A REACTOR NETWORK MODEL FOR PREDICTING NOx EMISSIONS IN AN INDUSTRIAL BURNER

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Abstract. *A chemical reactor network (CRN) is developed and applied to the modeling of a natural gas burner. The CRN development is based on experimental and CFD simulation results of the BERL 300 kW test. The CRN element arrangement, reactor volumes and flow splits are adjusted based on the best agreement with characteristic temperatures of the reactive flow, aiming to reproduce the experimental NOx emissions data. A sensitivity analysis and a parametric study of the CRN are carried out to evaluate its sensitivity to the flow splits adjustments and its ability to predict emission with air preheat and turndown operation. The NOx emissions predicted by CRN are in good agreement until 110ºC preheating and for a turndown ratio of 1,33.*

Keywords*: chemical reactor network, NOx emissions, BERL 300 kW*

1. INTRODUCTION

Detailed studies of reactive flows became possible in the late 20th century thanks to the development of advanced researching tools like the non-intrusive measurement techniques and the computational fluid dynamics. The CFD codes, based on the finite volume method, provide an accurate description of the flow field but its performance in the prediction of low concentration pollutants, like NOx, is limited by the computational power required to solve mass, energy, momentum and chemical species balances in refined meshes (Falcitelli et al., 2002). An alternative reactive flow modeling is known as chemical reactor networks (CRN), which involve chemical engineering models of ideal reactors, like the perfectly stirred reactor and the plug flow reactor, to describe the flow field as an arrangement of reactors where detailed reaction mechanisms can be applied.

According to Novosselov (2006), the use of chemical reactor models to study combustion processes started in 1953 with the work of S. L. Bragg, who modeled a gas turbine combustor as an association of a perfectly stirred reactor (PSR) followed by a plug flow reactor (PFR). In 1970 Swithenbank proposed the zonal combustion modeling, considering the combustor volume divided into zones represented by ideal reactors, improving the combustor design methods used so far, based on correlations. In the last two decades, the development of CRNs improved greatly with the aid of the CFD codes, which provide extensive information on turbulent mixing and residence time distribution of complex reactive flows, allowing the development of CFD-to-CRN algorithms (Falcitelli et al., 2002; Fichet et al., 2010).

Since there is no specific set of rules to determine the development of a CRN, the analysis of each reactive flow depends upon the amount of numerical and experimental information available. This paper presents an application of ideal reactors to the modeling of a natural gas burner. The CRN development is based on experimental and CFD simulation results of the BERL 300 kW test. The CRN element arrangement, reactor volumes and flow splits are adjusted based on the best agreement with characteristic temperatures of the reactive flow, aiming to reproduce the experimental NOx emissions data. A sensitivity analysis and a parametric study of the generated CRN are carried out to evaluate its sensitivity to the flow splits adjustments and its ability to predict emission with air preheat and turndown operation.

2. CHEMICAL REACTOR NETWORKS AND SIMULATION TOOL

The CRN elaborated in this work is formed by an arrangement of two kinds of ideal reactors: the perfectly stirred reactor (PSR) and the plug flow reactor (PFR). The PSR is an ideal reactor of arbitrary form that neglects mixing phenomena. In a system where either the mixing rates are high or the chemical reaction rates are slow, the chemical kinetics constrains the burning rates in the mixture and the residence time is the most important parameter of the reactor (Turns, 2000). The hypothesis of perfect mixing means that there is no dependence on flow parameters, hence there is no conservation of momentum. Also, with steady-state operation, the mass, energy and species balance equations are time independent. As a result of these assumptions, the PSR is described by a set of nonlinear algebraic equations instead of a system of nonlinear partial differential equations, and its solution is possible using a Newton-Raphson solver (Fogler, 2006).

The plug flow reactor is an ideal reactor that assumes steady, one-dimensional, inviscid flow of ideal gas. The main assumption implies that there is no mixing in the axial direction and each cross-section presents homogeneous

conditions. The hypotheses considered reduce the coupled, nonlinear, three-dimensional governing equations to a set of coupled ordinary differential equations.

The simulations of the CRN were carried out in the CHEMKIN 4.0™ software, a chemistry simulation tool which allows the adoption of extensive chemical reaction mechanisms and employs a modified damped Newton's method to solve the nonlinear algebraic equations (PSR) and the finite difference approximations of the coupled ordinary differential equations (PFR). The reaction mechanism adopted in this work is the GRI-Mech 3.0, which validation range covers the 1000 to 2500 K of temperature, 1,33 to 1013,00 kPa of pressure and 0,1 to 5,0 of equivalence ratio. This mechanism is adjusted for describing the natural gas ignition and flame propagation, including NO formation and reduction, but omits soot formation and selective non-catalytic reduction of NO (GRI-Mech 3.0).

3. BERL 300 kW TEST

The BERL 300 kW test is one of six tests realized in the SCALING 400 study, which was conducted during the 1990's to assess existing scaling methods and develop improved ones for combustion performance and emissions, based on input-output and detailed in-flame measurements (Hsieh et al., 1998). The velocities, temperatures and species measurements of the vertically fired BERL 300 kW test were used by several authors (Pember et al., 1996; Tiribuzi et al., 1997; Badur et al., 2003; Silva, 2003) to validate their CFD codes. Figure 1 presents the burner and the furnace and Table 1 shows the main parameters of BERL 300 kW test. Figure 2 exhibits the streamlines of the reactive flow.

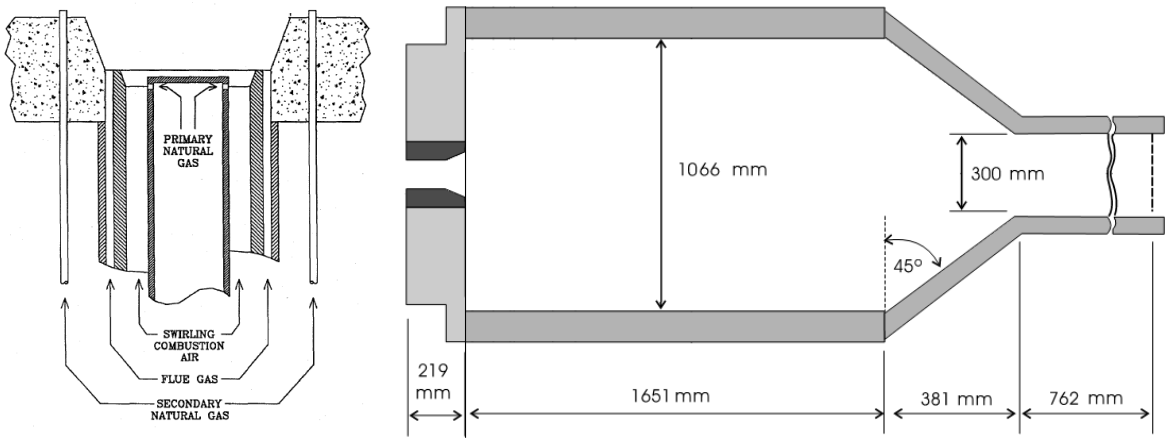

Figure 1. Diagrams of the burner and furnace of the BERL 300 kW test. Adapted from Fornaciari et al., 1994 and Silva, 2003.

Table 1. Operation parameters of the BERL 300 kW test.

Natural gas molar composition	0,965 CH_4 | 0,017 C_2H_6 | 0,020 C_3H_8 0,030 CO_2 | 0,013 N_2
Natural gas flow rate (kg/h)	22,7
Natural gas injection temperature (K)	308,15
Air flow rate – 15% excess (kg/h)	469,15
Air injection temperature (K)	312,15
Heat extraction (relative to thermal input)	41,9%
Flue gas exhaust temperature (K)	1386
NOx emission (g NOx/kg NG)	1,3

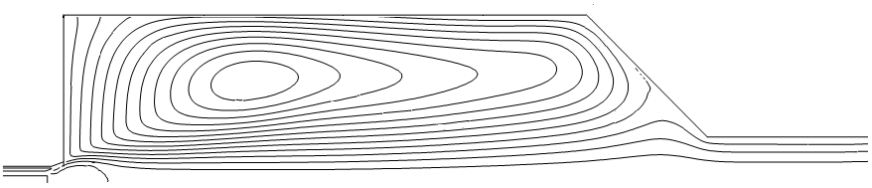

Figure 2. Streamlines of the reactive flow, considering symmetry in the centerline. Adapted from Silva, 2003.

4. CRN DEVELOPMENT AND RESULTS

The development of the CRN for the BERL 300 kW test uses insights gained from the measurements (Sayre et al., 1994) regarding temperature and emissions data, and several CFD and scaling models (Tiribuzi et al., 1997; Hsieh et al., 1998; Bollettini et al., 2000; Silva, 2003), regarding the flow topology and structure. The furnace CRN is constructed based on two main assumptions:

- The sum of reactor volumes is equal to the furnace volume.
- The flow splits and heat extractions are adjusted based on the best agreement with characteristic temperatures on the different regions of the reactive flow.

These assumptions allowed the development of three CRNs, starting with a fundamental PSR+PFR network and then developing new, improved versions of the model. Figure 3 presents the basic CRN network, composed by a PSR to describe the flame zone and a PFR comprising the remaining volume of the furnace. Table 2 exhibits the comparison of experimental data and modeling results for this two-element CRN.

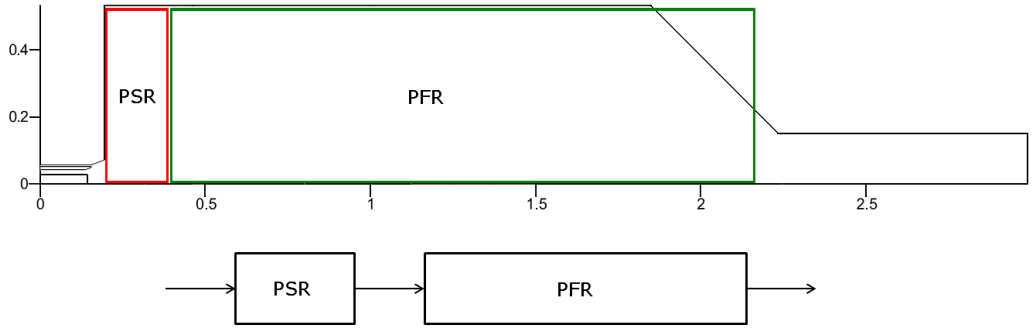

Figure 3. Two-element CRN for the BERL 300 kW test.

Table 2. Comparison of experimental data and modeling results of the two-element CRN.

Quantity	Experimental data	CRN data	Deviation
Flame temperature (K)	1945	1381	28,99%
Flue gas exhaust temperature (K)	1386	1386,4	0,028%
NOx emission (g NOx/kg NG)	1,3	0,023	98,23%

Considering the streamlines presented in Fig. 2, the two-element CRN is unable to describe the flow topology and residence time, hence yielding poor temperature and emissions predictions. The development of a second CRN was based on the work of Pedersen et al. (1997) regarding the characteristic regions of a furnace containing one swirl burner, especially the formation of internal and external recirculation zones. Figure 4 presents the flow topology described by Pedersen.

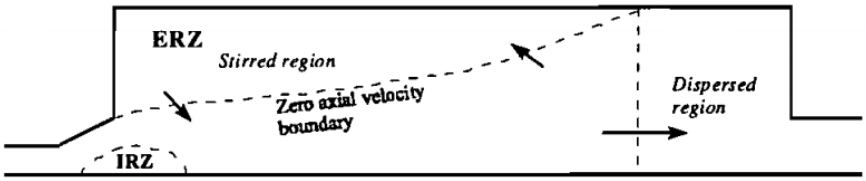

Figure 4. Characteristic regions of a furnace containing one swirl burner. Adapted from Pedersen et al., 1997.

According to the authors, the recirculation and dispersed zones are stirred regions, while the central, post-flame flow is more like a plug flow, this combination of reactors being able to describe the residence time distribution of the furnace. The application of this reactor arrangement, however, was not possible with CHEMKIN™ since its solver do not allow the recirculation of reagents between PSRs and PFRs. Considering this limitation, the second CRN was proposed with a PSR representing the post-flame, enabling the recirculation of reagents between the flow regions. Figure 5 presents this four-element CRN and Tab. 3 shows the experimental e modeling data.

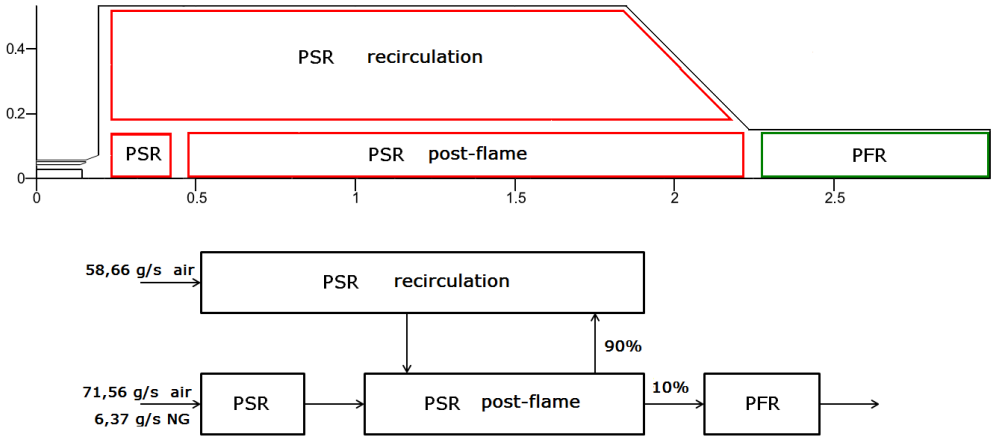

Figure 5. Four-element CRN for the BERL 300 kW test. The percentage values indicate the mass flow fraction in each flow split.

Table 3. Comparison of experimental data and modeling results of the four-element CRN.

Quantity	Experimental data	CRN data	Deviation
Flame temperature (K)	1945	1943,0	0,100%
External recirculation zone temperature (K)	1350	1346,5	0,074%
Flue gas exhaust temperature (K)	1386	1386,7	0,050%
NOx emission (g NOx/kg NG)	1,3	2,37	82,30%

The results presented in Tab. 3 shows a good temperature agreement with the experimental data but the NOx prediction is overestimated. A careful analysis of the CRN indicate that the first PSR, where the combustion starts, is, in fact, comprising the volume and containing the amount of reagents expected to be in the flame front and internal recirculation zone. Thus, the agreement with experimental data is not applied correctly, resulting in a larger amount of reagents reaching temperatures higher than 1900 K, triggering the formation of excessive thermal NOx.

To overcome this situation, a third CRN was developed, employing the strategy of Andreini and Facchini (2004) to describe the near burner region of non-premixed flames. Their CRN, developed to model a conventional diffusion flame gas turbine combustor, was based on the work of Broadwell and Lutz (1998) regarding the structure of jet flames. Figure 6 presents a sketch of the two-reactor model proposed by Broadwell and Lutz.

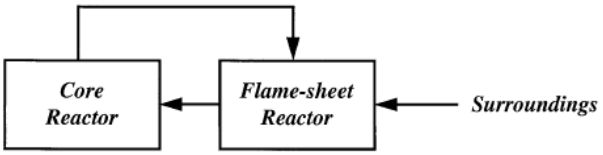

Figure 6. Two-reactor model for jet flames.

The incorporation of this concept in the second CRN resulted in the split of the flame zone PSR into two PSRs: one comprising the flame front, where the reagents are in stoichiometric condition; and one representing the internal recirculation zone, where the remaining mixture is injected. This enhancement resulted in a 5-element network, presented in Fig. 7. Table 4 presents the comparison of experimental data and modeling results.

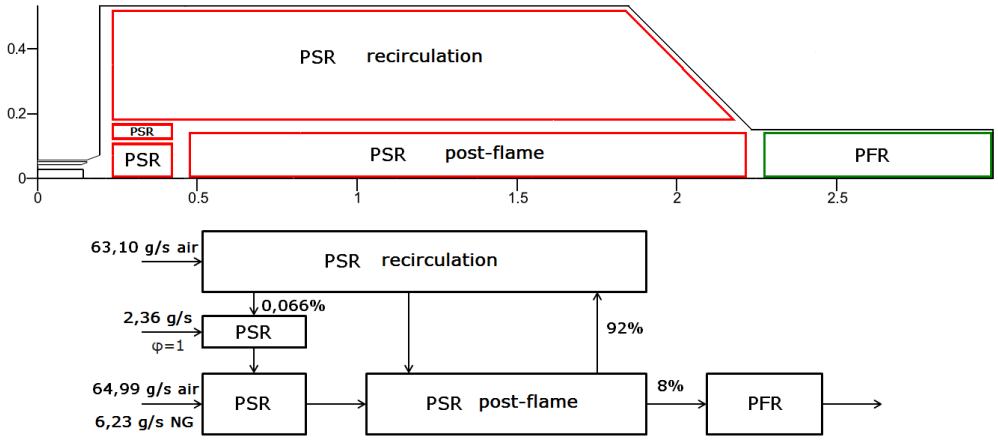

Figure 7. Five-element CRN for the BERL 300 kW test operating in nominal condition. The percentage values indicate the mass flow fraction in each flow split.

Table 4. Comparison of experimental data and modeling results of the BERL 300 kW test.

Quantity	Experimental data	CRN data	Deviation
Flame temperature (K)	1945	1944,4	0,030%
External recirculation zone temperature (K)	1350	1349,2	0,059%
Internal recirculation zone temperature (K)	1850-1900	1885,1	--
Flue gas exhaust temperature (K)	1386	1386,5	0,036%
NOx emission (g NOx/kg NG)	1,3	1,28	1,54%

Considering the accurate reproduction of measured temperatures and NOx emission, this five-element CRN is considered representative of the BERL 300 kW test and henceforward will be applied in sensitivity analysis and parametric study to evaluate its sensitivity to the flow splits adjustments and its ability to predict emission with air preheat and turndown operation.

5. SENSITIVITY ANALYSIS AND PARAMETRIC STUDY

In order to obtain a better understanding of the CRN capabilities and the constraints of the adopted methodology, a sensitivity analysis and a parametric study were conducted. In the sensitivity analysis, the influence of two flow splits on the NOx emissions was assessed: the air split between internal and external recirculation zones; and the amount of flue gas recycled from the post-flame to the external recirculation zone. The parametric study evaluates the models ability to reproduce NOx emissions for different operation conditions like air preheating and turndown operation. Figure 8 presents the results of the sensitivity assessments. Figure 4 shows the results of the parametric study.

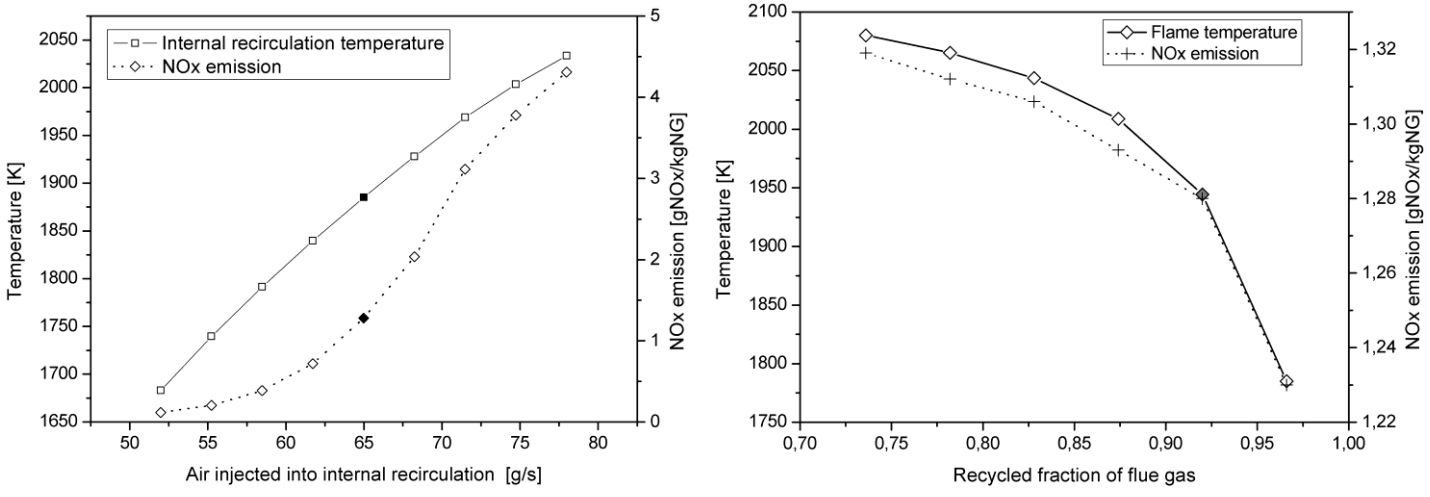

Figure 8. Sensitivity analysis of the CRN to air injection into internal recirculation (left) and to recycled fraction of flue gas (right). Filled markers indicate nominal condition.

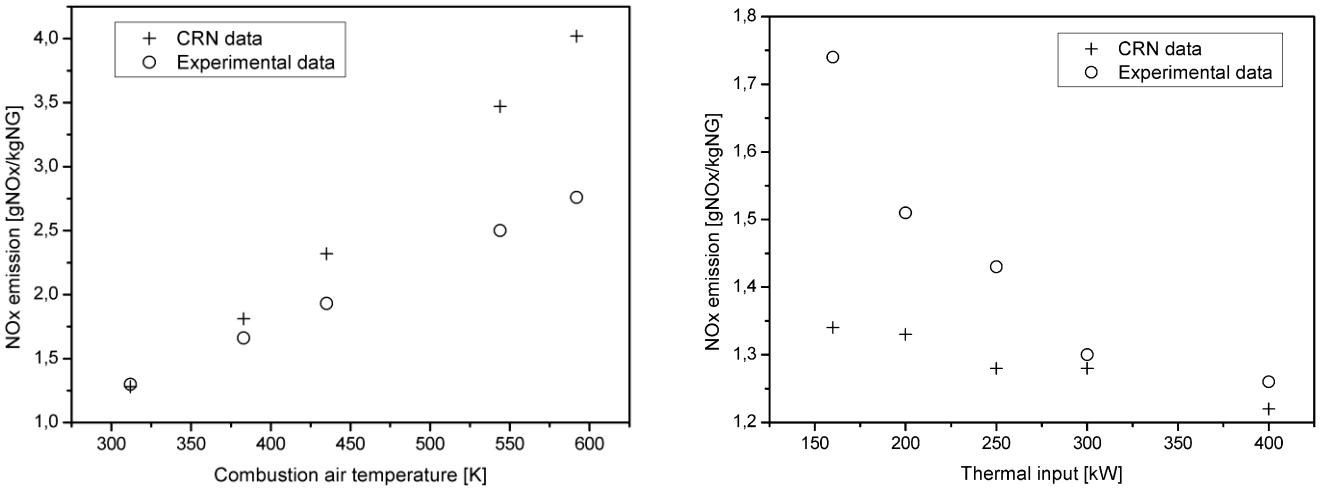

Figure 9. Effect of the air preheating (left) and turndown operation (right) on the NOx emissions of the CRN. Experimental data according to Hsieh et al., 1998.

The analysis of the Fig. 3 reveals the importance of the internal recirculation zone (IRZ) in the formation of NOx, since the total NOx emission decreases to 0,1 g NOx/kg GN when the temperature in the IRZ decreases from 1885 to 1683 K. Likewise, the NOx emission increases substantially with temperature, when the combustion in the IRZ approaches stoichiometric condition. The influence of the recycled gas fraction is minor. In the parametric study, Fig. 9, the CRN predictions are in good agreement until 110ºC preheating and for a turndown ratio of 1,33.

The results presented in Tab. 4 and Fig. 8 and 9 reveals the capabilities of the CRN tool, which is able to yield results in good agreement with the experimental data using a compact, 5-element reactor network model. Additionally, a calibrated CRN is a useful platform to evaluate burning systems modifications, as fuel substitution and fuel staging.

6. CONCLUSIONS

In the last decade several works were developed to simulate gas turbine combustors using CRN. The application of this tool to different combustion problems, like pulverized coal boilers, became more usual thanks to development of CFD-to-CRN post processing algorithms. This work presented a two premise methodology for the development of CRNs to reproduce the reactive flow in the BERL 300 kW test. Starting with the simplest CRN model (PSR+PFR), three CRNs were developed, employing the concepts of Pedersen et al. (1997) and Broadwell and Lutz (1998), to obtain a five-element CRN able to reproduce the overall flow structure, recirculation and flame zone temperatures and NOx emission, the latter with relative error within 2%. A sensitivity analysis and a parametric study of the five-element CRN were carried out to evaluate its sensitivity to the flow splits adjustments and its ability to predict emission with air preheat and turndown operation. The NOx emissions predicted by CRN are in good agreement until 110ºC preheating and for a turndown ratio of 1,33. These results reveal the capabilities of a compact, five-element reactor network and shows how a calibrated CRN could be a useful platform to evaluate burning systems modifications.

7. ACKNOWLEDGEMENTS

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