fraction history of selected species for stoichiometric  $\text{CH}_4/\text{air}$  autoignition at initial temperature 1600 K, pressure 1 atm.

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