

EXPERIMENTAL AND COMPUTATIONAL INVESTIGATION OF COMBUSTION
PHENOMENA IN MESOSCALE DUCTS

BY

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DISSERTATION

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Abstract

Experimental and computational investigations of the flame phenomenology in mesoscale tubes were performed. Particular emphasis was given to oscillating flames, a phenomenon in which a flame front undergoes a periodic cycle of upstream propagation, extinction, and downstream re-ignition. Fuel-rich methane-oxygen mixtures and straight quartz ducts with one end open to the atmosphere were used. The flame behavior was determined as a function of equivalence ratio and Reynolds number in order to produce maps of the phenomenology and identify the boundaries between behavioral regimes. Rich propane-oxygen mixtures and multiple tube lengths were also studied in order to examine the impact of fuel selection and geometry on the regime boundaries. Infrared thermometry was utilized in order to determine the wall temperature distributions for different flame phenomenologies and to test the hypothesis that the moving nature of an oscillating flame will more uniformly distribute enthalpy throughout the duct. Both quartz and steel tubes were studied during IR experiments in order to investigate the effects of material properties on the resulting temperature distribution and flame phenomenology. Analysis of the exhaust gases was performed in order to measure fuel leakage and incomplete combustion that may occur due to the gaps in combustion that exist in the oscillation cycle. Oscillating flames were found to have fuel leakage and pollutant emission no greater than conventional stationary flames, and to produce a nearly uniform temperature in the tube wall, aspects that make them highly suitable for small-scale power generation.

The results of the experiments were used in efforts to develop a high-level computational model that could predict the flame phenomenology. The experimental results strongly suggested a thermal driving mechanism underlying oscillations and revealed the interwoven nature of the wall temperature distribution and the flame phenomenology. The computational model

succeeded in qualitatively reproducing all observed flame phenomenologies. However, it was also determined that more complex mechanisms are needed in order to accurately replicate the experimentally observed boundaries between behavioral regimes.

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Chapter 1

Introduction

1.1 Motivation

Over the past few decades, miniaturization of devices has attracted growing research interest. Studies of small-scale combustion are a part of this trend, including fundamental examinations of miniature combustion and flame phenomena and applied efforts to develop usable systems on these scales. The high power density of hydrocarbon fuels as compared to alternatives such as electrochemical batteries makes them an attractive option to supply energy or thrust to microdevices and miniature vehicles [1]. Macroscale experience with combustion has shown the basic mechanism of combustion to be simple and reliable – useful traits for a small system.

However, development of miniature combustion systems has proven to be a formidable task [1]. Attempts to shrink existing systems have met with great difficulty. Miniaturized engines using conventional designs may not work at all, or at best have very low efficiency. Reducing the size increases the surface to volume ratio, exacerbating heat losses and changing the flow regime. Simply manufacturing the devices is troublesome, as the complex parts and mechanisms needed are very difficult to produce on small scales. Further complicating the issue, even our understanding of combustion itself has been challenged by observations of unusual phenomena and the defiance of concepts such as the quenching diameter.

Success in miniaturized power generation can only be attained through an understanding of the different rules and novel phenomena unique to these scales. New generation systems must

be developed with an eye toward simplicity and reliability. Development and improvement of miniature engines may be greatly aided by embracing the unusual characteristics of meso- and micro- scale combustion and harnessing the potential of the novel phenomena unique to these small scales. One such a phenomenon is the oscillating flame, in which a flame front propagates upstream through a mesoscale duct, extinguishes, and then re-ignites downstream in a periodic fashion. The moving nature of oscillating flames may more effectively distribute heat throughout a duct, which could be beneficial for techniques such as thermoelectric power generation. This thesis attempts to investigate the nature of oscillating flames and evaluate their potential for small-scale power generation.

1.2 Combustion in the Meso- and Micro-scale

The classical concept of the quenching distance or quenching diameter is regularly analyzed in combustion texts [2, 3], where it is generally described as the gap distance beyond which flame propagation is not possible due to excessive loss of heat and radicals to the walls. This quenching distance is estimated to be on the order of the flame thickness, and is the concept underlying the design of such devices as flash arrestors and flame holders. However, significant experimental and computational work [4-13] has demonstrated that flames can exist at scales below the quenching diameter. The true cause of quenching is not geometry, but rather thermal and radical losses [6, 14]. Thermal quenching can be mitigated by limiting the heat losses from the flame, through methods such as elevating the wall temperature [6], heat recirculation [15, 16], and external heating [13]. Radical quenching can be limited through methods such as catalytic combustion [11, 12]. Such methods allow combustion to be

established; however these small scales have effects on the fluid mechanics, heat transfer, and chemistry that present challenges to the design of miniature systems.

With regards to fluid mechanics, small-scale flows are almost invariably laminar [1]. The controlling parameter is the Reynolds number:

$$\text{Re} = \frac{\rho V D}{\mu} \quad (1.1)$$

Here Re is the Reynolds number, ρ is the fluid density, V is the fluid velocity, D is the duct diameter, and μ is the viscosity. In ducts, turbulent flow is observed for approximately $\text{Re} \geq 2300$. Turbulence has some advantages such as promoting mixing. This can be a useful consideration, especially given that fuels and oxidizers are generally stored separately for reasons of safety. However, decreasing diameters keep Re low, essentially limiting micro-systems to laminar flow unless unusually high pressures or densities are involved.

Heat losses in small systems are generally much more adverse than in conventional large-scale applications [1]. The reason for this is that heat generation in the flame is a function of volume, whereas heat losses to the environment are a function of surface area. Reducing the system size increases the surface area to volume ratio, magnifying the proportion of energy lost as heat. However, there are also thermal advantages to the reduction in scale. As a result of the smaller volume, hardware can be heated to its operating temperature more quickly, and potentially more uniformly. This can be beneficial to equipment such as fuel cells, which require certain operating temperatures (SOFCs work at 700°C), and thermoelectrics, which are often efficient only in narrow temperature ranges. Smaller sizes also make heat recirculation easier to implement. Heat recirculation can boost efficiency by reducing heat losses [17], but in large systems requires additional equipment and increases the system complexity. In small-scale systems, some upstream propagation of heat is essentially automatic, and can be promoted

through methods such as arrays of burners, or even an arrangement as simple as two parallel opposed flames [18]. Heat recirculation in the walls has been shown to promote stabilization of flames in small channels and improve resistance to extinction [19, 20]. The magnitude of heat losses to the walls affects the reaction zone thickness and the extinction limits [10], but upstream heat transfer can also promote stability and improve power density [9], and can even allow unusual phenomena such as superadiabatic combustion [7, 18, 21].

In conventional, large-scale systems, residence times tend to be much longer than reaction times, ensuring reactions run to completion. Large dimensions relative to the flame thickness also minimize the impact of hardware on the combustion process. In small scales, however, this is not necessarily the case. Small spaces limit residence times, and the reduction of turbulence hinders mixing, further slowing reactions and potentially reducing combustion efficiency [1]. Heat losses, noted above to be a major concern on these scales, also slow reactions by limiting the system temperature. Equipment on the same scale as the flame thickness produces the potential for geometry to affect reactions through mechanisms such as radical quenching. These issues can drastically lower combustion efficiency from the high levels expected in conventional applications, increasing waste and pollution. Even so, there are potential benefits for catalysis, as a small space makes it easier to bring more of the reacting fluid into contact with catalytic material.

1.3 Micropower Generation

Theoretical analysis has demonstrated that even with the increasing heat losses associated with reduced size, practical microcombustion heat engines can be developed with length scales on the order of 1mm, if not smaller [22]. Attempts to develop small-scale combustion systems

for power generation have followed two general approaches: the miniaturization of existing systems, and the development of novel designs.

Several projects have taken the approach of reducing existing systems to small scales [23-27]. This approach has advantages, as such devices have proven operation and significant background of research and optimization that have led to a good understanding of the devices and relatively high efficiencies, albeit at larger scales. However, they also share common challenges [1]. The manufacture of such equipment on small scales is very difficult, for several reasons. Machinery for conventional systems is often complex, requiring numerous moving parts of varying sizes. Producing the required parts on a small scale in general is difficult, and made more so by the tolerances needed for parts to interface. Even with satisfactory components, assembling these devices in the miniature is a challenge in itself. Problems continue after assembly. Current materials, already pushed to the limit in conventional large-scale systems, may be unable to withstand the thermal and material stresses of operation with reduced size and thickness. In addition, conventional designs are greatly hindered by the differences of the micro-scale discussed above: reductions in turbulence hinder mixing, and residence times that are sufficient on a large scale may no longer be adequate. As a result of these issues, such devices generally suffer from low efficiencies or an inability to produce positive power. Some promising projects in this category are as follows.

Efforts to develop a miniature gas turbine have been underway at MIT by Epstein and associates [23, 24]. The device has a volume of 300 mm^3 and operates at over 1 million rpm. While the project demonstrates the great potential of MEMS design and fabrication techniques, there has been great difficulty in producing positive power from the device. Fernandez-Pello and associates at UC Berkeley have worked on the development of miniature Wankel engines [25].

Two sizes are under development, one with a 10 mm rotor and 30W output, and a smaller micro-rotor with a 3 mm rotor and output in the mW range. Machining has proven to be a great challenge for these devices, in particular achieving proper seals to prevent fuel leakage, though the larger size has demonstrated positive power output. The efficiency is only ~0.2%, low because of a relatively low compression ratio and fuel leakage issues. Piston-engine systems have been pursued at Georgia Institute of Technology [26] and the University of Michigan [27]. The Georgia Tech model employs a piston oscillating in a magnetic field with 13.4 mm³ displacement. Positive power has been generated, though with low efficiency, again due to sealing issues. The Michigan project intends to use a swing engine with a rotating piston, in order to form multiple combustion chambers within the housing for a relatively simple design. However, it remains unproven.

Other researchers have pursued the alternate approach of developing novel designs, rather than attempting to shrink existing ones. This has advantages in that the unique characteristics of combustion on small scales can be embraced and exploited. However this approach involves developing new systems from scratch, without the benefits of knowledge, experience, and optimization that go along with the use of existing hardware. Rather than trying to develop an entire system at once, projects using this approach have tended to focus on the development of effective burners, which can then be coupled with heat engines or possibly with alternative methods of energy production that could be simpler or more efficient, such as fuel cells and thermoelectrics. Some promising efforts in this category are as follows.

At the University of Southern California, efforts by Ronney and associates to develop miniature Swiss-roll combustors have shown great potential [28-31]. Originally proposed by Lloyd and Weinberg [17], Swiss-roll designs help to recirculate heat from the flame and exhaust

to the unburned reactants, which can offer a substantial boost to efficiency [32]. This is especially beneficial on these small scales as it helps to counteract the heat losses inherent to the meso- and micro-scale. The burner has been demonstrated with an external combustion heat engine [29] and with fuel cells [30]. The thermal output of the burner can keep the fuel cell at its operating temperature, and versions of the combustor including a solid oxide fuel cell and burning propane-oxygen mixtures have demonstrated power densities of up to 420 mW/cm^2 [30]. The addition of catalytic material allows the burner to operate at much lower temperatures and Reynolds numbers [31]. The Swiss-roll combustor developed at USC is three dimensional, but a smaller, two-dimensional burner using the Swiss-roll concept with catalytic materials has also been developed at Princeton [33]. When coupled with thermoelectric modules, this burner has demonstrated positive power output, albeit with a low efficiency of $\sim 0.5\%$.

Other projects have focused on the use of catalytic methods to improve performance, with the downside of requiring scarce and expensive precious metals. Researchers at Yale developed a 16 cm^3 mesoscale catalytic reactor combining fuel electrosprays with stacked catalytic grids [34]. The combination of high dispersion and catalytic reaction allows even longer-chain hydrocarbons such as dodecane and jet fuel to be burned with combustion efficiencies of $\sim 97\%$. A larger (400 cm^3) model coupled with a Stirling engine exhibited an overall conversion efficiency of fuel chemical energy to electricity equal to 22%, which is the largest of the combustion-based micro-engines proposed so far [35].

The development of technologies capable of using more complex fuels is an important step in the path toward miniature power generation, as the fuels envisioned for practical devices are liquids such as methanol and ethanol, rather than simple gaseous fuels such as the hydrogen and methane most commonly used in research. Combustion-driven catalytic reformers that can

be coupled with fuel cells or other equipment are under development at MIT [36, 37] and Pacific Northwest National Laboratory [38]. Such systems are well suited to use with fuel cells, as the reformer can provide the fuel cell with both hydrogen gas and the heat needed to reach operating temperature.

It is still too early to tell what strategy will ultimately prove most effective in the development of small-scale energy systems. In the meantime, there is still great need for fundamental research to promote a better understanding of combustion in the meso- and micro-scales. Novel designs that seek to work within the restrictions imposed by small scales have shown the best results so far. Where possible, these designs may be improved through the exploitation of phenomena specific to these small scales. Oscillating flames may offer such an opportunity.

1.4 Oscillating Flames

As noted previously, an oscillating flame is a phenomenon in which a flame front undergoes a periodic cycle of upstream propagation, extinction, and downstream re-ignition. Figure 1.1 shows an example of an oscillating flame in a curved duct. Understanding of them is limited, as oscillating flames in small ducts are a recently observed phenomenon. To our knowledge, this is the first doctoral dissertation to be devoted to the topic. The first reports of oscillations were made in 2005 by Richecoeur and Kyritsis [39] and independently by Maruta et al. [40]. Data from both experimental studies were used for the theoretical/computational analysis of [41]. In both cases, the oscillations consisted of a flame front within a mesoscale quartz tube that would propagate upstream, extinguish, and re-ignite downstream in a periodic fashion. These oscillations demonstrated a new form of unsteady yet stable combustion that

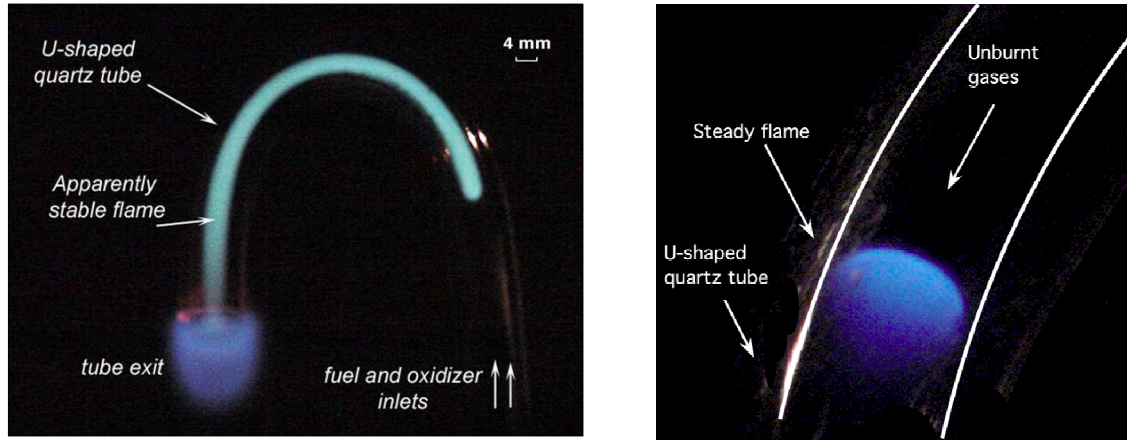


Figure 1.1. Sample images of an oscillating flame (left) and a stationary flame front (right) [39].

could be maintained indefinitely. Interestingly, the two groups produced this phenomenon through very different techniques. Maruta et al. [40] employed lean mixtures of methane-air and propane-air in a quartz duct that was given a temperature gradient by means of an external heat source. Oscillations were found to occur with both fuel types. Richecoeur and Kyritsis [39] made use of rich mixtures of methane-oxygen, and found that both a rich mixture and undiluted oxygen as the oxidizer were essential in order to establish oscillations. However, the oscillations produced by this method were self-sustaining, not requiring the external heating of [40]. Both curved and straight ducts were examined, and the duct geometry was found to influence the stability of the flames. Multiple modes of oscillation were observed, including varying propagation distances as well as stable flame fronts within the tube and at the open end. Thermoacoustic sound emissions were present for many oscillatory modes.

Since these initial observations, expanded investigations of both methods of producing oscillations have been conducted. Evans and Kyritsis [42] continued experimental studies of the self-sustaining oscillations from [39]. They verified the existence of self-sustaining oscillations for propane-oxygen flames and compared them to methane-oxygen flames by mapping out the flame behavior for a range of equivalence ratios and Reynolds numbers. Multiple tube lengths

and materials were also studied. Similar behavioral regimes were seen for both fuels, though the layout of the boundaries between them differed greatly. Oscillations were less stable and more difficult to produce in longer tubes, and it was observed that only ducts made of materials with low thermal conductivity, such as quartz, were capable of producing oscillations. The experimental and analytical results of this study supported a thermal driving mechanism. These experiments are described in detail in chapter 3.

Similar behavioral maps of flame behavior for lean methane-air flames have been produced by Fan et al. [43] in quartz channels with external heating – the same method of producing oscillations employed in [40]. With a constant fuel flow rate, increasing the air flow rate caused the flame to exhibit first blowout, then oscillations, then quenching within a narrow range of air flow rates, then stationary flame fronts, then blowout again. This pattern is the same as what was observed in [42] for rich flames, with the addition of a narrow quenching band between the oscillation and stationary flame front regimes. Multiple channel widths were studied, and the range of stable flame conditions shrank with smaller channel size. Only in the smallest channel was the narrow quenching band between the oscillation and stationary flame front regimes observed. Since reducing channel size increases the surface to volume ratio and with it the effective heat loss, these results also point to a thermal driving mechanism. Subsequent computational and experimental investigations of propane-air mixtures have produced splitting flames, in which two propagating flame fronts exist [44, 45]. Ignition occurs due to heating from the wall, after which the flame separates into two fronts. One flame front propagates upstream, while a weaker second flame propagates downstream until both extinguish and the process repeats.

Several computational investigations and simulations have also been performed [41, 46-50]. Jackson et al. [41] developed a theoretical model capable of capturing flame oscillations of the type seen experimentally using a solely thermal model. Other simulations [46, 47] with methane-air mixtures for stoichiometric and lean mixtures support the idea of a thermal driving mechanism, primarily an interaction with the heated duct walls. In particular, the results of [46, 51] emphasize the importance of the wall temperature gradient to the evolution of the flame behavior. A computational model by Pizza et al. [48, 49] has succeeded in producing oscillations in lean hydrogen-air flames in microchannels of various sizes. Further work with this model has indicated that the addition of a catalytic coating to the channel walls suppresses the activity of unsteady combustion modes such as oscillations [50].

As studies of mesoscale combustion have expanded, so have observations of oscillating flames and other phenomena that contain elements of oscillation. Kim et al. [52] observed oscillations in Swiss-roll burners. Continuously propagating flames have been observed in radial microchannels [53, 54]. Like oscillations, these flames are stable but not stationary. Oscillation-like flame cells of repetitive ignition and extinction events have also been observed in alumina channels [55]. These too produced thermoacoustic sound emissions.

The findings of the studies above suggest that oscillations are driven by thermal interactions with the duct walls. Oscillations themselves appear to be a series of repetitive flashbacks, in which a flame front propagates upstream until it reaches an area for which the wall temperature is too low to sustain a flame, heat losses become excessive, and the flame extinguishes. The mixture will re-ignite when it reaches a hotter portion of the tube that transfers enough heat to the reactants to initiate combustion. This process is quite different from what is expected in a conventional macroscale burner. In conventional burners the ducts are quite large

compared to the flame, and are generally made of materials with high thermal conductivity such as metals. In such a case, the duct wall acts as a heat sink. In mesoscale ducts, however, a low conductivity material such as quartz can more easily take and hold an elevated temperature, allowing it to act not just as a heat sink, but also as a heat source to reactant gases. A duct temperature high enough to re-ignite the mixture can be achieved through a diffusion flame burning excess fuel at the duct exit, as in [39]. Alternately, a temperature gradient can be imposed on the duct through external heating, as in [40]. External heating can allow oscillations to occur in near-stoichiometric mixtures [56].

1.5 Research Objectives and Structure of the Thesis

The work described above has made great progress toward understanding oscillating flames and mesoscale combustion, but has also raised many questions and left much unexplored. Gaps in the research and efforts that were undertaken to address them are as follows:

Initial investigations of oscillating flames [39] were limited to studies of rich methane-oxygen mixtures in quartz ducts. The early results of these studies revealed multiple flame behaviors, and suggested the importance of such factors as equivalence ratio, Reynolds number, and the system geometry in determining the flame behavior and the characteristics of oscillations. However, these factors were not studied in depth. Additionally, Maruta's work [40] demonstrated oscillatory behaviors in propane as well as methane for tubes with externally imposed thermal gradients, but it was not known if self-sustaining oscillations could be established for propane flames.

These questions were addressed by examining and mapping the flame phenomenology in mesoscale ducts. This involved experimental characterization of methane and propane flame

behavior over a range of equivalence ratios and Reynolds numbers. Multiple tube lengths were examined, and several tube materials were tested to determine whether oscillations are possible only in materials of low thermal conductivity. These efforts are discussed in chapter 3.

One of the most interesting aspects of oscillating flames is that they are stable, yet not stationary. A moving flame has the potential to effectively distribute heat throughout a system, which may be useful for some types of power generation such as thermoelectrics. However, previous studies have not examined whether this potential is actually realized within the system – that is, whether an oscillating flame does in fact heat the duct more than a stationary flame. Examination of the extent to which oscillations distribute heat through a tube is essential to evaluating their potential for small-scale power generation. In addition, it has been proposed that re-ignition of the flame front after extinction occurs due to heat input from a high-temperature region near the tube exit, however this has not been experimentally verified. These issues were addressed through the use of infrared thermography to analyze the temperature distribution of the tube wall. These efforts are described in chapter 4.

Self-sustaining oscillations have been produced only in rich mixtures. A consequence of this is that complete combustion cannot occur within the propagating flame front, leading to the production of many pollutant species. Generally, the excess fuel supports a diffusion flame at the tube exit that helps to complete the combustion process and eliminate these pollutants. However, high-speed videos have indicated that when the propagating flame front extinguishes, so does the diffusion flame at the exit [42]. Relighting occurs soon after, but it is possible that a gap in the combustion exists during which fuel or combustion intermediates may leak out. Alternately, the diffusion flame at the exit may not vanish completely, but may enter a non-luminous state instead. These issues were addressed by performing chemical analysis of the

combustion products using Gas Chromatography and Mass Spectrometry in order to quantify the percentage of fuel consumed and remaining. These efforts are described in chapter 5.

Experimental observations of the phenomenology points to the importance of developing theories of the driving mechanisms underlying the flame behaviors, and suggests the influence of factors not easily controlled experimentally, most notably the thermal properties of the duct. A computational model would aid understanding of the underlying physics and allow phenomenology maps to be produced for system configurations that cannot be produced in the lab (e.g. for tubes with arbitrary thermal conductivity). Previous computational work [41, 46-50] focused on low-level modeling that is very computationally intensive and not suited to perform the thousands of time-varying simulations necessary to generate behavioral maps. Here “low-level” modeling is used to mean computations that focus on detailed modeling of more fundamental aspects of the system, including such elements as detailed chemistry, solution of the full Navier-Stokes equations, etc. The use of low-level mechanisms, while essential to accurately model such aspects as the shape of a flame front, introduce a level of complexity that may be unnecessary to model higher level concepts such as the flame behavior and can obscure simpler mechanisms that would achieve the same results. These issues were addressed through the development of a simple, high-level computational model. Multiple iterations of the model were developed. The initial version employed fixed temperature distributions for the wall and reactants. The second version allowed for dynamic variation to the reactant temperatures and included the ability to model the effects of reactant preheating on the flame properties. The final version allowed dynamic variation of the wall temperature distribution and modeled the heat transfer within the wall and between the wall and environment. The development of the model is discussed in chapter 2, while the computational results are discussed in chapter 6.

In summary, the following objectives were pursued in order to address perceived gaps in the research.

- To determine whether oscillations are produced via solely thermal mechanisms, or if fluid mechanics is also a factor.
- To explore the range of oscillatory behaviors that can be produced, and examine the variations in flame phenomenology that occur with changes to key parameters.
- To examine the wall temperature distributions produced by oscillations and other flame behaviors, search for evidence of a high temperature region near the tube exit that provides heat for re-ignition, and examine whether an oscillating flame provides a more uniform distribution of enthalpy along the duct.
- To determine whether the extinction-ignition cycle of oscillations leads to gaps in the combustion, and if this leads to leakage of fuel or combustion intermediates.
- To develop a computational model to predict the flame phenomenology using high-level mechanisms.

Addressing these questions required a variety of experimental and computational techniques that are described in chapter 2. Investigations of the flame phenomenology and parametric studies of the effects of tube length and duct material are described in chapter 3. Infrared thermometry measurements of the wall temperature distributions are described in chapter 4. Analysis of the emissions and fuel consumption are discussed in chapter 5. The development of the computational model is described in chapter 2, and the results of simulations are discussed in chapter 6. Finally, a summary of the major findings and suggestions for the direction of future research are contained in chapter 7.

Chapter 2

Experimental and Computational Methodology

2.1 Flame Phenomenology Studies

The apparatus, depicted in Fig. 2.1, consisted of a straight quartz tube mounted vertically on a brass plate and copper block, with one end open to the atmosphere. Premixed fuel and oxygen flowed through a channel in the copper block and into the tube, where combustion occurred. Combustion products exited the tube through the open end. Three tube lengths were studied: 35 mm, 70

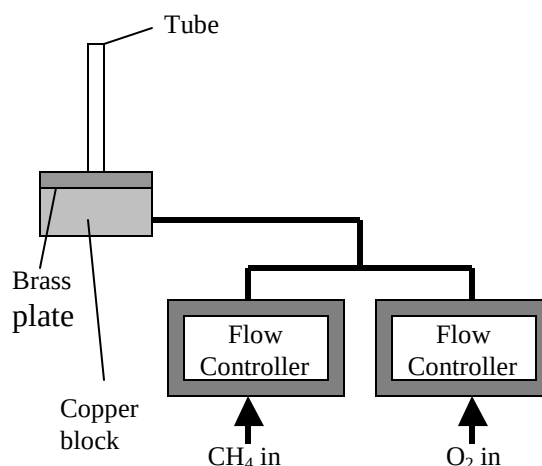


Figure 2.1. Apparatus for phenomenology studies.

mm, and 210 mm, each with an inner diameter of 4 mm and outer diameter of 6 mm, giving a wall thickness of 1 mm. Several additional tubes were made from different materials (glass, quartz, stainless steel, brass, and copper) in order to examine the effects of the duct material properties. For each material, the tube dimensions were the same: 70 mm length, 4 mm inner diameter, and 6 mm outer diameter. The duct was positioned vertically, with the top end open to the atmosphere and the bottom end attached to a brass plate (1 in. x 1.5 in., 0.25 in. thick). The tube-plate assembly was mounted on a copper block (1 in. x 1.25 in. x 1 in.) containing a 0.25 in NPT connection and internal ducts to deliver gas to the base of the tube. Two fuel mixtures were studied: methane-oxygen and propane-oxygen. For the methane-oxygen case, methane flow

control was established using an Omega FMA5510 mass flow controller and oxygen flow control with an Omega FVL-2617A flow controller. For the propane-oxygen case, propane flow control was established using an Omega FVL-2617A flow controller and oxygen flow control with a Cole-Parmer PMR1-010270 rotameter. The fuel and oxygen gas streams were mixed together in a T-intersection, then delivered to the copper block and tube.

The experimental procedure was as follows: a quartz tube of the desired length was installed in the apparatus, and a steady flow of fuel at the desired rate was established, initially with no oxygen input. The fuel was ignited at the open end of the tube so that a diffusion flame was initially formed. Oxygen was then slowly introduced into the flow until a change in the phenomenology was observed. With the flow rate of fuel held constant, the oxygen flow rate was incrementally increased and the flame phenomenology observed at each step until combustion terminated due to flashback and extinction. At each oxygen flow rate, the flame behavior was monitored and allowed to develop until the observed behavior remained consistent for at least 60 seconds, at which point the behavior was categorized and recorded. This process was repeated for both fuels with all three tube lengths. The effect of tube material on oscillations was explored qualitatively for five materials (glass, quartz, steel, brass, and copper) using a methane-oxygen mixture and similar technique.

2.2 Infrared Thermometry Studies

This study employed a setup similar to that used in the phenomenology studies. Ducts of two different materials, steel and quartz, were used for this experiment. In both cases the tube length was 70 mm, and the reactant mixture was composed of methane-oxygen. The only significant modification was the connection of four 0.01 in. diameter K-type thermocouples to

the tube at various locations along the length, in order to provide reference temperature measurements.

Infrared images were captured using an Electrophysics PV320-L2 camera positioned 20.5 in from the tube and equipped with a 50 mm germanium broadband lens (F1.0) transparent to wavelengths in the 2–14 μm range. The gas mixture consisted of premixed methane and oxygen, controlled in same manner as in the flame phenomenology studies.

The experiments were performed with two ducts made of different materials. In one case the duct was made of stainless steel, and in the other case the duct was made of quartz. For the steel duct, the four thermocouples were welded to the outer surface of the tube 2 mm, 17 mm, 34 mm, and 57 mm down from the duct exit. For the quartz tube, attachment of the thermocouples at the surface was not feasible; as an alternative, holes were drilled into the wall of the tube and the thermocouples were inserted into these. The holes were 0.5 mm deep (penetrating halfway through the wall thickness), and had diameters comparable to the thermocouple beads, allowing the thermocouples to fit snugly inside. For the quartz tube, the thermocouples were located 2 mm, 15 mm, 31 mm, and 55 mm down from the duct exit.

A major aim of this study was to compare the duct temperature profiles for different flame phenomenologies with the same chemical energy input. To accomplish this, the fuel flow rate was kept constant throughout an experiment, and the oxygen flow rate was varied in order to make mixtures with different equivalence ratios and thereby produce different flame phenomenologies. In each experiment, a fixed fuel flow rate was established, and the equivalence ratio was varied by adjusting the oxygen flow rate. For each mixture, the flow was established and the flame was ignited at the tube exit, then the system was allowed 5 minutes to achieve steady state. If steady state was not achieved within this time (as evidenced, for

example, by significant changes in the thermocouple readings over several seconds), additional 5 minute periods were allowed as necessary until steady state was achieved. In nearly all cases, steady state was achieved within the initial 5 minute period, and even in the cases slowest to reach steady state, it was achieved within 30 minutes. Once steady state was achieved, the flame phenomenology was noted, the temperatures of the thermocouples were recorded, and images of the infrared signal were captured.

As the camera was unable to capture the full temperature range observed over the experiment (from 25°C to 700°C and higher) with any single configuration, two images were taken for each case, each with different gain and level settings. One configuration captured the lower portion of the temperature range, while the other configuration captured the upper portion of the range, with some overlap. For quartz, the lower portion of the temperature range was captured with level 52 and gain 71, while the upper portion of the temperature range was captured with level 15 and gain 12. Typically, there is an exponential dependence of luminous gain to the gain setting of intensified cameras, but this was not confirmed by the manufacturer of the particular imager. For all quartz images, a sensitivity of 1 and gain parameter of 51 were used, and the iris was set in a fixed position at the beginning of the experiment such that the maximum temperature produced in the quartz duct (~780°C) produced an intensity below saturation with the “high temperature” settings (level 15, gain 12). For steel, the lower portion of the temperature range was captured with level 64 and gain 76, while the upper portion of the temperature range was captured with level 21 and gain 19. For all steel images, a sensitivity of 1 and gain parameter of 50 were used, and the iris was set in a fixed position at the beginning of the experiment such that the maximum temperature produced in the steel duct (~550°C) produced an intensity below saturation with the “high temperature” settings (level 21, gain 19).

Trials were conducted for several fuel flow rates (28.7, 46.6, 64.6, 75.3, 86.1, 96.9, and 107.6 mL/min), allowing a range of mixture flow speeds and Reynolds numbers to be studied. For each trial, the oxygen flow rate was first configured to produce a mixture equivalence ratio of 4.0. After taking data, the oxygen flow was increased in order to reduce the equivalence ratio to the next data point. Data were taken for equivalence ratios in increments of 0.5, to an equivalence ratio of 2.5. For the remaining data points, the equivalence ratio was then reduced in increments of 0.1, continuing until extinction occurred. The process and mixtures studied were identical for both the steel and quartz tubes, the only difference being that extinction sometimes occurred at slightly lower equivalence ratios for the quartz duct, producing a few additional data points.

2.3 Emissions Studies

This experiment employed a reactor setup similar to the ones used for the phenomenological observations and the infrared thermography measurements, with additional equipment for gas sampling and speciation. The apparatus is depicted in Fig 2.2. A 70mm long quartz duct was used in the burner, and the reactant mixture was composed of premixed methane-oxygen. Samples of the exhaust gases were taken with a 1mm ID, 2mm OD quartz probe positioned vertically over the axis of the duct. Measurements of the exhaust species were performed with an Agilent 6890N Gas Chromatograph (GC) and 5973N Mass Spectrometer (MS). A Robinair CoolTech 4 CFM two stage vacuum pump provided suction to the probe, allowing gas samples to be drawn through the probe and into the GC/MS. A cylindrical sleeve of pyrex glass with 75 mm diameter and 4.5 inch length was positioned around the flame front in

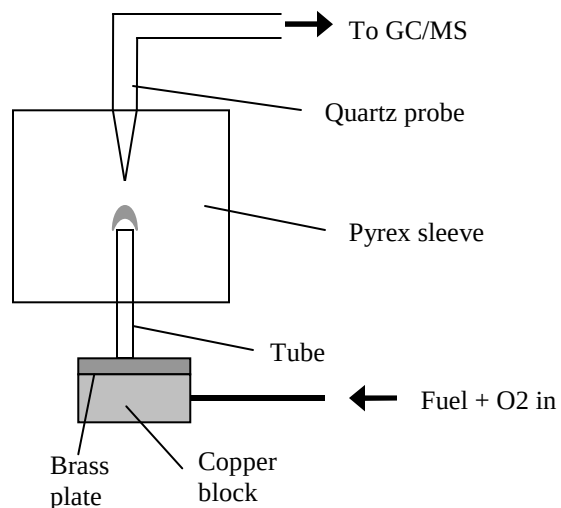


Figure 2.2. Apparatus for emissions studies.

order to shield the flame from air currents while still allowing air to reach the flame at the tube exit. The lower edge of this sleeve was positioned 30 mm below the exit of the tube.

The GC/MS was configured to use an Agilent HP-PLOT Moleseive column, allowing separation of methane, oxygen, and carbon monoxide. To ensure proper vacuum in the MS, a 0.1 mm ID, 0.25 mm OD fused silica restrictor column was connected in series with the Moleseive column. Sample gases flowed continuously from the probe and through a 1 mL sampling loop before being expelled through the vacuum pump. This sampling loop was maintained at 100°C to prevent condensation of water. At the start of each experiment, the contents of the sampling loop were redirected into the GC inlet via a pneumatic valve. Each analysis was performed with a constant gas flow of 1.5 mL/min through the column, and an oven temperature cycle as follows. The oven temperature was maintained at 60°C for 3 minutes, then increased at a rate of 20°C/min to 150°C. At the end of each analysis, the oven was heated to the column conditioning temperature of 300°C for 5 minutes in order to flush the column and remove potential contaminants. The total time per test was 16.5 minutes, not including cooldown time between runs.

The experimental procedure was similar to that followed in the IR thermometry experiments. Four fuel flow rates were studied: 46.6, 64.6, 75.3, and 96.9 mL/min. Multiple equivalence ratios were studied for each fuel flow rate, starting at 2.5 and decreasing in increments of 0.1 until the extinction limit was reached. The oxygen flow rate was adjusted in order to produce different equivalence ratios while allowing the fuel flow rate to remain constant. A major aim of this study was to compare the fuel leakage for different flame phenomenologies, so for each combination of fuel flow rate and equivalence ratio, samples were taken at the same probe position for both a burning and non-burning mixture, to allow the percentage consumption of fuel to be determined. The probe was positioned slightly downstream of the luminous region of the flame and kept in this position for both the reacting and non-reacting cases. For each experiment, the reactant flow rates were established, the probe was positioned, and the system was allowed 5 minutes to reach a steady or periodic state (an amount of time found to be sufficient in the IR thermometry studies). Once steady state was achieved, the vacuum pump was turned on and 5 minutes were allowed for the gases to reach the sampling loop. At that time, the GC/MS analysis sequence described above was activated and the sample was analyzed.

2.4 Computational Studies

2.4.1 Initial Version of the Model

A model was implemented that tracked the movement of the flame front in one dimension, along the axis of the tube. Nomenclature for this section is described in Appendix A. Fluid flow through the tube was modeled as plug flow. The model employed a marching scheme that used the flame front as the point of reference and followed it in each time step. Time was

discretized, and a constant time step size was used for each simulation. Heat transfer between the tube wall and flame front was assumed to occur with a constant Nusselt number of 4.36, which corresponds to laminar flow [57].

Simple one-step chemistry was employed within the model, with reactants changing immediately to products at the flame front. However, the adiabatic flame temperature and speed for each equivalence ratio were calculated for an equilibrium state in CHEMKIN using the GRI 3.0 mechanism, allowing us to more accurately model the thermal properties of the system and the movement of the flame front.

The model environment consists of three main entities: the tube, the inlet flow, and the flame front. The tube defines the system geometry, and has a specified length, inner diameter, and temperature distribution along its length. The inlet flow defines the unburned mixture properties, and has a specified equivalence ratio, flow speed, and gaseous properties as needed. The tube and inlet flow characteristics remain constant throughout the simulation. The flame front properties are determined dynamically at each step, and include the position, temperature, speed, and gaseous mixture properties. Heat and mass exchange with the surroundings was not modeled. Figure 2.3 provides an illustration of the computational space.

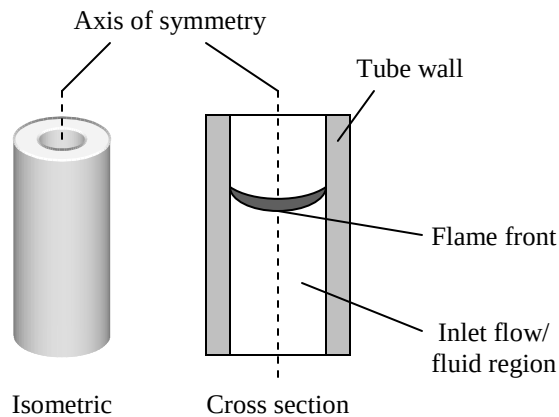


Figure 2.3. Geometry and components of the computational space.

2.4.1.1 Theoretical Description of the Model

Three phenomena are required for an oscillating flame: propagation, extinction, and re-ignition. These three components were modeled as follows.

Propagation. For the case of a flame front in a duct with constant reactant flow rates, the reactant mixture will flow downstream as a result of its momentum, while the reacting flame front will move upstream as it propagates into the unburned reactants. This creates a competition between the momentum of the flow, which will push the flame front downstream, and the speed of the deflagration, which will move the flame front upstream. In the code, the net velocity of flame propagation (with respect to the stationary duct) is calculated as the net difference between the flow speed and the flame speed. At each time step, the flame front moves a distance equal to the product of this net difference and the time step size.

$$\Delta z = (U_{in} - f)\Delta t \quad (2.1)$$

Extinction. Extinction of the flame front can occur via two mechanisms in our model. The first is thermal quenching due to volumetric heat loss. This is evaluated using a method described in the text by Law [58]:

The upstream heat loss in the preheat zone can be calculated as:

$$L^- \approx \frac{\rho C_p f}{k} q^- \quad (2.2)$$

Where the heat loss flux is given by:

$$q^- = h(T_u - T_{wall}) \quad (2.3)$$

Downstream heat loss in the cooling zone is similar:

$$L^+ = \frac{\rho C_p f}{k} q^+ \quad (2.4)$$

With:

$$q^+ = h(T_f - T_{wall}) \quad (2.5)$$

Heat loss from the reaction zone is neglected due to its small thickness. For modeling purposes, constant Nu_D heat transfer for laminar flow was assumed, based on the experimentally observed laminar nature of the flow. This produced the following relation for the heat transfer coefficient, with $Nu_D = 4.36$ [57]:

$$h = \frac{Nu_D k}{D} \quad (2.6)$$

The volumetric heat loss terms are non-dimensionalized using the following relation:

$$\tilde{L}^\pm = \frac{k}{\rho^2 C_p \varepsilon_{ad}^2 f_{ad}^2 q_c Y_u} L^\pm \quad (2.7)$$

Where ε_{ad} is calculated with the relations:

$$\varepsilon_{ad} = \frac{\tilde{T}_{ad}^2}{\tilde{T}_a} \quad (2.8)$$

$$T_a = \frac{E_a}{R_u} \quad (2.9)$$

Temperature is non-dimensionalized as follows:

$$\tilde{T} = \frac{(C_p T)}{(q_c Y_u)} \quad (2.10)$$

The non-dimensional volumetric heat loss terms are combined into an aggregate heat loss:

$$\tilde{L}_v = \tilde{L}^+ + \tilde{L}^- \quad (2.11)$$

This can be used to find the non-dimensional flame speed via the relation

$$-\tilde{L}_v = \tilde{f}^2 \ln(\tilde{f}^2) \quad (2.12)$$

For the flame to remain stable, $\tilde{L}_v$ must fall in the range $0 \leq \tilde{L}_v \leq e^{-1}$, limiting $\tilde{f}$ to the range $e^{-1/2} \leq \tilde{f} \leq 1$. Using the $\tilde{L}_v$ value calculated above, Eqn. 15 was solved for $\tilde{f}$ using the bisection method. If $\tilde{f}$ falls outside the stable range, extinction occurs. Otherwise, the non-adiabatic flame speed and temperature of the stable flame can then be found from the non-dimensional flame speed $\tilde{f}$ as follows:

$$\tilde{f} = \frac{f}{f_{ad}} \quad (2.13)$$

$$\tilde{T}_f = \tilde{T}_{ad} - \varepsilon_{ad} \frac{\tilde{L}_v}{\tilde{f}^2} \quad (2.14)$$

In this manner, the mechanism is used not only to evaluate stability, but also to determine the non-adiabatic temperature and speed of stable flame fronts at each time step.

Extinction of the flame front will also occur if the flame reaches the base of the tube. This is based on observations from the physical system, which beyond the base of the tube becomes a much narrower duct through a copper block. This acts as a heat sink and remains much cooler than the tube, causing the flame front to quench upon reaching the base for essentially all stable configurations.

Re-ignition. The flame front exists at the interface between the leading (downstream) edge of the reactant mixture and the trailing (upstream) edge of the product mixture. After the flame front is extinguished, this interface continues to move through the tube at the flow velocity. As warmer areas of the tube are encountered and the reactant gases heat up, the first and most likely place at which re-ignition can occur is this interface, at the leading edge of the reactant mixture. This portion of the reactant flow is the first to encounter warmer regions of the

tube as it flows downstream and may also carry some residual thermal energy from the now-extinguished flame front. It has the same location as the former flame front, so when extinction occurs this location continues to be tracked by the code as it propagates downstream at the inlet flow speed. Upon extinction of the flame front, the temperature of the leading edge of the inlet flow is set to the local wall temperature as an initialization. As it moves toward the exit, the change in temperature of this location as a result of thermal interaction with the duct walls is evaluated using the reactant specific heats and the heat transfer from the walls with the following equation:

$$\Delta T_f = \frac{\dot{Q}_{wall} \Delta t}{V_{edge} \left((y\rho C_p)_{methane} + (y\rho C_p)_{oxygen} \right)} \quad (2.15)$$

Where the heat transfer between the leading edge and the wall is

$$\dot{Q}_{wall} = hA_{edge} (T_f - T_{wall}) \quad (2.16)$$

The area and volume of the leading edge are defined as

$$V_{edge} = \frac{\pi}{4} D_{tube}^2 \Delta z \quad (2.17)$$

$$A_{edge} = \pi D_{tube} \Delta z \quad (2.18)$$

Combining produces:

$$\Delta T_f = \frac{4h(T_f - T_{wall})\Delta t}{D_{tube} \left((y\rho C_p)_{methane} + (y\rho C_p)_{oxygen} \right)} \quad (2.19)$$

This shows that the temperature change is independent of the leading edge thickness. If the temperature of this leading edge exceeds the ignition temperature of the mixture, the extinction mechanism is run to determine whether a stable flame can be established or if the flame front will fail to ignite due to excessive heat loss.

Domain limits. The computational domain is limited to the tube wall and the fluid region it contains. The movement of the flame front is restricted so that it remains within the area of the tube. If the flame front passes beyond the tube base, this consists of extinguishing the flame and setting its position to the tube base. If it moves beyond the tube exit, the flame is anchored at the tube exit. If the flame is extinguished and the leading edge of the inlet flow passes beyond the tube exit, the simulation can be ended, as re-ignition will not occur.

2.4.1.2 Characterization

Characterization of the behavior of the flame front is also performed within the code. At the end of each time step it is noted whether an ignition or extinction event has occurred as well as the position of the occurrence. At the end of the simulation, the event history and current state of the flame are examined, and the flame behavior is classified as follows:

Extinction: the flame front has extinguished, and the leading edge of the inlet mixture has propagated to the exit of the tube or beyond without re-igniting.

Blowout: the flame front is currently active, and seated at the tube exit, and did not move from this position during the last time step.

Stationary flame front: the flame front is currently active and positioned within the tube, and did not move from this position during the last time step.

Oscillation covering the full tube length: the flame front has consistently been igniting at the tube exit and extinguishing at the tube inlet. It is not necessary for it to have done this throughout the entire simulation, but the pattern must exist by the end.

Oscillation covering part of the tube length: the flame front has been igniting at a consistent position and extinguishing at a consistent position. The ignition must not be at the

tube exit and/or the extinction must not be at the tube inlet. As before, this pattern does not need to exist throughout the simulation, but must be present by the end.

These regimes are shown schematically in section 3.1, where the phenomenology is discussed in detail. In the case of the stationary flame front and both oscillatory classifications, which require the flame front to occupy identical positions at successive times, slight variation in the exact location is permitted, as discretization and other issues make it unlikely that the flame front will indeed occupy exactly the same position numerically even though a macroscopic perspective would consider the locations to be essentially the same. Successive ignition/quenching locations were considered to be the same location if they differed by less than 1mm. Flame fronts were considered to be stationary if they had an effective velocity of less than 0.1mm/s.

2.4.1.3 Program Execution

The flame front is initialized to a specified location within the tube, in either a burning or extinguished state. If burning, initial non-adiabatic flame temperature and speed are determined using the mechanism from [58]. If extinguished, the position is noted as the leading edge of the flame front.

The main program loop is as follows. First, time is advanced by one step. The flame front, or the leading edge of the inlet flow if the flame is extinguished, is moved to a new position according to Eqn. 2.1. The new position is then verified as being within the tube boundaries, or else corrective action is taken as described previously. Next, if a flame front is present, the extinction mechanism from [58] is solved to determine whether the flame extinguishes given the wall temperature at the new location, and to solve for the new temperature

and speed if it remains stable. Alternately, if there is no flame front present, the heat transfer between the leading edge of the reactant flow and the wall is determined, and the new temperature of the leading edge is calculated using Eqns. 2.16-2.19. If this meets or exceeds the ignition temperature, the extinction mechanism is solved to determine if a stable flame front can be established, and the properties if a stable flame is possible. If a stable flame can exist, the flame front re-ignites; otherwise it remains unlit. The time step is then complete, and the loop begins again.

If enough time steps are processed, the system will eventually reach either a steady state or a stable periodic cycle. When the simulation is completed, characterization is performed as outlined above.

2.4.1.4 Procedure for Studies with Initial Version

Plots of the phenomenology were produced by performing multiple simulations for equivalence ratios from 1-3.5 and Reynolds numbers from 0-250. 1024 evenly spaced combinations of equivalence ratio and Reynolds number were selected from these ranges to

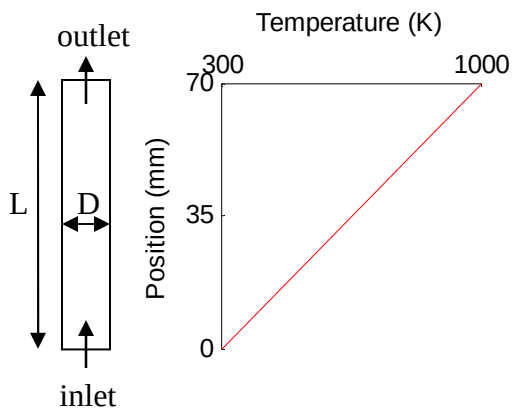


Figure 2.4. Tube geometry and wall temperature distribution for computational studies.

generate a map of the flame behavior for each configuration. All simulations were run for 5000 time steps with a step size of 0.001 seconds. The tube was given a length of 70mm and an inner diameter of 4mm, as in our physical setup. We specified a wall temperature profile for the tube that decreased linearly with axial position from 1000 K at the

tip (exit) to 300 K at the base (inlet). This profile was chosen in order to have a tip that was hot enough to ignite the mixture, making oscillations possible, and to have a general shape that approximates the expected profile of the physical system. The temperature profile of the tube remained constant throughout each simulation. Figure 2.4 illustrates this geometry and temperature profile.

2.4.2 Version 2 of the Model: Inclusion of Reactant Preheating

A major limitation of the first version of the model was that the adiabatic flame temperature and speed were calculated as a function of equivalence ratio only. In the real world, the temperature of the unburned gases also has an effect on these properties. This may be important in the actual system: the reactants enter the tube at ambient temperature, but encounter warmer tube walls as they flow through the tube. Heat transfer between the incoming gases and the tube wall causes the reactants to be preheated before entering the flame, potentially boosting the flame temperature and speed. The second version of the model sought to account for this phenomenon.

Several additions to the model were necessary to account for the effects of reactant preheating on the flame properties, including finite element meshes of the wall and fluid region, routines to calculate the temperatures in the fluid region at each time step, and expanded data for the flame temperature and speed taken as a function of both the equivalence ratio and the unburned gas temperature.

2.4.2.1 Changes to the Model

2.4.2.1.1 Finite Element Meshes

Finite element meshes are necessary in order to track the temperature distribution in the fluid region. Figure 2.5 illustrates the division of the computational domain into the finite element arrays. Two finite element meshes are created: one for the fluid region, and one for the tube wall surrounding it. Each of the two FE meshes was designed to be individually one-dimensional: divisions are made between elements in the axial direction (the z -dimension, along the tube axis), but not in the radial direction. The FE array of the fluid region occupies the central space along the tube axis, and the FE array of the tube wall surrounds it in the radial direction. The number of elements in the fluid region is given as an input to the program, and during initialization the fluid region is divided into the requested number of equally sized elements. The tube wall is divided into the same number of elements, ensuring that the cells line up for heat transfer calculations.

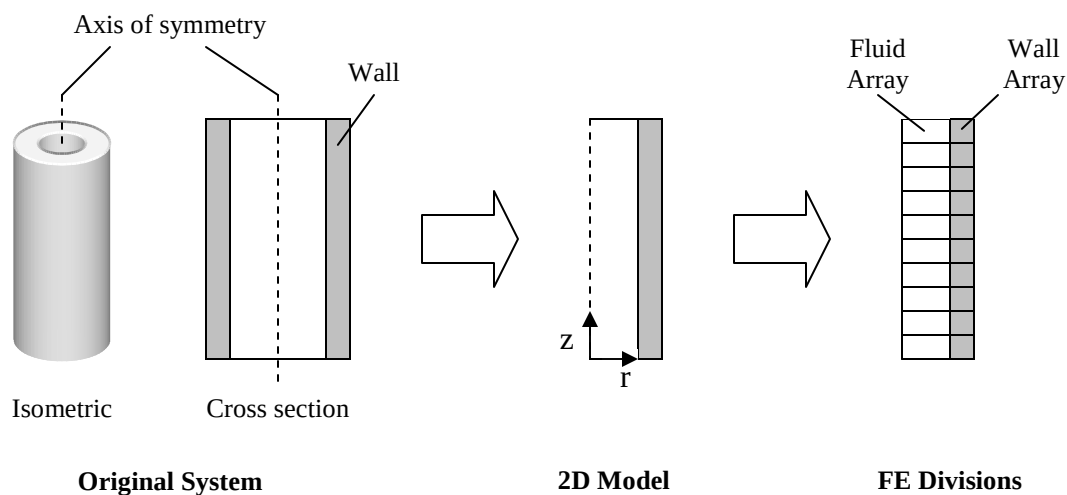


Figure 2.5. Formation of the computational domain.

In addition to position and geometry data, each element in the FE arrays tracks temperature values for three locations in the element: the top surface, the bottom surface, and the interior. The bottom (upstream) surface is the element inlet, and the temperature at this surface corresponds to the temperature of the fluid as it enters the element. The top (downstream) surface is the element outlet, and the temperature at this surface corresponds to the temperature of the fluid as it leaves the element. The interior of the element is the region between these two surfaces, and is modeled as having a uniform temperature throughout. This interior temperature is equal to the average of the temperatures at the top and bottom surfaces.

2.4.2.1.2 Heat Transfer and Element Temperature Calculations

The temperature distribution of the tube wall is specified at the beginning of the simulation and remains fixed throughout. However, the temperatures of the fluid elements can change due to heat transfer with the tube wall and interaction with the flame front. At each time step, the temperatures of the fluid elements are solved as follows. Figure 2.6 provides an illustration of the standard solution procedure for each element.

The program begins solving temperatures as the bottom of the tube and works its way up through the elements of the array. Solving starts at the bottommost element, located at the tube inlet. The bottom surface of this element is assigned a temperature equal to the specified temperature of the inlet flow. The time needed for the fluid to traverse the element is calculated from the element height and flow speed. Using this time value, the temperature change of the fluid after traversal is calculated using Eqn 2.19, and the new temperature is assigned to the top surface of the element. The internal temperature of the element is then calculated as the average of the top and bottom surface temperatures. The top surface temperature is then used as the

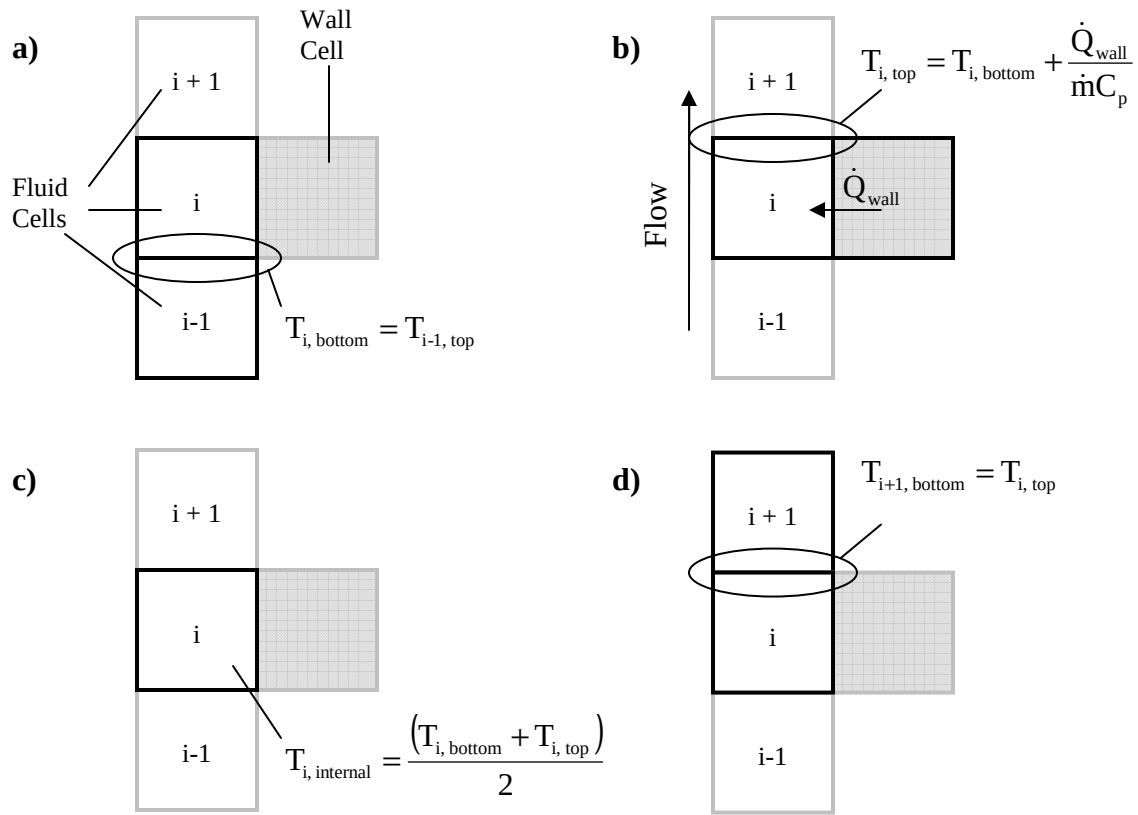


Figure 2.6. Standard method used to solve the temperatures of each fluid element. Simplified equations are used for illustration.

bottom surface temperature for the element above, and solution of that element's temperatures is handled in the same way. In this manner, the program "climbs" from the bottom to the top of the FE array, solving the element temperatures as it goes. A slight variation on this procedure occurs for an element that contains a flame front. If a flame front is present in the element, then the top surface temperature is set to the temperature of the flame front, and Eqn 2.19 is not used.

2.4.2.1.3 Determination of Adiabatic Flame Properties

The inclusion of reactant preheating allows the adiabatic flame temperature and speed to be calculated as a function of both the equivalence ratio and the unburned gas temperature.

These properties were calculated in CHEMKIN using the GRI3.0 mechanism for a range of equivalence ratios and unburned gas temperatures in order to create lookup tables.

Although the equivalence ratio remains constant throughout the simulation, the temperature of the unburned gases entering the flame front can vary from moment to moment. Because of this, new adiabatic flame properties were determined at each time step as follows. First, the position of the flame front is used to determine what fluid element it currently occupies. The temperature of the bottom (upstream) surface of that element is used as the unburned gas temperature. If the flame has blown out of the tube, it will be seated at the tube exit and not within any fluid element; in this case, the temperature of the top surface of the topmost fluid element is used as the unburned gas temperature. With the equivalence ratio and unburned gas temperatures known, the adiabatic flame temperature and speed are determined from the lookup tables via bilinear interpolation of the surrounding points.

2.4.2.2 Procedure

Phenomenology maps were determined using the same procedure employed for the previous version of the code, described in section 2.4.1.4. For the finite element meshes, 70 fluid and 70 wall elements were used, giving a 1 mm length per element.

2.4.3 Version 3 of the Model: Dynamic Calculation of Wall Temperature

2.4.3.1 Changes to the Model

The flame behavior and the temperature distribution of the tube wall are strongly linked, a phenomenon discussed in depth in chapter 4. These factors are interwoven: the flame heats the wall to produce local temperature elevations, and the local wall temperature affects the heat loss

from the flame and provides heat recirculation to the reactants, thereby influencing the flame speed, temperature, and stability. To improve the model's ability to accurately capture the flame behavior, it is necessary to model this flame-wall thermal interaction and dynamically calculate the temperature distribution in the tube wall.

The finite element mesh necessary to model the temperature distribution in the wall was implemented in the previous version of the model, described in section 2.4.2. Modeling the changes in the wall temperature distribution required calculating the heat transfer and enthalpy change experienced by each element at each time step. The modes of heat transfer experienced by a typical wall element are illustrated in Fig. 2.7, and were calculated as follows:

Conduction. Conduction occurs between adjacent solid cells. A typical element in the solid array will experience conduction with one element above and one element below. As noted above, the wall array is one-dimensional, and has divisions only along the tube axis. Because of this, conductive heat transfer occurs only in the axial direction, and not in the radial direction. The conductive heat transfer with each adjacent element is calculated using Fourier's Law:

$$Q_{conduction} = kA_{contact} \frac{\Delta T}{\Delta x} \quad (2.20)$$

Here the thermal conductivity k has a value based on the tube material, and $A_{contact}$ is the contact area between adjacent solid elements, equal to the cross sectional area of the tube wall. ΔT is the temperature difference between the two elements, and Δx is the distance between their center points (also equal to the length of one element).

Convection: inner surface. The inside surface of the tube wall is subject to forced convection with the fluid flowing through the duct. The convective heat transfer is then:

$$Q_{convection} = hA_{surface} \Delta T \quad (2.21)$$

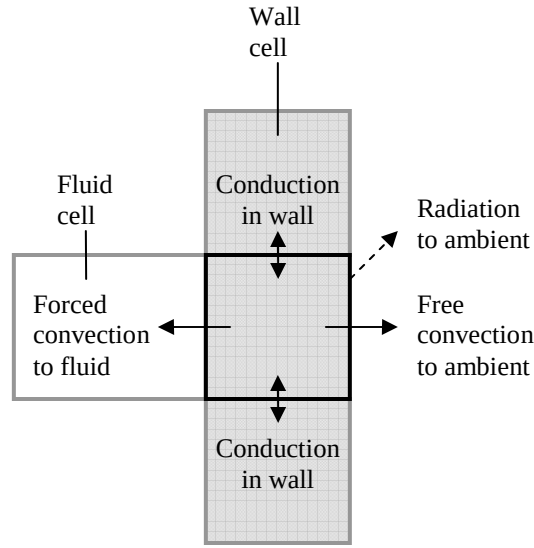


Figure 2.7. Heat transfer modes experienced by a typical solid element.

As noted above, this heat transfer is modeled as occurring with the constant Nusselt number for laminar flow through a tube ($Nu = 4.36$) [57], allowing h to be calculated using Eqn. 2.6. The area $A_{surface}$ is the area of the inside surface of the solid element, and ΔT is the temperature difference between the wall and fluid at this axial position.

Convection: outer surface. The outer surface of the wall experiences free convection with the ambient air. Equation 2.21 is also used for this calculation, but with slightly different values: h is the free convection heat transfer coefficient, $A_{surface}$ is the area of the outside surface of the solid element, and ΔT is the temperature difference between the wall element and the ambient air. For our simulations, a general value of $10 \text{ W/m}^2\text{-K}$ was used as the heat transfer coefficient for free convection.

Radiation. The outer surface of the tube wall experiences radiative heat losses to the environment, calculated using the following equation:

$$Q_{radiation} = \epsilon \sigma A_{surface} (T_{wall}^4 - T_{surroundings}^4) \quad (2.22)$$

Here ε is the material emissivity, σ is the Stefan-Boltzmann constant, $A_{surface}$ is the area of the outside surface of the solid element, T_{wall} is the temperature of the wall element, and $T_{surroundings}$ is the temperature of the surroundings.

The magnitude and direction of each component of heat transfer are computed separately. Once all heat transfer components have been calculated, the components are aggregated and the temperature change of the element is determined with the following equation:

$$\Delta T = \frac{t \Sigma Q}{\rho V C_p} \quad (2.23)$$

Here ΔT is the temperature change of the element, t is the time step size, ΣQ is the sum of the heat transfer components, ρ is the material density, V is the element volume, and C_p is the material specific heat. Where the duct material properties were needed in the above equations, the properties of quartz were used by default: thermal conductivity 1.38 W/m-K, density 2220 kg/m³, specific heat 745 J/kg-K, and emissivity 0.75.

There are two special cases for which the heat transfer calculations are slightly different: the bottom surface of the bottommost element, and the top surface of the topmost element. In the actual system, the bottom surface of the tube is in contact with a brass plate and large copper block that may be assumed to generally have the same temperature as the surrounding air. To approximate this, the heat transfer at the bottom surface of the bottommost element is modeled as conduction with an element with a fixed temperature equal to the ambient, using Eqn. 2.20. The topmost element is more complicated. It will experience heat transfer with the diffusion flame if one is present, or the ambient otherwise. Heat transfer from the diffusion flame to the tip of the tube is likely to involve substantial transfer by radiation, however this is not a simple thing to calculate; modeling the radiative heat transfer from a flame could itself constitute a doctoral thesis. Instead, a simple, flexible method of modeling the heat transfer from the diffusion flame

to the tube was devised. This heat transfer was modeled as a combination of free convection and radiation. The convection and radiation components were calculated using equations with the form of Eqns. 2.21 and 2.22, with user-defined values for the convection coefficient h and the emissivity ε . A convection coefficient of 10 W/m^2 and an emissivity of 0.4 were found to give satisfactory temperature elevations to the tip and were used as the defaults for simulation. In the situation when no diffusion flame is present, heat transfer to the ambient at the top surface is modeled as occurring via free convection and radiation in the same manner as for the outer surface of the tube, using Eqns 2.21 and 2.22. The temperature of the diffusion flame was calculated in CHEMKIN using the GRI 3.0 mechanism; temperatures were calculated for an equilibrium state, with an initial mixture consisting of the rich inlet methane-oxygen mixture combined with enough air to bring the equivalence ratio to 1.

2.4.3.2 Procedure

The method of computing phenomenology maps for the final version of the code was similar to that used for previous versions, described in section 2.4.1.4. However, some modifications to this procedure were necessary in order to deal with the greater complexity and computational intensity of dynamically calculating the wall temperature distributions.

The first change was an increase to the simulation time. To allow the system to evolve from the starting conditions to a final steady or periodic state, simulations were run for 5 virtual minutes, rather than the 5 virtual seconds used in previous versions. The 5 minute figure was derived from the IR thermometry tests, in which this was found to be a sufficient time to reach a final state. The time step size kept a constant value of 0.001 s, as in previous versions.

The second change was a decrease in the resolution of the phenomenology maps. Previous versions used 32 divisions of equivalence ratio and 32 divisions of Reynolds number, for a total of 1024 computational cases per map. The greater computational intensity and time requirements of the final version necessitated a reduction in the total number of cases per map. 16 divisions of equivalence ratio and 8 divisions of Reynolds number were used, for a total of 128 cases per map. More distinct equivalence ratios were tested on the basis of experimental data suggesting that this is the primary factor that determines the phenomenology, discussed in chapter 3.

It was quickly observed that the initial wall conditions affected the final solutions for this version of the model, so for each computational case, two different initial wall temperature distributions were used. In one case, the wall was initially given the linear temperature distribution of Fig. 2.4, with the temperature decreasing linearly from 1000 K at the tip to 300 K at the base. The purpose of this distribution was to simulate a preheated tube. In the other case, the initial wall temperature was set to the ambient temperature of 300 K throughout, in order to simulate a cold tube.

Chapter 3

Flame Phenomenology Studies

3.1 Flame Stabilization Phenomenology

For the case of a premixed laminar flame with constant equivalence ratio in a non-adiabatic, large-scale duct, three modes of behavior are commonly observed. (By “large” here, we mean of large diameter in comparison to the flame thickness.) The exhibited behavior is determined by the relative values of the flow velocity and the laminar flame speed. At a critical flow speed that equals the flame speed, a stable, stationary flame front will exist within the duct. If the flow speed is below this critical value, the flame speed exceeds the flow speed and the flame front propagates upstream in a flashback; if the flow speed is above the critical value, it exceeds the flame speed and the flame blows out and takes on a conical shape at the duct exit.

In “mesoscale” ducts, i.e. ducts with diameter on the order of the flame thickness, an additional oscillatory mode is observed in the results that are reported here. These oscillating flames were first observed in our group [39], and have previously been subjected only to limited study. To our knowledge, no other theses to date have been devoted to this phenomenon. During oscillations, a flame front propagates upstream, while at the same time the excess fuel maintains a diffusion flame at the tube exit. After propagating some distance, the moving flame front extinguishes. High-speed visualization has revealed that when this happens, the diffusion flame at the exit extinguishes at roughly the same time, for at least some cases. Shortly afterward, the flame reignites at the tube exit and the process repeats. Since these oscillatory phenomena appear to be repetitive flashback, extinction, and re-ignition events, it would be expected that oscillations can occur only when the flow speed is less than the flame speed.

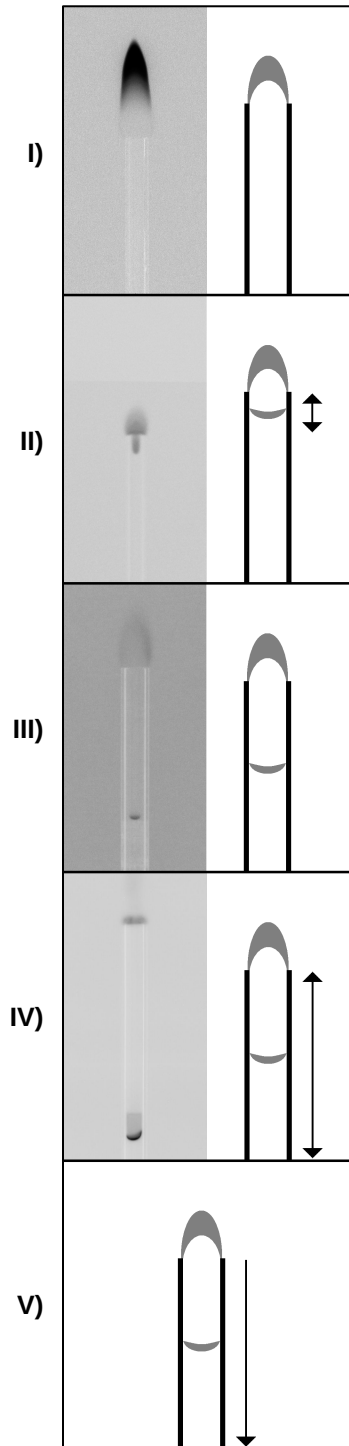


Figure 3.1. Inverted color photographs and sketches of characteristic flame behaviors. I) blowout; II) oscillations covering part of the tube length; III) stationary flame front in the tube; IV) oscillations covering the full length of the tube; V) flashback and extinction (sketch only).

Figure 3.1 shows the characteristic flame behaviors of the mesoscale regime. There, five behavioral regimes are introduced and indicated with Roman numerals (I-V). They will be used extensively in the discussion of the results that will follow. The observed mesoscale flame phenomenologies are the same ones described in brief in section 2.4.1.2:

I) Blowout. In this case, the flame front remains seated at the tube exit. No propagation into the tube occurs.

II) Oscillations covering part of the tube length.

Oscillations occur in which a flame front propagates into the tube, but extinguishes prior to reaching the base of the tube. Re-ignition occurs at the tube exit, producing a stable periodic cycle.

III) Stationary flame front in the tube. A flame front propagates some distance into the tube, comes to rest, and then remains at that location.

IV) Oscillations covering the full length of the tube.

Oscillations occur in which a flame front propagates into the tube and travels all the way to the tube base before extinguishing. Re-ignition occurs at the tube exit, producing a stable periodic cycle. Quenching of the flame

at the base is nearly certain, because below the base of the tube the fluid passage changes to a narrow duct through the copper block. This generally extinguishes flame fronts that propagate to the base of the tube.

V) Flashback and extinction. A flame front propagates some distance into the tube, after which both the propagating flame front and the exit diffusion flame extinguish, and re-ignition does not occur.

In all oscillating cases, a diffusion flame is maintained at the tube exit as a result of the excess fuel. It is recognized that, in principle, similar oscillatory phenomena can be induced in larger scale ducts, e.g. with an appropriate manipulation of the wall boundary conditions. Indeed, the results of [41] that theoretically described such oscillations are expressed in terms of non-dimensional geometric parameters. However, what is unique about the mesoscale regime is that the necessary temperature gradients on the tube wall occur spontaneously without the need to be imposed externally as e.g. in [40].

3.2 Fuel Effects (Methane vs. Propane)

Methane and propane flames were found to differ in several key areas. Propane flames require much more oxygen on a molar (volume) basis per kmol of fuel than methane flames – 2.5 times as much for stoichiometric combustion. Methane is a lighter fuel, with lower density and higher diffusivity than propane. Methane is lighter than oxygen, while propane is heavier. As the flames studied here are fuel-rich and oxygen-limited, this means that for the methane case the limiting reactant is the heavier one, while for propane the limiting reactant is the lighter one. This affects the Lewis number for the mixture, defined by Eqn 3.1.

$$Le = \frac{D}{\alpha} = \frac{D\rho C_p}{k} \quad (3.1)$$

Here Le is the Lewis number, D is the diffusion coefficient, α is the thermal diffusivity, ρ is the density, C_p is the specific heat, and k is the thermal conductivity (all properties are for the aggregate mixture). A comparison of the Lewis numbers for each fuel at various equivalence ratios is presented in Table 3.1. Diffusion coefficients were calculated using the method described in [59], with the following equation:

$$D = \frac{0.00266T^{3/2}}{PM_{AB}^{1/2}\sigma_{AB}^2\Omega_D} \quad (3.2)$$

Mixture-averaged thermal conductivities were calculated using a relation from [60]:

$$k = \frac{1}{2} \left(\sum_{n=1}^N X_n k_n + \frac{1}{\sum_{n=1}^N \frac{X_n}{k_n}} \right) \quad (3.3)$$

These relations are described in detail in their relevant texts. As shown in Table 3.1, Le is roughly 1 for the methane-oxygen case, but more than twice as high for propane-oxygen.

Increasing the equivalence ratio also increases Le for both fuels, though the effect is marginal in the methane-oxygen case and much more pronounced for propane-oxygen.

Table 3.1. Lewis numbers for methane-oxygen and propane-oxygen mixtures at various equivalence ratios. Values are calculated for a “cold” mixture at 300 K and atmospheric pressure.

Equivalence Ratio	<i>Le</i> Methane-oxygen	<i>Le</i> Propane-oxygen
1	1.02	2.41
1.5	1.03	2.57
2	1.03	2.72
2.5	1.03	2.84
3	1.04	2.96
3.5	1.04	3.06

Figures 3.2 a-f depict the relationship between equivalence ratio, Reynolds number, and flame behavior for methane and propane flames. The Reynolds number was calculated using the tube diameter and cold flow properties and used as a means of non-dimensionalizing the flow velocity and to facilitate comparison of the different fuels, without assuming that it is the controlling parameter of the flow. The flow controllers used in these experiments have an error of $\pm 1.5\%$, which produces some uncertainty in the measured equivalence ratios. This uncertainty ranges from $+0.066/-0.064$ for the case with the smallest flow rates to $+0.059/-0.058$ for the case with the largest flow rates.

A general trend can be seen in the behavior of both methane and propane flames. For a fixed Re , a sufficiently rich mixture will produce blowout and generate a diffusion flame at the tube exit. As the mixture is made gradually leaner, oscillations and stationary flames will be observed within the tube, with propagation depth increasing as the equivalence ratio decreases. When the mixture becomes close enough to stoichiometric, flashback and extinction will occur. This oscillatory pattern makes sense from a thermal perspective. As the flame penetrates into the duct, it loses heat to the non-adiabatic wall and ultimately extinguishes. At the same time the diffusion flame at the tube exit keeps the tip of the tube hot enough to re-ignite the mixture after the moving flame front is extinguished. It has to be stressed that the diffusion flame at the tube exit extinguishes simultaneously with the propagating flame front, so the heat for re-ignition must be supplied by the tube wall. This assertion can be supported by careful measurements of wall temperature (based on IR thermometry, as reported in chapter 4) but our focus here will remain on the description of phenomenology, leaving detailed measurements of temperature and flow fields as a possibility for future study.

A mixture that is too rich cannot produce a self-sustaining flame front, and will blow out to form a diffusion flame. On the other hand, a mixture that is close to stoichiometric can sustain flame propagation, but since both reactants are almost depleted at the flame, the partially

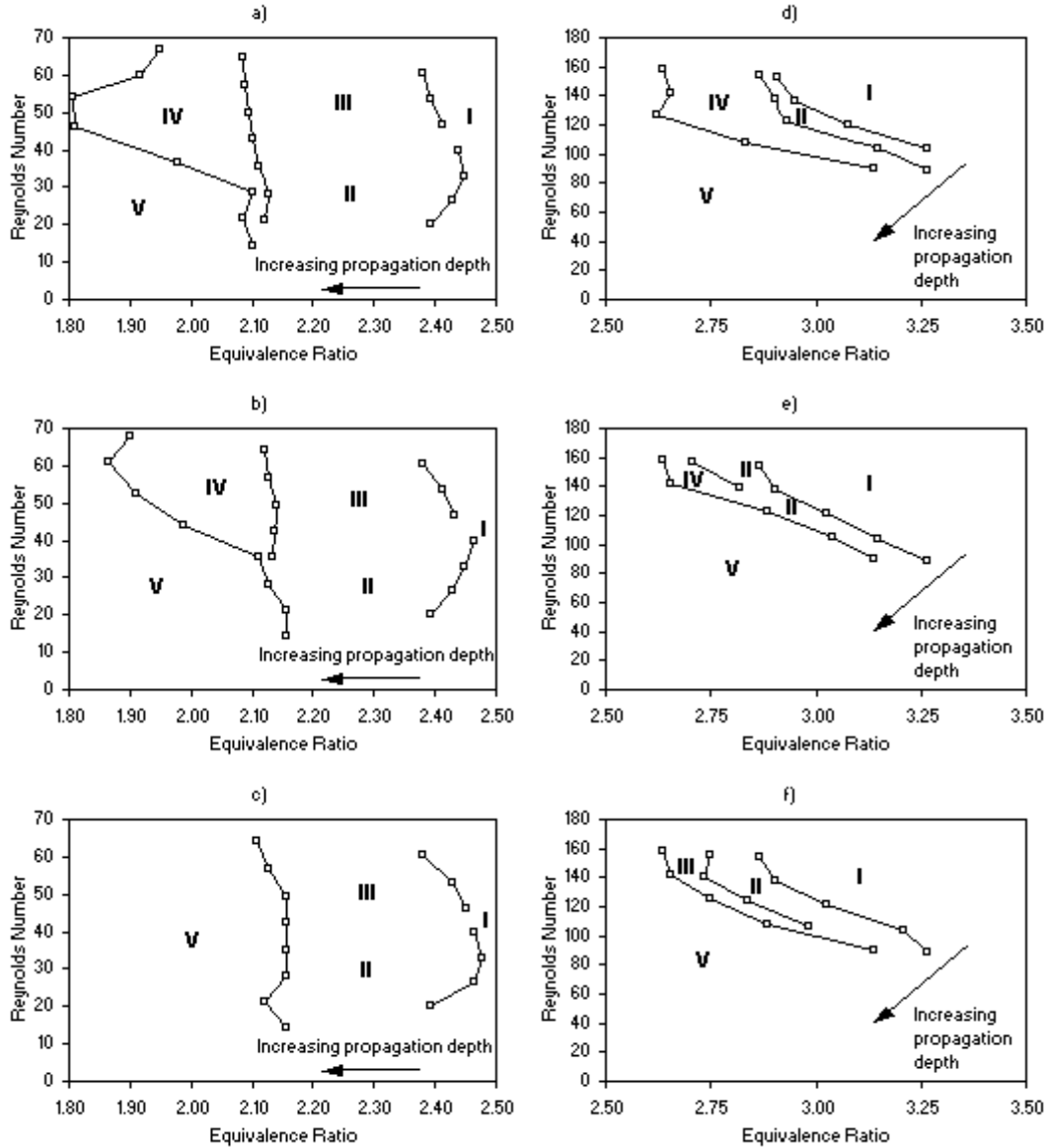


Figure 3.2. Flame behavior plotted against Re and ϕ for methane and propane with multiple tube lengths. Behavioral regimes are labeled with Roman numerals as follows. I: blowout; II: oscillations covering part of the tube length; III: stationary flame front in the tube/oscillations covering part of the tube length; IV: oscillations covering the full length of the tube; V: flashback and extinction. Lines mark experimentally observed boundaries between regimes. a) methane, 35 mm tube; b) methane, 70 mm tube; c) methane, 210 mm tube; d) propane, 35 mm tube; e) propane, 70 mm tube; f) propane, 210 mm tube.

premixed flame at the tube exit does not burn vigorously and does not generate enough heat to warm the tip of the tube enough for re-ignition to occur. So, only at intermediate equivalence ratios can stable oscillations be sustained.

For the methane case, boundaries between operational regimes are defined almost solely by equivalence ratio, a trend that suggests that oscillatory phenomena are thermally driven for this case. Only the flashback/extinction limit is affected by Re , and then only for the shorter tube lengths. Re does appear to determine at what point stationary in-tube flame fronts can exist; only for Reynolds numbers greater than approximately 40 were stationary flame fronts observed within the ducts. The shape of the flashback/extinction limit boundary is noteworthy. For low Re , the line is essentially vertical, but as Re increases the limit shifts and stable oscillations can exist for mixtures closer to stoichiometric. The explanation for this is thermal: for a fixed equivalence ratio, higher Re means fuel is being burned at a greater rate, corresponding to greater heat release. This warms the tube more quickly and to a higher overall temperature, increasing the likelihood that the tube will be warm enough at the tip to re-ignite the flame after it flashes back to the base of the tube and extinguishes – thus full-tube oscillations can be supported for mixtures closer to stoichiometric. The fact that this trend in the flashback/extinction limit is not seen for the 210mm tube, which has the greatest area and therefore the most demanding energy requirements for heating, supports this explanation. For the shortest tube at high Re , the flashback/extinction limit moves back toward the rich side. It was observed that at this point the flame front actually becomes seated at the base of the tube, which suggests that the tube became hot enough along its length that re-ignition could occur closer to the base and not only at the tip, as in other cases.

Similar behavior and oscillatory phenomena were observed for propane flames. However, as Figs. 3.2 d-f show, the boundaries between regimes were highly dependent on Reynolds number as well as equivalence ratio. This suggests that fluid mechanical factors may play a much greater role in determining the behavior of propane as opposed to methane flames. Oscillations were also observed at more fuel rich and higher Re conditions than for methane flames. The range for which oscillations covering the entire tube length could be established was smaller for propane flames, indicating that these oscillations are less stable.

Figures 3.3 a-f show the behavioral regimes plotted against mean flow velocity and the calculated adiabatic flame speed. Flame speeds were determined computationally in CHEMKIN 4.1.1 using the GRI 3.0 mechanism and the measured equivalence ratios. The disagreement between the theoretical macroscale predictions and the experimental results highlights the unique nature of the mesoscale regime. All of the studied methane mixtures have predicted flame speeds that exceed the flow velocity and would be expected to flash back, which allows oscillations to occur when a sufficient thermal gradient is present and the flammability limit is not exceeded. However, stationary flame fronts within the ducts were also observed, contrary to what would be expected in a non-adiabatic analysis given that the predicted flame speeds exceed the flow speeds. Propane flames exhibit better agreement with computational predictions, with the slopes of the curves approximately following the line of equality between flame speed and flow speed. However, the computed flame speeds for propane are less than the flow speeds, which should make flashback, and thus oscillations, impossible. Conceivably, there is heat input from the warm tube to the flame that increases the flame speed, although this point deserves further, possibly computational, investigation.

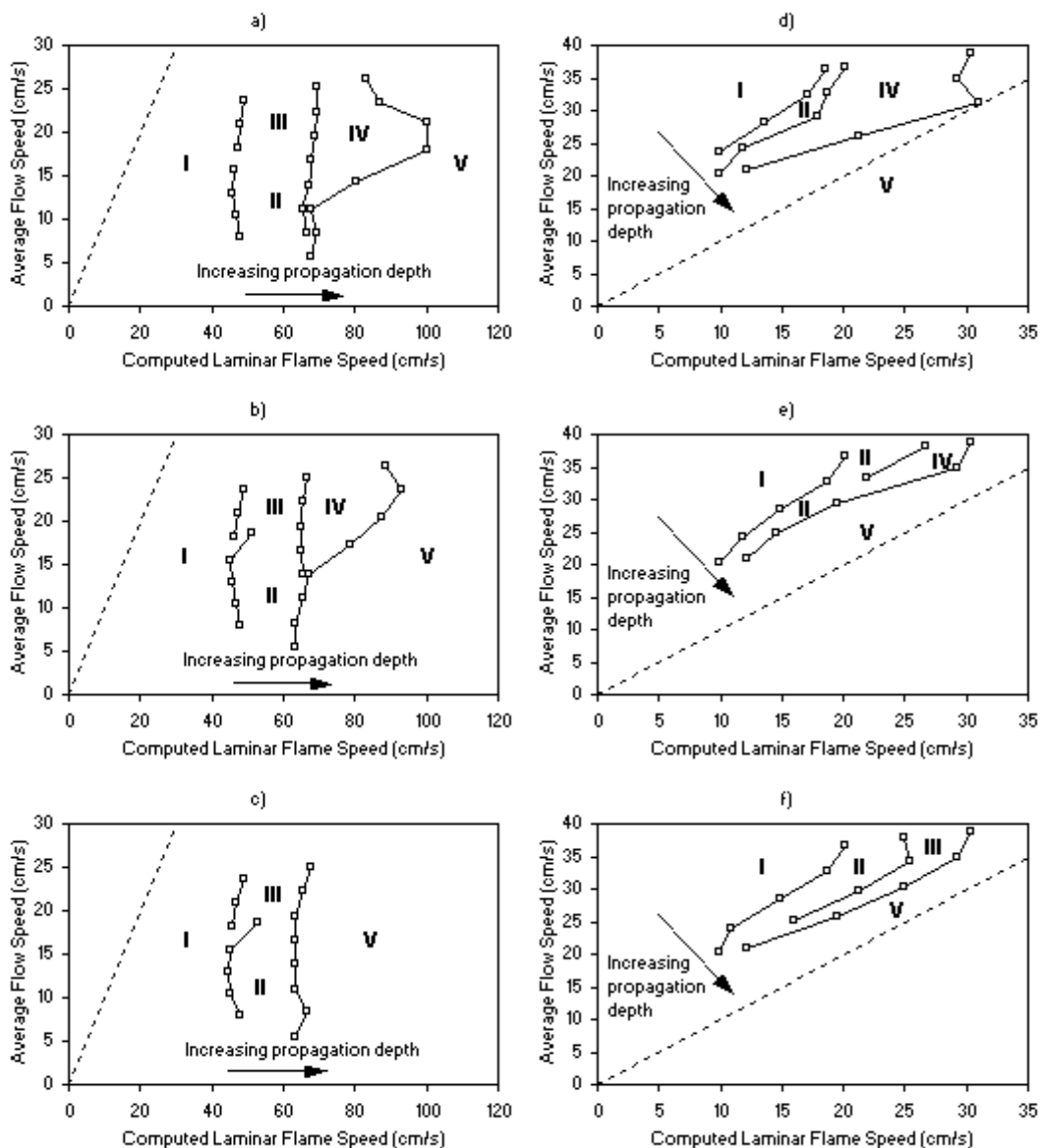


Figure 3.3. Flame behavior plotted against the measured mean flow speed and Chemkin-calculated theoretical flame speed for propane and methane with multiple tube lengths. Behavioral regimes are labeled as in Fig. 4. Lines mark experimentally observed boundaries between regimes. Plots include a dashed $y = x$ line for which the flow speed and flame speed are equal. a) methane, 35 mm tube; b) methane, 70 mm tube; c) methane, 210 mm tube; d) propane, 35 mm tube; e) propane, 70 mm tube; f) propane, 210 mm tube.

3.3 Effect of Tube Length

Changing the length of the duct affects the acoustic frequency, as well as the area for heat exchange between the flame front and the duct. If thermal effects are important and a warm tube is necessary for oscillations, a longer tube may inhibit stable oscillations since more energy is required to heat the tube and to maintain an elevated temperature. As Figs. 3.2 a-c show, this is exactly what was observed in the methane studies. As the tube length increases, the least fuel-rich section of the stable oscillation regime shrinks, and for the 210 mm tube it vanishes completely, giving a flashback/extinction limit that is nearly vertical. Moreover, no oscillations that propagated over the full length of the tube could be established for the 210 mm methane case. Little variation was seen in the rich boundaries of the oscillation and stable flame front regimes, which depend only on a thermal gradient near the tip of the tube.

This same trend is apparent for propane in Figs. 3.2 d-e as the tube length is increased from 35 mm to 70 mm. The flashback/extinction limit moves more toward the rich side, and the conditions for which stable full-tube oscillations can be established are drastically reduced. The effect of increasing the tube length from 70 mm to 210 mm is less clear. Although no stable full-tube oscillations can be established for the 210 mm tube, Fig. 3.2 f shows that the flashback/extinction limit appears to stretch into less rich mixtures in the 210 mm case. Stationary flame fronts within the tube were observed only for the 210 mm case. This is a notable departure from the behavior of methane flames, for which stationary flame fronts could be established for all three tube lengths. These phenomena suggest that fluid mechanics plays a greater role in the oscillatory behavior of propane flames.

3.4 Effect of Tube Material

Table 3.2 compares the thermal properties of the tested materials and whether each is capable of sustaining oscillations. No oscillations could be established for the materials of highest thermal conductivity and thermal diffusivity. Apparently, for highly conductive materials the notion of quenching diameter applies, while for materials of lower conductivity, novel, oscillatory behaviors are observed. Tubes made of low conductivity material have two important properties. First, they have the ability to maintain steep temperature gradients in their walls. The behavior of the tubes in this situation is analogous to that of a fin: a high temperature is produced at one end of the duct by a diffusion flame, and convective heat dissipation occurs along the length. The degree to which temperature changes with position along the fin can then be expressed in terms of the fin parameter βL . β is calculated with the following relation:

$$\beta^2 = \frac{hP}{kA_c} \quad (3.4)$$

Here h is the convection heat transfer coefficient, P is the perimeter of the fin, k is the thermal conductivity, and A_c is the cross-sectional area of the fin. Higher values for the fin parameter indicate greater influence of convective losses, and a sharper gradient in temperature along the length of the fin.

Second, tubes of low thermal conductivity follow temperature fluctuations in their boundary with significant time delay, quantified as a small Fourier number.

$$Fo = \frac{\alpha \cdot t}{L^2} \quad (3.5)$$

Here Fo is the Fourier number, α is the thermal diffusivity, t is the characteristic time, and L is the length of the tube.

Table 3.2. Comparison of thermal properties and capacity to maintain stable oscillations for tested tube materials. k is the thermal conductivity, α is the thermal diffusivity, βL is the fin parameter, and Fo is the Fourier number.

Material	k (W/m-K)	α (m ² /s)	β (m ⁻¹)	βL	Fo	Can establish stable oscillations?
Glass	1.40	7.54e-07	158	11.06	1.54E-05	Yes
Quartz	1.38	8.34e-07	159	11.14	1.70E-05	Yes
Steel	15.1	3.91e-06	48	3.37	7.98E-05	Yes
Brass	110	3.39e-05	18	1.25	6.92E-04	No
Copper	401	1.17e-04	9	0.65	2.39E-03	No

Calculation of the fin parameter for each material is complicated by the fact that convective losses occur from two surfaces: from the outer surface of the tube via free convection, and from the inner surface via forced convection. A modified version of Eqn. 3.4 was used to account for this:

$$\beta^2 = \frac{h_{outside} P_{outside} + h_{inside} P_{inside}}{k A_c} \quad (3.6)$$

Here the “outside” and “inside” subscripts denote the values for the outer and inner surfaces of the duct. For the outer surface, a reasonable estimate for the free convection heat transfer coefficient $h_{outside}$ of 10 W/m²-K was used. The flow inside the tube may be considered laminar and fully developed, and assuming a uniform heat flux between the fluid and wall, a constant Nusselt number Nu of 4.36 can be used, similarly to the well-established relation for constant-density flows [57]. Equation 3.7 can then be used to determine h_{inside} .

$$Nu_D = \frac{hD}{k} \quad (3.7)$$

Here D is the tube inner diameter.

As Table 3.2 shows, the fin parameter is smallest for the most thermally conductive materials, and increases with decreasing thermal conductivity. A larger fin parameter indicates a greater influence of convective heat losses, which should result in a steeper temperature gradient.

Examining the theoretical temperature distribution along the duct verifies this. In this specific case, one end of the duct is heated by a diffusion flame, while the other end is attached to a copper block that acts as an effective heat sink, effectively making the temperature at that end equal to the ambient. With the fin parameter known, the non-dimensional temperature at each point in the duct can then be calculated using a modified form of the equation for a fin with a prescribed temperature at one end [61]:

$$\theta = \frac{\sinh \beta(L - x)}{\sinh \beta L} \quad (3.8)$$

Here L is the duct length, x is the axial position, and θ is the non-dimensional temperature, defined as:

$$\theta = \frac{T - T_{\infty}}{T_b - T_{\infty}} \quad (3.8)$$

Where T is the temperature at a given point, T_{∞} is the ambient temperature, and T_b is the temperature at the heated end of the duct. The temperature distribution for each material is shown in Fig. 3.4. The highly conductive materials have nearly linear temperature distributions due to the predominance of conduction heat transfer, while for the less conductive materials a rapid decrease in temperature occurs near the heated end, which departs significantly from the linear profile predicted by a simple conductive transport. Because of these temperature patterns, tubes with lower thermal conductivity have convective heat losses that are effectively concentrated near the heated end.

It is pointed out that the results of Fig. 3.4 are in terms of non-dimensional temperature, with the temperature at the edge of the tube towards the diffusion flame remaining essentially unknown. This unknown temperature is not constant for all materials. What is instead essentially constant is the input of heat from the diffusion flame, which must be balanced by

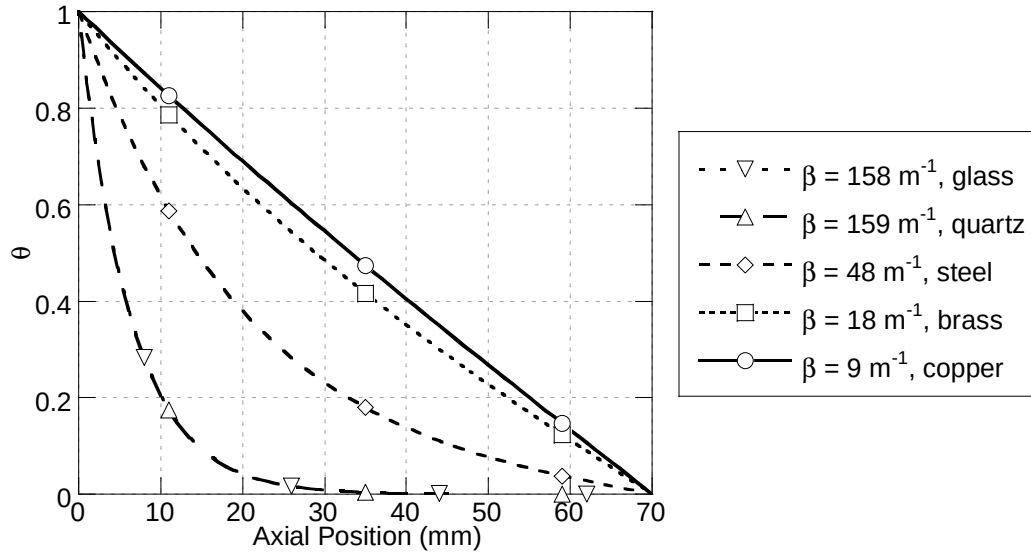


Figure 3.4. Predicted non-dimensional temperature distributions along the duct, based on the fin parameter for each material. The glass and quartz curves nearly overlap.

losses elsewhere in the system. The linear temperature distribution of highly conductive materials results in elevated temperatures, and by extension greater convective losses, farther from the heated end, along with conductive losses to the copper block at the tube base (which may be dominant). For less conductive materials, elevated temperatures occur only near the heated end. To achieve the same convective losses over this smaller area, the temperature of the duct in this region must be higher. As such, a tube made of low thermal conductivity material should produce not only a substantial temperature gradient, but also a higher temperature near the exit. Both are conducive to re-ignition of a flame during oscillation.

Fo was calculated for each material by using the tube length for L and the time between oscillations for t ; a reasonably fast oscillation frequency of 10 Hz (based on experimental observations) was used, for a characteristic time of 0.1 s. Table 3.2 summarizes the approximated Fo values. For the particular configuration of a tube that is heated at one end by a diffusion flame and cooled at the other through contact with a copper block at ambient

conditions, a high fin parameter leads to a substantial temperature gradient along the tube length and a high temperature region near the tip that could supply heat to re-ignite an extinguished mixture. At the same time, a low Fourier number indicates that this hot region will be more capable of holding its temperature after the flame extinguishes until fresh fuel and oxygen are delivered, at which instance, re-ignition will occur. Thus for a flame that is oscillating at several Hz, a stable situation can be established in a tube with low thermal conductivity that is impossible for a highly conductive tube. As Table 3.2 shows, the lower conductivity materials have higher fin parameters and lower Fo and fit this model more closely, enabling them to support stable oscillations. The higher conductivity materials have much lower fin parameters and higher Fo , indicating that they maintain smaller temperature gradients and do not hold temperatures well; as a result, they fail to maintain stable oscillations. For quartz and copper, respectively the least conductive and most conductive materials, the difference in fin parameter and Fo is multiple orders of magnitude.

The arguments based on the fin parameter and Fo point to the importance of the wall temperature distribution for combustion in small-scale ducts. This was pursued experimentally with infrared thermometry, as described in chapter 4.

Chapter 4

Infrared Thermometry Studies

4.1 Image Processing

The infrared camera described in chapter 2 captured grayscale images of the infrared intensity along the tube. To construct distributions of the tube wall temperature profiles from the IR images, the image grayscale intensity values needed to be converted to temperatures. This was done by correlating the thermocouple temperature readings to the image grayscale intensities at the thermocouple locations. As noted earlier, two images were taken for each mixture composition: one capturing the “low temperature” range and another capturing the “high temperature” range. As a result, two calibration curves needed to be produced. Each was done separately.

The “low temperature” images were used to produce the “low temperature” calibration curve. Each image was processed as follows. First, the image background intensity was measured and subtracted from all non-saturated pixels in the image. Next, making the assumption of radial symmetry in the temperature profile, a single representative intensity value was determined for each location on the tube axis. This was done by recording the intensity of the brightest pixel in each row, beginning at the row corresponding to the tube exit and continuing line by line to the row corresponding to the tube base. This produced a profile of the grayscale intensity along the length of the tube. A single representative value for each row, rather than the row average, was necessary because normal wear on the ducts resulted in the development of regions of varying emissivity, in which the pixel intensity varied despite the

temperature being uniform. This phenomenon is illustrated in Fig. 4.1. Seeking only the brightest pixels on each row ensured that the emissivity at each location had a consistent, high value.

Once all images were processed and an intensity profile was established for each image, the calibration curve was constructed. For each experiment, the grayscale intensity at each thermocouple location was taken from the intensity profile and matched to the thermocouple reading for that experiment, in order to produce a correlation of grayscale intensity to temperature. Data for which the thermocouple location had a grayscale intensity of zero, or for which it was saturated (intensity = 255), were omitted. All temperature-intensity data for a given material across all experiments were aggregated, and a third order polynomial was fitted to the data. This relation was then used to convert the intensity profile for each image into a temperature profile. After the “low temperature” profiles were completed, the “high temperature” images were processed in the same way, a new correlation was determined, and

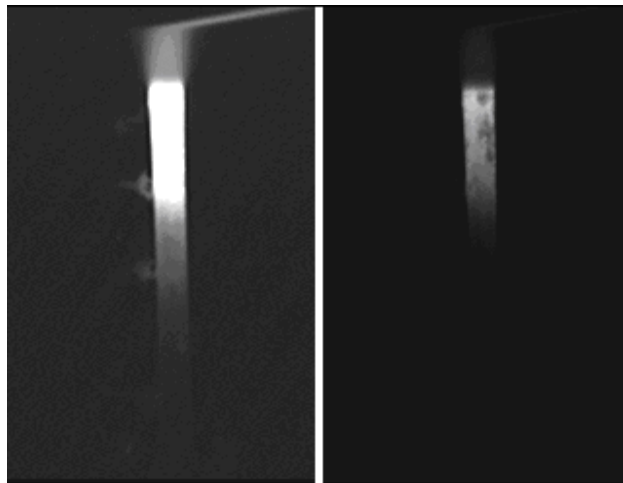


Figure 4.1. Sample pair of grayscale images of the IR signal. The conditions of the duct are the same for both images. The left image captures the lower portion of the temperature range (“low temperature” image). Hotter areas show saturation. The right image captures the upper portion of the temperature range (“high temperature” image). Cooler areas have intensity too low to be detected. Variations in the emissivity resulting from normal wear are visible in the right image.

“high temperature” profiles were produced.

Because the two images taken for each case each captured only part of the temperature range, the two partial temperature profiles were combined in order to produce a full temperature profile for the tube. Combining the profiles also helps to reconcile differences in the temperature measurements that can occur as a result of the two different correlation equations. The combination of the “high temperature” and “low temperature” profiles into a single temperature profile was done by applying the following rules at each axial position. When both the “high temperature” and “low temperature” images had intensities in the valid range (greater than zero and less than saturation), the temperature values from the two profiles are averaged. The differences between the two temperature values in such cases was different for each material. For steel, the average difference was ~ 7.5 K. The difference was <20 K for over 97% of cases, and <10 K for 72% of cases. Larger temperature differences were observed in the quartz case, perhaps due to the larger temperature range and steeper temperature gradients for this material (discussed later in this chapter). For quartz, the average difference was ~ 19 K. The difference was <50 K for over 97% of cases, and <30 K for 81% of cases. When the “low temperature” image is saturated, the temperature value from the “high temperature” profile is used. Likewise, when the “high temperature” image has zero intensity, the temperature value from the “low temperature” profile is used. If the “low temperature” image has zero intensity, the minimum temperature from the correlation equation is used (corresponding to either ambient temperature or the minimum temperature that the camera can observe.). In the event that the “high temperature” image has maximum intensity (saturation), the maximum temperature from the correlation equation is used (corresponding to the maximum temperature the camera can observe).

4.2 Flame Phenomenology

The observed flame phenomenologies in the tubes were classified according to the categories described in the phenomenology experiments of chapter 3 and illustrated in Fig. 3.1:

- (1) blown out flame at the duct exit,
- (2) stationary flame front,
- (3) oscillating flame propagating over part of the tube length,
- (4) oscillating flame propagating over the full length of the tube, and
- (5) extinction.

Maps of the flame phenomenologies for the quartz and steel tubes are presented in Fig. 4.2. It should be noted that because of the opaque nature of the steel tube, determining the penetration depth of oscillating flame fronts was not feasible, so no distinction was made between oscillations that propagate over only part of the tube length and oscillations that propagate over the full tube length.

The basic pattern of behavioral regimes for both the steel and quartz tubes is the same as what was reported in [42]. Blown out flames occur at high equivalence ratios (ϕ), oscillations and stationary flame fronts occur when ϕ drops below a certain threshold, and extinction occurs when ϕ is too low. Flame behavior appears to be determined by equivalence ratio rather than Re . Stationary flame fronts, however, are observed only at sufficiently high Re .

There are notable differences between the flame phenomenology in the quartz and steel tubes. The most significant difference is that for the quartz tube, stable oscillations and stationary flame fronts can be established for a wider range of ϕ than in the steel tube. The onset of oscillations and stationary flame fronts occurs at an equivalence ratio that is 0.1 higher for the quartz tube compared to the steel tube. This onset generally occurs at around $\phi = 2.4$ for the

quartz tube and $\phi = 2.3$ for the steel tube. The extinction limit is also more generous for the quartz tube. For the $Re = 40$ to $Re = 70$ range, extinction occurs at equivalence ratios 0.1 lower in the quartz tube than in the steel tube, a notable increase in the stable range. As a result of these two effects, for most of the studied fuel flow rates the stable oscillating/stationary flame front regime covers a 0.2 broader range of ϕ for the quartz duct compared to the steel duct. Also noteworthy is that many more stationary flame front cases are observed for the quartz tube. In the steel tube, these same mixtures generally produce oscillating flames instead.

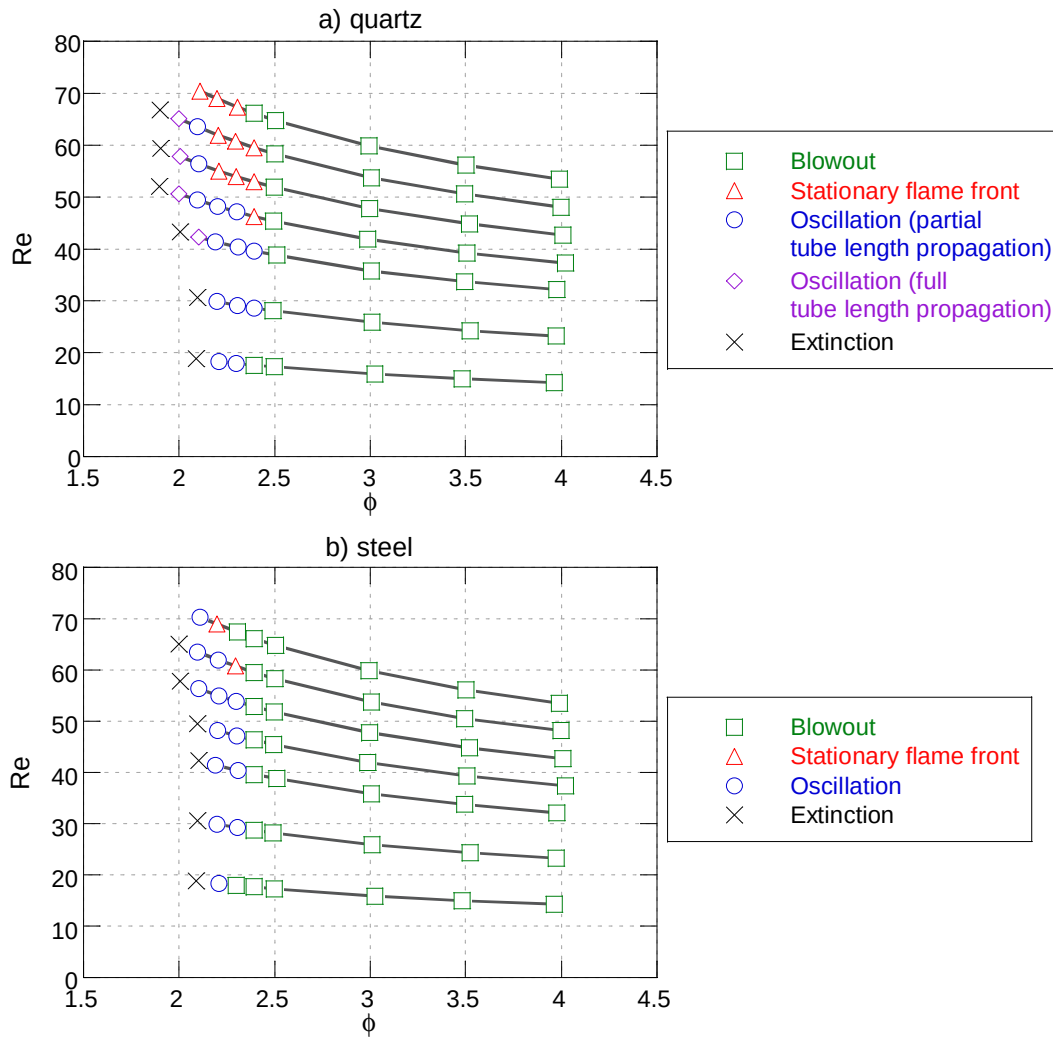


Figure 4.2. Flame phenomenology maps. Lines connect experiments with equal fuel flow rates. Since the penetration depth of oscillations in the steel tube could not be observed, no attempt was made to distinguish between oscillations that propagated over part of the tube length and oscillations that propagated over the full tube length.

4.3 Effects of Equivalence Ratio/Flame Phenomenology on the Wall Temperature

Figures 4.3 and 4.4 show the temperature distributions for each duct material and fuel flow rate. Within each plot, the fuel flow rate is the same for all curves. One point deserves mention here. As an unavoidable effect of the different emissivities of the two tube materials, and the different camera settings used in each case, the minimum temperature at which the camera is capable of detecting the IR emissions from the tube is different for quartz and steel. As a result, the minimum plotted temperature (seen as the point at which the temperature curve becomes a vertical line) is higher for the steel tube. This is not indicative of a higher ambient temperature in the steel experiments, and does not suggest that the actual minimum temperature in a given steel case is necessarily higher than the actual minimum temperature of a given quartz case.

In general, the temperature is highest at the tube exit and lowest at the tube base for all flame behaviors. This is expected, as the tube exit holds a diffusion flame and experiences relatively low thermal losses compared to the tube base, which is far from the flame and in contact with a brass plate and copper block that act as a heat sink. Another general trend is that the cases with lower equivalence ratios, for which the flame exhibits oscillatory or stationary flame front behavior, have much higher temperatures farther from the tube exit, while the exit temperature is approximately unaffected.

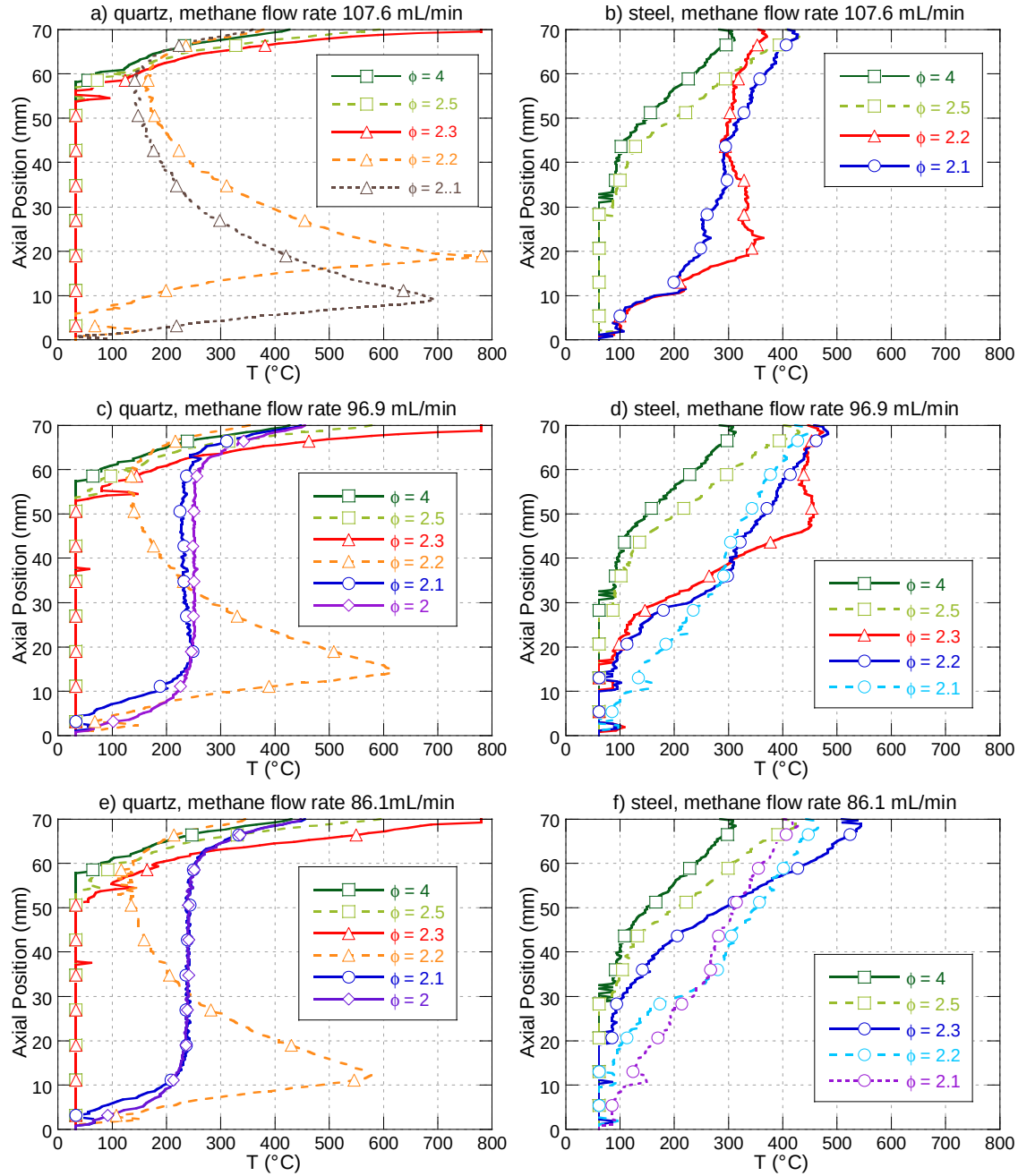


Figure 4.3. Temperature distributions for each duct material and fuel flow rate. Each curve represents the steady state temperature distribution for a single equivalence ratio. The fuel flow rate is uniform for all curves within each subplot. The flame phenomenology for each equivalence ratio is indicated using the symbols defined in Fig. 4.2.

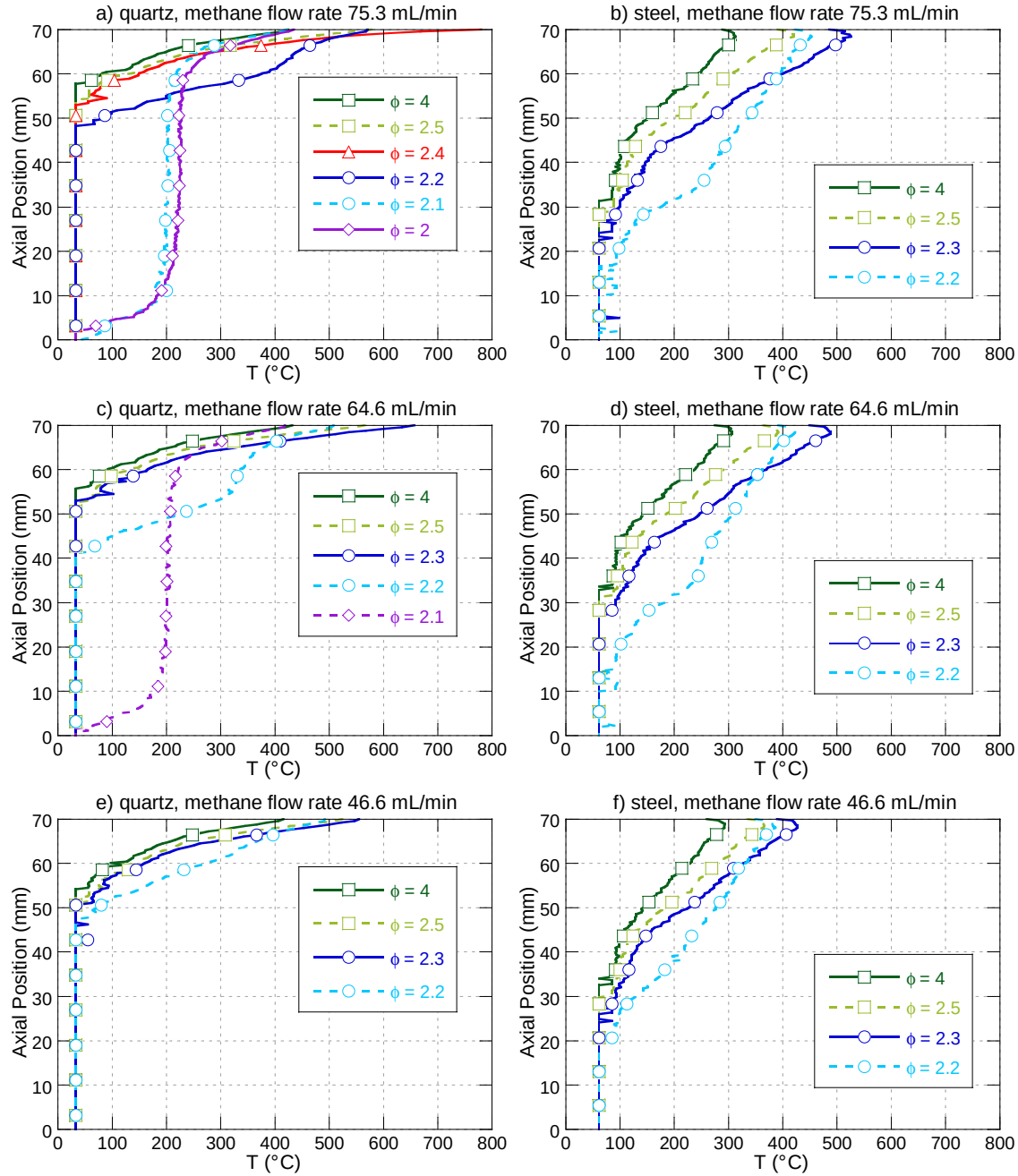


Figure 4.4. Temperature distributions for each duct material and fuel flow rate. Each curve represents the steady state temperature distribution for a single equivalence ratio. The fuel flow rate is uniform for all curves within each subplot. The flame phenomenology for each equivalence ratio is indicated using the symbols defined in Fig. 4.2.

The highest equivalence ratios correspond to cases in which the flame is blown out of the tube. Starting at the tube exit and moving downward, the temperature is initially at a maximum, but decreases rapidly farther from the tube exit. The temperature decrease is much sharper in the quartz case, which is an expected consequence of the lower thermal conductivity. Decreasing the equivalence ratio effectively shifts the temperature curve to the right, increasing the peak temperature at the exit, the temperature throughout the heated area, and the depth at which elevated temperatures are observed. However, this effect is far less pronounced than the impact of changing from a blowout to an oscillatory mode of operation, or to a stationary flame front within the tube.

When Re is high enough, equivalence ratios slightly lower than those for blowout conditions will produce stationary flame fronts. The most notable aspect of the temperature profile for this mode of operation is that two temperature peaks exist. One always lies the tube exit, which hosts a diffusion flame. The other peak occurs at the location of the stationary flame front. In some cases this flame front may rest very near the tube exit, and the combined output of these two flames causes massive elevation in the local wall temperature, for example in the quartz tube in Fig 4.3.c) for $\phi = 2.3$. When the flame front is farther from the tube exit, one of the two temperature maxima tends to be larger than the other. For the quartz tube, the higher wall temperature is observed near the stationary flame front. For the steel tube, the higher wall temperature is generally observed at the tube exit, where the diffusion flame exists. Moving away from either flame front, the temperature decreases.

The lowest equivalence ratios for which stable combustion was established produced oscillating flames. It should be emphasized that although the flame front moves within the tube during oscillations, the temperature distribution of the duct wall remains constant at steady state.

The wall temperature profile for an oscillating flame configuration can have multiple patterns. The optical accessibility of the quartz tube allowed us to determine that the shape of the temperature profile is dependent on the depth of penetration of the oscillating flame front. For flame fronts that have a shallow penetration depth (propagating only near the tube exit), the shape of the temperature profile is essentially identical to that produced by a blown out flame. As the equivalence ratio will be lower than in a blown out case, the temperature curve is shifted to the right as noted above. This pattern can be observed in the quartz tube in Fig. 4.4.c) for $\phi = 2.3$. When the flame front propagates farther into the tube, but not all the way to the base, the wall temperature profile resembles a piecewise function. Moving down from the tube exit, wall temperature decreases gradually with distance until the depth of penetration of the flame front. Below this point, the temperature decreases much more rapidly with distance. This pattern can be observed in the quartz tube in Fig. 4.4.c) for $\phi = 2.2$ and in the steel tube in Fig. 4.4.d) for $\phi = 2.2$. When the flame front propagates over the full tube length, different patterns are observed in the quartz and steel tube. In the quartz case, the tube wall achieves a nearly uniform temperature for most of its length. Deviations are observed at the ends, with temperature sharply increasing to a maximum at the tube exit and sharply decreasing to a minimum at the base. This pattern can be observed in the quartz tube in Fig. 4.3.e) for $\phi = 2$. The region of uniform wall temperatures was not observed in the steel tube, even for oscillating flames that were believed to propagate over the full length of the tube. What is observed instead in the steel tube is a nearly constant decrease in temperature with distance over the tube length. Near the tube base, the temperature may decrease more rapidly. This pattern can be observed in Fig. 4.3.f) for $\phi = 2.1$. It is noted that in a low conductivity material, flame oscillation is compatible with a constant temperature along the tube wall, which is particularly attractive for coupling with direct energy conversion

modules, such as thermoelectrics or thermophotovoltaics. Such devices are often effective only for narrow temperature ranges, and would benefit from a system that maintains a nearly uniform temperature.

For non-oscillating cases, the higher thermal conductivity of the steel tube promotes more uniform distribution of enthalpy along the duct, whereas the lower thermal conductivity of the quartz tube creates more localized high temperature areas. Even so, the same low thermal conductivity of quartz, notably, promotes better enthalpy distribution and near-uniform wall temperatures for oscillating cases. In addition, because the low thermal conductivity of quartz promotes localized high temperature areas, identical flame conditions will produce a higher temperature at the duct exit for the quartz tube compared to the steel tube. A higher wall temperature at the tube exit facilitates re-ignition of the flame front, which promotes stable oscillations. This explains why oscillating flames can occur at lower ϕ in the quartz tube compared to the steel tube. This higher exit temperature may also be the reason why oscillating and stationary flame fronts are observed at higher ϕ in the quartz tube. High temperatures near the exit will preheat the reactant gases prior to entering the flame, potentially boosting the flame speed high enough that upstream propagation into the tube becomes possible. In this manner, the more generous ϕ limits on both sides of the oscillatory regime in the quartz duct can potentially be attributed to the lower thermal conductivity of the material.

4.4 Comparison of the Effectiveness of the Flame Behaviors for Duct Wall Heating

Table 4.1 summarizes the material properties of each duct. The effectiveness with which each flame behavior transfers energy to the duct can be evaluated both in terms of the magnitude of transferred energy, and in terms of the resulting distribution of energy. Since the wall

Table 4.1. Duct properties.

Material	ρ (kg/m ³)	C_p (J/kgK)	k (W/mK)	Duct mass, m (kg)	α (m ² /s)	Energy transfer needed to change average wall T , mC_p (J/K)
Quartz	2220	745	1.38	9.76e-3	8.34e-7	7.27
Steel	8055	480	15.1	3.54e-2	3.91e-6	17.01

temperature corresponds to its energy content, the relative magnitude of the energy transfer can be quantified in terms of the average temperature elevation of the duct above the baseline temperature of a cold tube at the beginning of the experiment. This was calculated for each fuel flow rate-equivalence ratio combination by computing the mean temperature of the distribution and subtracting the baseline temperature for the material. The uniformity of the energy distribution can be quantified in terms of the standard deviation of temperature for each wall temperature distribution. These data are summarized for each material and fuel flow rate in Fig. 4.5. As the energy input to the system is the same for each fuel flow rate, the effectiveness of the energy transfer for different phenomenologies can be compared directly.

Blown out flames, which occur at high ϕ , produce the smallest elevations in the average temperatures, and are least effective at heating the tube. Since combustion begins at the tube exit, much of the heat release occurs beyond the tube, giving little opportunity for wall heating. The standard deviation of wall temperature in such cases is low, largely because the bulk of the tube experiences little to no heating and has a temperature near ambient. What heating does occur is very nonuniform, being concentrated entirely at the tube exit, so while the standard deviations are among the lowest observed in these experiments, they are not very low in an absolute sense. Decreasing ϕ will cause combustion to occur earlier and move the flame closer to the tube, resulting in a slightly higher average temperature. The standard deviation also increases by a similar amount.

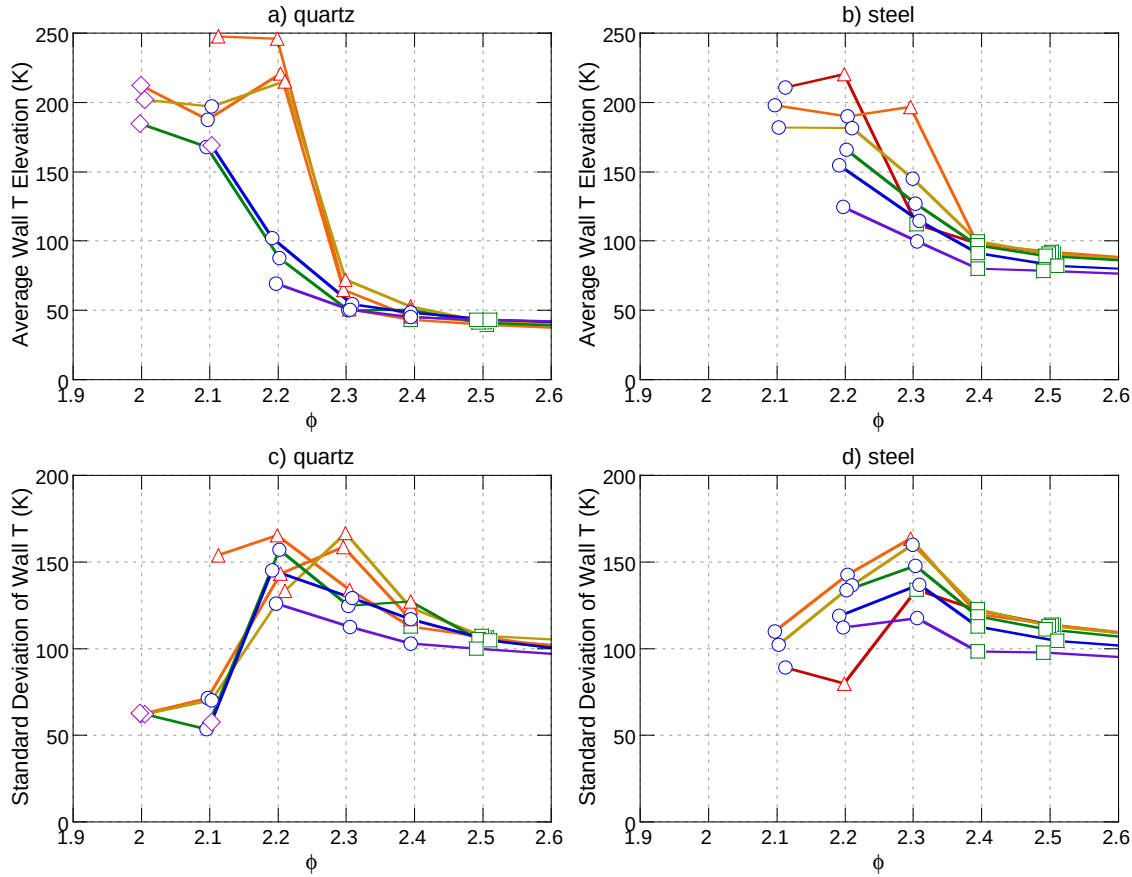


Figure 4.5. Average elevation of the wall temperature above the baseline and standard deviation of wall temperature for each experimental condition. The flame phenomenology at each point is indicated using the symbols defined in Fig. 4.2. Lines connect conditions with identical fuel flow rates.

When ϕ becomes low enough, a flame can move fully into the tube in the form of a stationary or oscillating flame front (around $\phi = 2.4$). When this occurs, there is a substantial increase in the average wall temperature elevation, as the wall becomes much more exposed to the energy release of combustion. Compared to a blown out flame, the average wall temperature elevation can be as much as 4 times as high in a steel tube, and more than 8 times as high in a quartz tube. How much of an increase occurs depends on the maximum depth of the flame front: if the flame front is deep within the tube, the hot products will have much more time to transfer heat to the duct walls before exiting than if the flame front is very near the tube exit. As noted previously, the maximum depth of the flame front increases as f decreases, so the average wall

temperature increases with decreasing ϕ as well. Changing from a blown out flame to an oscillating or stationary flame front is associated with an increase in the variance of the wall temperature. This is an expected consequence of increasing the heated area of the wall while still primarily heating the exit. This trend continues as ϕ decreases, but only to a point. When the flame fronts travel far enough into the tube, the heating becomes more uniform and the standard deviation of the wall temperature begins to decrease with decreasing ϕ . This is especially apparent in Fig. 4.5.d).

Some differences are apparent in the trends for stationary and oscillating flame fronts. The highest average wall temperatures occur with stationary flame fronts, particularly those residing deep within the tube. A stationary flame front should theoretically provide more heating to the wall than an oscillating flame that penetrates to the location of the stationary flame front. Since the stationary flame front always has the same depth, all of its hot products will have the same amount of time in contact with the walls before they exit the tube. In comparison, an oscillating flame front reaches that depth only momentarily, and will spend a large portion of the time closer to the tube exit, which will give the hot products less time to transfer heat to the walls before exiting. The standard deviation of temperature is generally very high for these deep stationary flame fronts, and the temperature profiles for these cases, for example in quartz in Fig. 4.3.a) for $\phi = 2.1$ and $\phi = 2.2$, show a very nonuniform distribution, with most of the temperature elevation occurring in a large peak at the stationary flame front. An exception to this trend is seen in the steel tube in Fig. 4.3.b) for $\phi = 2.2$, where the stationary flame front rests at a depth that happens to produce a relatively low wall temperature variance. Oscillating flame fronts are capable of producing average wall temperatures nearly as high as stationary flames, and can produce a much more uniform temperature distribution. The lowest temperature standard

deviations of all were observed for oscillating flames that propagate over the full length or nearly the full length of the tube, for example in quartz in Fig. 4.3.e) for $\phi = 2$.

A few differences exist for the tube materials. On average, the mean wall temperature elevation is similar for quartz and steel tubes. The range is greater for the quartz tube however, with both the highest and lowest average wall temperature elevations occurring in this material. For steel, the average wall temperature elevation is somewhat more consistent. The standard deviation of wall temperature is generally similar for the two materials, although the lowest observed variance is seen in the quartz tube, for several cases in the $\phi = 2.0 - 2.1$ range.

While temperature elevations are an effective metric for comparing the effectiveness of energy transfer for the different flame behaviors, temperature alone does not present a complete picture of the duct energy content. The properties of the duct are also a major factor, and as noted in Table 1, the different density and specific heat values of the duct materials give rise to very different energy requirements to raise the temperature of the duct by the same amount. Expanding on the analysis above, the total energy added to the duct in each case was calculated using Eqn. 4.1:

$$\Delta H = \rho C_p V \overline{\Delta T} \quad (4.1)$$

Here ΔH is the change in the enthalpy of the duct, ρ is the material density, C_p is the material specific heat, V is the duct volume, and $\overline{\Delta T}$ is the average temperature elevation of the entire tube above the baseline measurement of a cold tube (the data presented in Fig. 4.5 a) and b)). Figure 4.6 shows the energy added to the tube for each experimental condition and duct material. The shapes of the curves are identical to what is seen in the plots of average temperature elevation in Figure 4.5 a) and b), but the magnitudes are very different. In all cases, the energy content of the steel tube is much greater. For the poorly-heating blowout cases, the energy added

to the steel tube is generally 4 times greater than a quartz tube. For the oscillating and stationary flame front cases, which provide better heating, the energy added to the steel tube is at least double that of the quartz case.

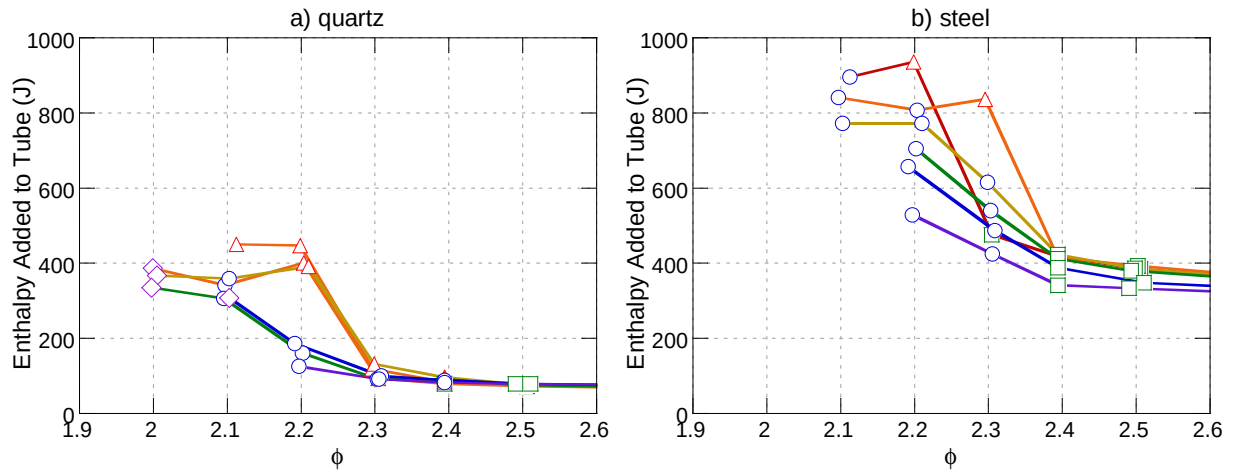


Figure 4.6. Energy added to the tube for each experimental condition. The flame phenomenology at each point is indicated using the symbols defined in Fig. 4.2. Lines connect conditions with identical fuel flow rates.

Chapter 5

Emissions Studies

5.1 Flame Phenomenology

The observed flame phenomenology for these experiments is shown in Fig. 5.1. The fuel flow rates studied in this investigation are a subset of those used in the IR thermometry experiments of chapter 4. These flow rates were selected based on their observed ability to produce a variety of novel flame phenomena including a number of oscillating cases. Although the mixture conditions were identical in this study and the IR thermometry experiments, slightly different phenomenology was observed in some cases. Most notably, the three highest fuel flow rates produced blown out flames at $\phi = 2.4$ in this study, whereas in the IR studies they produced stationary and oscillating flame fronts within the tube. This variation is likely due to different tubes being used in each study. Unlike the tube of the IR experiments, the tube used for this study appeared to have a slight constriction at the open end as a result of machining. This could cause small changes in the fluid flow and thermal situation, which may produce the slight boundary shifts observed here. Interference from the probe is considered unlikely, as it was positioned in the exhaust region beyond the luminous region of the flame at all times. Aside from this minor variation, the phenomenology matches what was observed in the IR experiments and is consistent with the patterns observed in the phenomenology studies of chapter 3: blown out flames at the highest ϕ , stationary and oscillating flames at lower ϕ , and extinction when ϕ is too low.

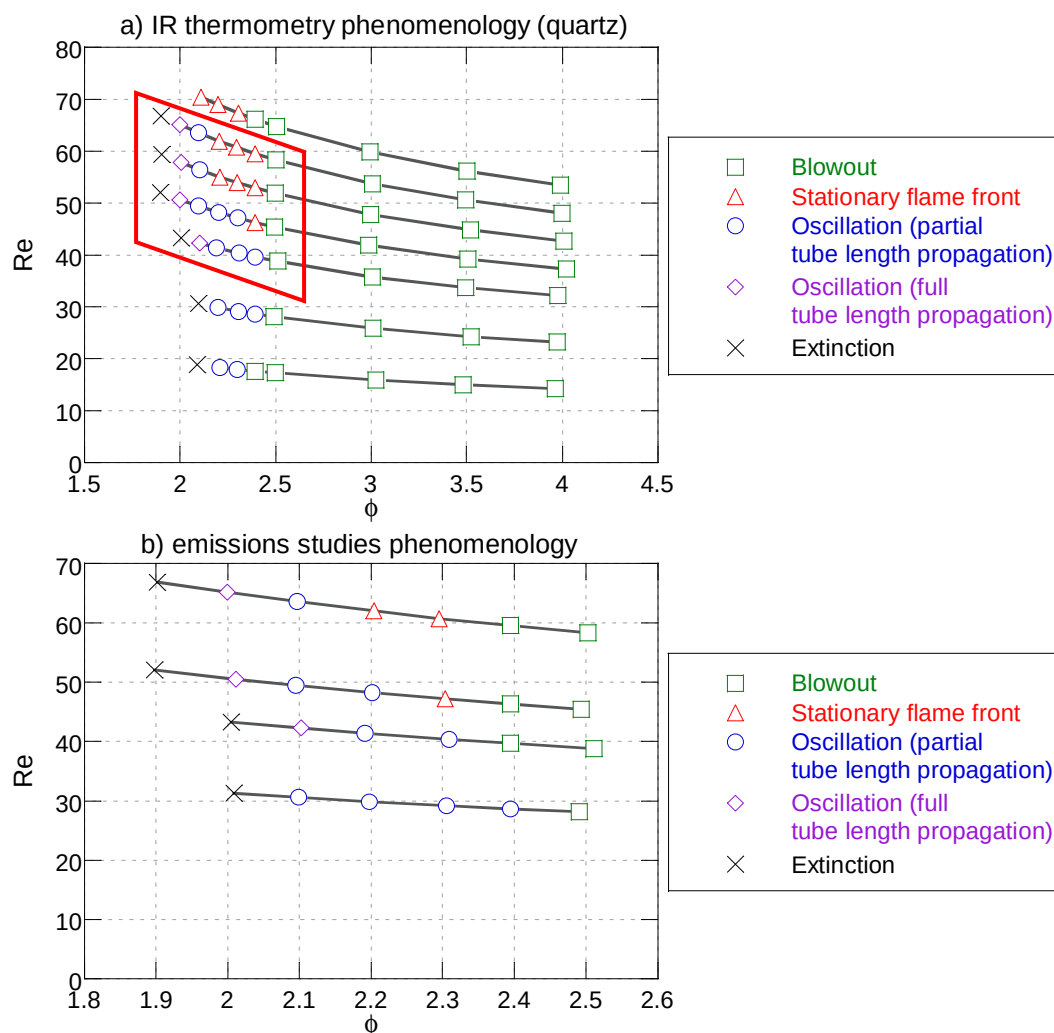


Figure 5.1. Flame phenomenology maps. Lines connect experiments with equal fuel flow rates. Panel a) shows the phenomenology observed in the IR thermometry experiments of chapter 4. The frame outlines the subset of conditions used in the emissions studies. Panel b) shows the phenomenology observed in the emissions studies described here.

5.2 Calibration

The areas of the chromatogram peaks were determined using the integrator of the MS analysis software. The area of each chromatogram peak corresponds to the abundance of a particular species. Experiments involving combustion products sampling with similar equipment have shown that this relationship is approximately linear [62]. Assuming this holds true for methane over the range measured in this experiment, the fuel consumption could be determined

by taking a ratio of the methane peak area in a reacting case to the peak area in the corresponding non-reacting case.

In order to verify this linearity assumption, a simple experiment was conducted. A connection was made between the GC inlet and the line that supplied the fuel-oxygen mixture to the tube. This arrangement is shown in Fig. 5.2. This allowed samples of the reactant mixture to be taken into the GC directly, without using the probe. A constant flow of oxygen was established, and the methane flow rate was varied in order to produce mixtures with chromatogram peak areas that covered the range observed in the experiments. As the samples in the experiments were taken in the exhaust region, the methane concentrations were generally low relative to other species, even in non-reacting cases (due to entrainment of air). Producing similarly low methane concentrations for the linearity tests required a very high flow rate of oxygen relative to the methane. Suitable concentrations of methane were produced with an oxygen flow rate of 100 mL/min, while the methane flow rates were less than 12 mL/min.

The results of the linearity tests are shown in Fig. 5.3. The methane peak area is very closely linear with methane flow rate (with a regression coefficient equal to 0.99964). Because of this, good accuracy can be obtained by calculating the methane consumption as a ratio of the chromatogram peak areas for a reacting and non-reacting case.

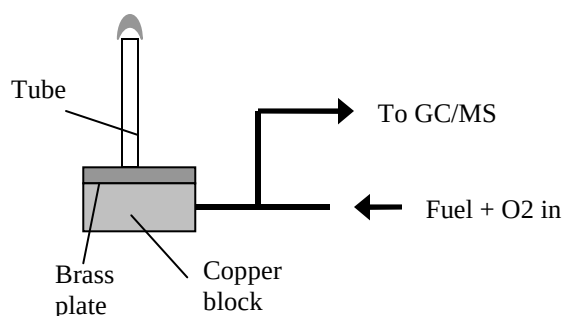


Figure 5.2. Modified apparatus used in linearity tests.

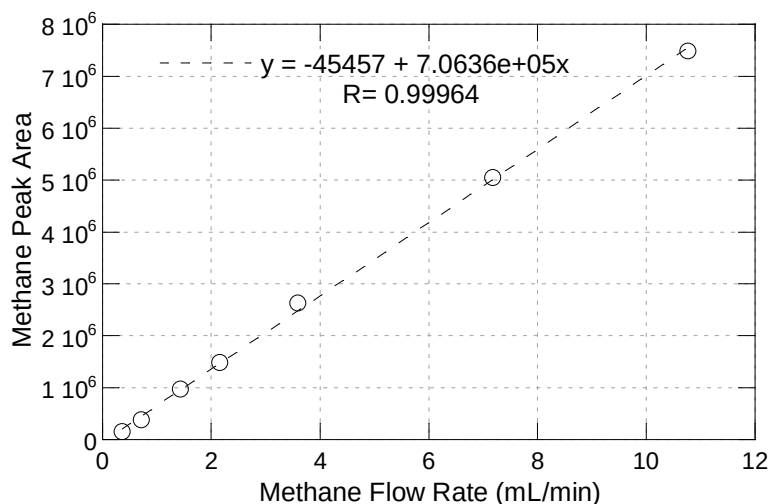


Figure 5.3. Tests showing a linear relationship between the methane abundance and the area of the chromatogram peak for methane.

5.3 Fuel Consumption and Leakage

The portions of fuel that are consumed and that remain after escaping combustion were calculated for each of the cases of Fig. 5.1 using the method described in the previous section, by taking a ratio of the peak areas for a reacting and non-reacting case. It is emphasized that the GC/MS analysis employed here is strictly an averaged sampling technique, and does not provide instantaneous data. Several minutes are required to collect a gas sample, and analysis of the sample takes an additional 20 minutes. By comparison, the oscillating flames studied here had a minimum oscillation frequency of several Hz. The results for each fuel flow rate are shown in Fig. 5.4. Panel 5.4.d) merits special discussion. Experiments with the highest fuel flow rate of 96.9 mL/min, were conducted first, while the experimental technique was under development and not yet refined. Instead of repositioning the probe near the flame for each new equivalence ratio, the probe was positioned high enough to be in the exhaust region for all ϕ and left in that position throughout the experiments with this flow rate. In addition, a slightly different GC/MS

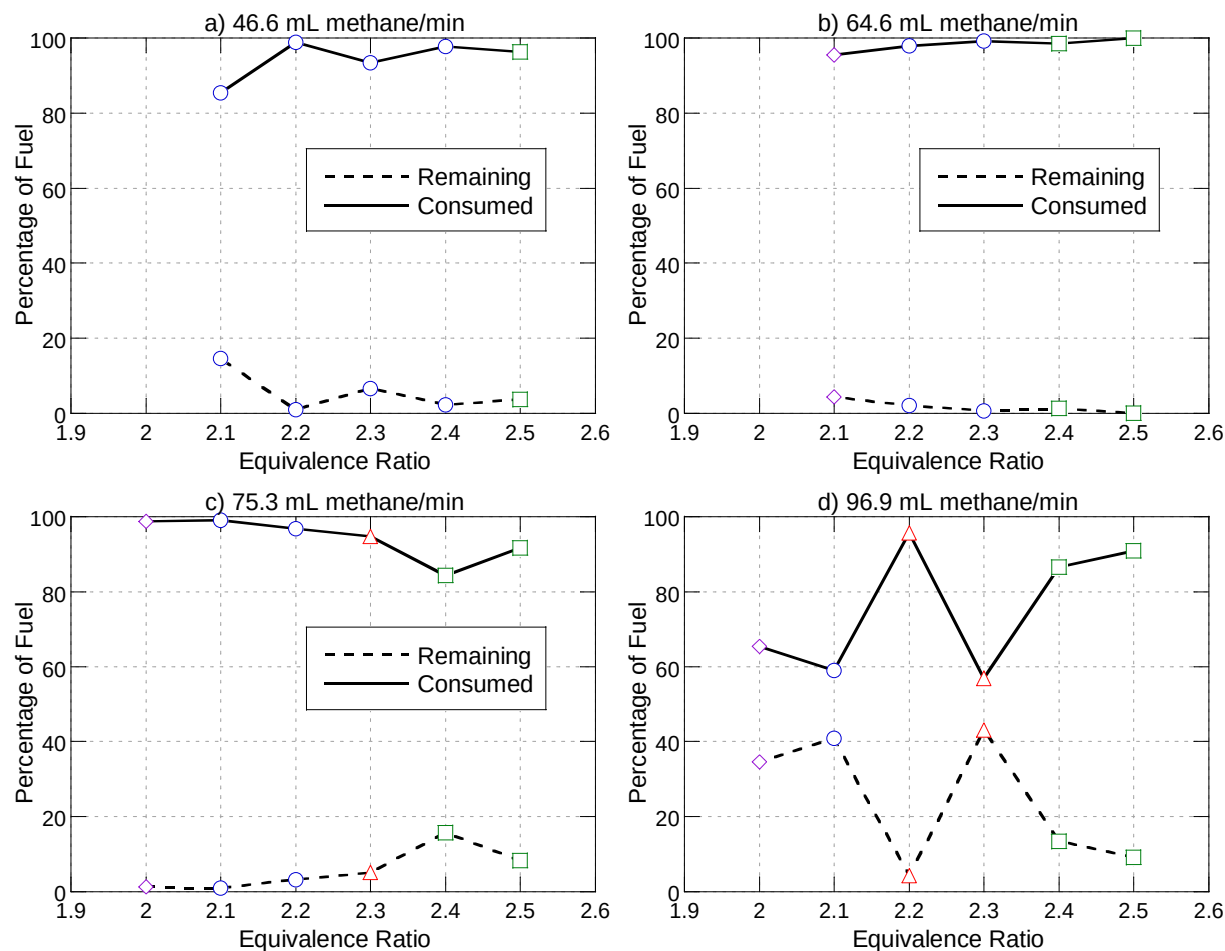


Figure 5.4. Percentage of methane consumed and remaining for each experimental condition. Each subplot shows the results for one fuel flow rate. The values of the two curves sum to 100 at each point. Flame phenomenology at each point is indicated using the symbols defined in Fig. 5.1.

analysis program was used that did not include heating the oven to the column conditioning temperature for 5 minutes at the end of each run (an aspect intended to help clear impurities). The high variation in the results for this flow rate suggest that the older procedure was sub-optimal, however this did not become apparent until after the data were analyzed and there was not sufficient time to repeat the experiment. As such, the results in Fig 5.4.d) are considered to be of limited validity, but are included for completeness.

Panels 5.4 a-c, which correspond to lower fuel flow rates of 46.6, 64.6, and 75.3 mL/min show consistently high values for the fuel consumption percentage and low values for the

percentage of fuel remaining after combustion. In addition, no measurable quantities of CO or other combustion intermediates were detected. This pattern holds throughout all experiments; there is no strong evidence of variation with flow rate or flame phenomenology. The combustion reaction appears to cause depletion of the parent fuel regardless of the flame behavior. (It is noted that the flame phenomenology at each point is identified using the symbols defined in Fig. 5.1.) Moreover, the absence of CO from the products leads to the inescapable conclusion that independent of phenomenology the combustion is complete! This runs counter to intuitive expectations.

As noted in chapter 3, high-speed video recordings of oscillating flames have revealed that when the propagating flame front extinguishes, the diffusion flame goes out as well. This would seem to create the possibility for unburned fuel to escape from the tube before re-ignition occurs. Even if the flame re-ignites at the very moment when the fresh reactant mixture finally reaches the tube exit, there will still be a gap in combustion during which incomplete combustion products of the propagating flame could escape. The fact that these species are not observed in any appreciable quantity suggests that even this form of leakage does not occur, which raises the question of how the combustion reaction is brought to completion. There are at least two possible answers to this.

The first possibility is that, despite appearances, the diffusion flame does not truly extinguish at all. When the propagating flame front extinguishes, the diffusion flame may simply become non-luminous (or of luminosity that could not be detected with the particular settings of the high-speed camera) while it burns the incomplete products left behind by the propagating flame front. This state could last until fresh reactants reach the tube exit, at which time normal combustion resumes and the flame returns to its luminous state. The problem with

such an explanation is that it is difficult to imagine why the diffusion flame would behave differently without a change in chemistry. The partially-burned mixture that fuels the diffusion flame should have essentially the same composition both while the propagating flame is present and after it extinguishes. Even if unusual species that could evoke such a change are produced by the propagating flame at the moment of extinction, it would still take time for them to travel to the diffusion flame at the tube exit. However, the change in the diffusion flame – its apparent extinction – occurs immediately upon extinction of the propagating flame front; there is no delay. This possibility therefore seems unlikely.

The second possibility is that the diffusion flame does extinguish and allow incomplete combustion products to leak out, but these products are consumed after the flame re-ignites. Observations of the shape of the diffusion flame support this possibility: when re-ignition occurs, the diffusion flame is observed to leap above the tube, reaching a height much greater than what is seen in any other condition. This is only a momentary occurrence, as the next extinction occurs shortly after, but this brief extension in the diffusion flame may represent a downstream propagation of the flame front that consumes escaped combustion intermediates. In such a case, incomplete products will indeed escape from the tube, but will only exist until they are consumed in the next re-ignition cycle. The net reaction would then be one of complete combustion. The reason why the diffusion flame extinguishes along with the propagating flame front remains unknown. Perhaps the extinction of the propagating flame front causes a sudden pressure change and therefore an expansion that quenches the diffusion flame.

5.4 Products of the In-tube Flame

One additional test was performed in order to examine what species are produced by the rich-burning flame fronts that exist within the tube during stationary and oscillating flames. An experiment was conducted to take a sample of the gases in between the premixed flame front in the tube and the diffusion flame at the exit. A stationary flame front condition was chosen for the purpose, as the steady nature of this phenomenology allowed adequate time for the lengthy sampling procedure. It is expected that the results can be generalized to oscillating flame fronts as well. With methane and oxygen flow rates of 96.9 and 88 mL/min, respectively, a stationary flame front was produced 4.5 cm below the tube exit. The equivalence ratio for this condition was 2.2. The quartz probe was inserted into the tube through the open end; the probe inlet was positioned 2 cm below the tube exit. The insertion of the probe produced no discernible effects on the stationary flame front. The system was allowed 5 minutes to reach equilibrium, after which a sample of the gas was taken and analyzed as described in section 2.3. For comparison purposes, an additional sample of the gases at this position was taken for a non-reacting mixture with the same flow rates.

In the reacting case, the major detected species were O_2 , N_2 , CH_4 , CO , ethane, ethylene, and trace amounts of argon. The presence of N_2 , Ar, and unreacted O_2 is indicative of an air leak in the sampling line, but this should have little impact on the species profile as the sampled gases are expected to be rapidly cooled while traveling through the quartz probe. CO was the most abundant combustion species in the sample. The methane was measured to be 85.4% consumed at the point of sampling, leaving 14.6% unreacted methane. A small fraction of ethane is normally present in the methane supply. Comparison of the ethane peaks in the non-reacting and reacting samples shows a net consumption of ethane. Ethylene, on the other hand, was not

detected in the non-reacting sample in any measurable quantity, and may be assumed to be a product of reaction.

Perhaps the most major finding of this experiment was the dominance of CO in the products of the in-tube flame. The absence of this species in the post-diffusion flame exhaust confirms that complete combustion occurs for all flame phenomenologies, even in the case of oscillations. The presence of CO almost certainly indicates simultaneous generation of hydrogen during the reaction. This has incredible potential, as both of these products can potentially be used by fuel cells. Attempts to produce H₂ through combustion of fuel-rich mixtures have been previously attempted [63], with modest results. The scale here is much smaller than in the previous experiments, which is less suitable for large-scale hydrogen production. However, it may be ideal for point-of-use generation of small quantities of these gases for use in a micropower system. A rich stationary flame front like the one studied here could be coupled with a solid oxide fuel cell (SOFC). The rich flame front would provide in-system generation of CO and H₂ for the fuel cell to consume, as well as providing the heat needed to keep the fuel cell at operating temperature.

Chapter 6

Computational Studies

Nomenclature for this chapter is described in Appendix A.

6.1 Initial Version of the Model

The first version of the model employs fixed temperature distributions for the tube wall that remain constant throughout the simulation, and does not model temperature changes experienced by the fluid as it flows through the tube.

6.1.1 Base Case

Figure 6.1 shows the phenomenology map for the standard system depicted in Fig. 2.4. Six main regions are apparent. The reader is encouraged to compare with the phenomenological regimes of Fig. 3.1. Such a comparison will be discussed in detail below.

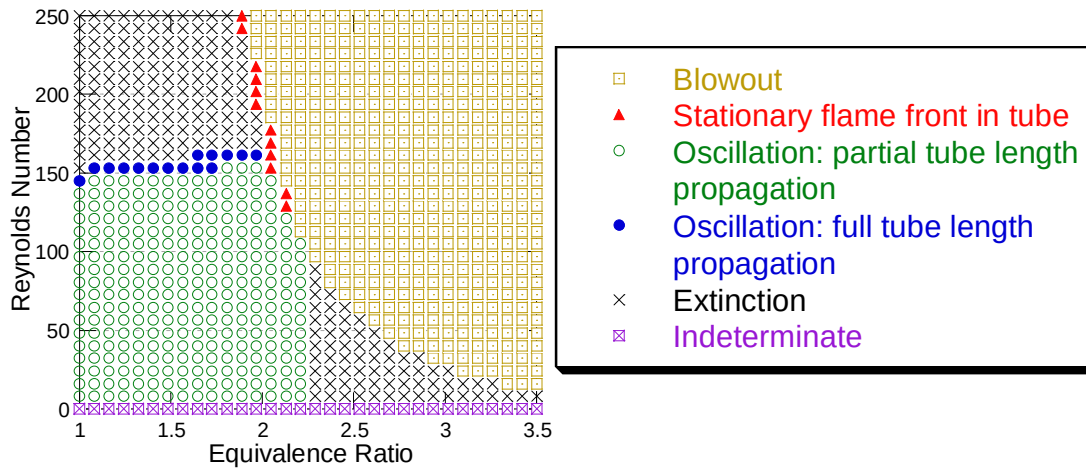


Figure 6.1. Phenomenology plot for a 70 mm length, 4 mm diameter tube with a wall temperature decreasing linearly from 1000 K at the exit to 300 K at the inlet. “Indeterminate” phenomenology occurs for simulations that do not reach a steady or periodic state within the time step limit.

“Blowout” region (upper right): In this region, the flow speed exceeds the flame speed, so the flame front cannot propagate into the tube. Instead it will anchor at the tube exit in a blowout configuration. This is a region of high equivalence ratios and Reynolds numbers. Regardless of the equivalence ratio, blowout will occur when the flow speed exceeds the flame speed. However, as equivalence ratio decreases and approaches unity, the flame speed increases substantially, so higher Reynolds numbers are required for this to occur.

First “Extinction” region (lower right): In this region the Reynolds number is too low to produce blowout, but the equivalence ratio is very rich and so the heat generation at the flame front is relatively low. Propagation into the tube is possible due to the low flow speed, but once inside the flame cannot remain stable even while in contact with the hottest parts of the tube. As a result, the flame front extinguishes. Re-ignition is not possible, because even though the wall temperature near the tube exit (where extinction occurs) is higher than the ignition temperature, the heat losses to the wall are still too high for the rich flames of this region to stabilize.

“Oscillation: partial tube length propagation” region (lower left): When the equivalence ratio drops below a value of ~ 2.25 , the heat generation of the flame becomes sufficient to overcome the heat losses to the walls, at least in the warmest portion of the tube. Below this threshold value, stable propagation into the tube is possible. This allows the establishment of oscillations that propagate over at least part of the length of the tube. The exact range of the oscillation is determined by the relative locations at which extinction and re-ignition occur.

“Oscillation: full tube length propagation” region (middle left): In this region the equivalence ratio is low enough that the flame front can remain stable when in contact with walls at 300 K, the lowest temperature in the tube. This allows the flame front to propagate down the entire length of the tube, causing extinction to occur at the tube base. The Reynolds number is at

a critical value such that reactant gases traveling from the inlet to the exit will receive sufficient heat to ignite exactly as they reach the exit, causing re-ignition to occur there. This combination of an equivalence ratio below a threshold value and a Reynolds number at a critical value produces oscillations that propagate over the full length of the tube, extinguishing at the inlet and re-igniting at the exit.

Second “Extinction” region (upper left): In this region, the equivalence ratio is such that the initial flame front will propagate into the tube some distance and then extinguish. However, the Reynolds number is above even the critical value for which re-ignition occurs at the tube exit. As a result, the flow speed is high enough that the reactant gases reach the tube exit before there has been enough heat transfer to bring them to the ignition temperature, so they flow out of the tube without re-igniting.

“Stationary flame front in tube” region: When the equivalence ratio is low enough, the flame front is able to propagate some distance into the tube. As it does so, it comes into contact with cooler walls that cause heat loss and reduce the flame speed below its adiabatic value. If this non-adiabatic flame speed equals the flow speed, the flame front will remain stationary but stable, as seen in this region. As outlined in chapter 2, the flame speed can have a value in the range $f_{ad}e^{-1/2} \leq f \leq f_{ad}$, depending on the local heat losses, while still maintaining a stable flame. This flexibility allows a small range of Reynolds numbers to produce a stationary flame front for each equivalence ratio.

“Indeterminate” region (bottom row): This region has a Reynolds number of 0, indicating that there is no flow through the tube. In this case the flame will propagate some distance into the tube and extinguish, after which no movement will occur. The software is not able to properly identify this behavior, hence the “Indeterminate” label.

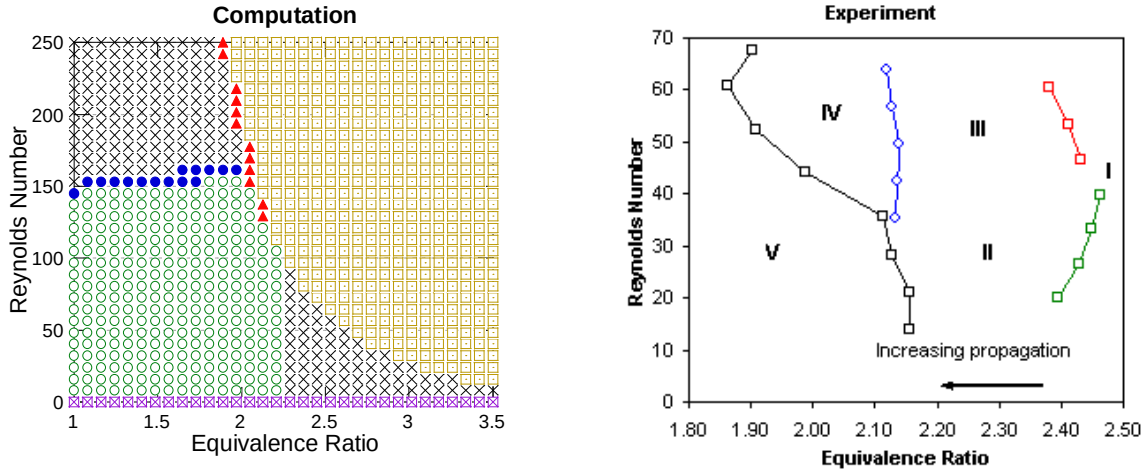


Figure 6.2. Computational and experimental maps of flame phenomenology for methane-oxygen combustion in a 70 mm long tube.

Figure 6.2 provides a side-by-side comparison of the computational and experimental results for this case. As it relates to the phenomenology observed experimentally and described in chapter 3, the computational case succeeds in producing all of the same behaviors as the experimental case: blowout, stationary flame fronts, oscillations with propagation ranges up to the entire length of the tube, and extinction. The basic trend of blowouts at richer equivalence ratios giving way to oscillations at lower equivalence ratios is also consistent between the two cases. Moreover, the depth of propagation for oscillations increases in the computations just as it does in the experiments, the difference being that the point of re-ignition is consistently at the tube exit for the experimental case. Though not identical, the threshold equivalence ratio below which oscillations occur is quite similar for both cases: ~ 2.25 in the computations and ~ 2.4 in the experiments. The threshold for oscillations covering the full tube is similarly close: ~ 2 for the computations and ~ 2.1 for the experiments.

The differences in the experimental and computational cases are also quite apparent. A major finding of our phenomenology studies was that the boundaries between behavioral regimes are almost entirely a function of equivalence ratio, and not of Reynolds number. This is not true

for the computations. During oscillations, re-ignition consistently occurred at the tube exit for the experimental tests, not within the tube as in some of the computations. The lower right extinction region of the computational map was not observed in experimental studies over similar ranges. At least some of the discrepancies can be attributed to the different wall temperature profiles in the experimental and computational cases. Here a linear wall temperature distribution has been imposed on the wall and remains constant throughout the simulation, whereas in the actual system the wall temperature will change dynamically due to thermal interaction with the flame front, and may ultimately be nonlinear, as noted in chapter 4.

6.1.2 Effects of Operational Parameters

6.1.2.1 Tube Diameter

In addition to the base case of a tube with diameter 4mm, phenomenology maps were also produced for tubes of 2 mm and 4 mm diameter. All other parameters were held constant. As the diameter increases, the coefficient of heat transfer decreases according to Eqn. 2.3, reducing the both the heat losses experienced by the flame front and the heat transfer to the reactant gases. Increasing the tube diameter, and thereby reducing the heat transfer, should make both extinction and re-ignition more difficult. Figure 6.3 shows the phenomenology maps for the three tested cases.

One notable effect of a tube diameter increase is that the boundary between the lower left oscillatory region and the lower right “Extinction” region shifts to the right, toward more rich equivalence ratios. This is expected, as the decreased heat losses in a wider tube make it possible for richer flames with less heat generation to propagate into the tube for at least some distance. It is also observed that as diameter increases, the Reynolds number for which

oscillations propagate over the full tube length decreases, and with it the boundary above which extinction will occur. Both of these effects are consistent with the expectations for diameter variations. Heat transfer causes the extinction of the propagating flame front, but is also required to re-ignite it. Changes in the geometry can alter the thermal situation and disrupt this delicate balance, preventing oscillations from occurring.

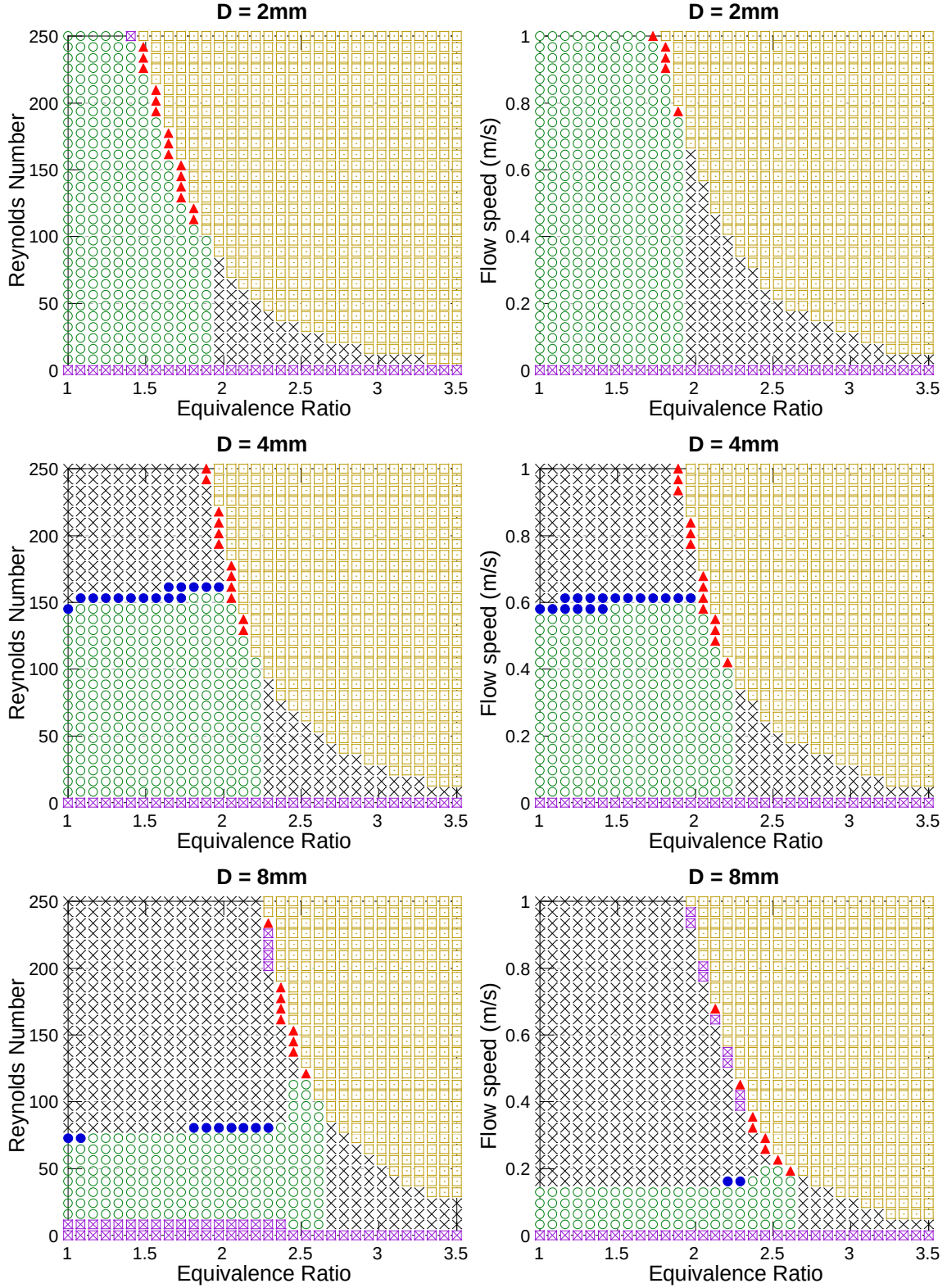


Figure 6.3. Phenomenology plots for 70 mm length tubes of three diameters, for equal Reynolds number and flow speed ranges. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

6.1.2.2 Tube Length

Three tube lengths were examined: 35mm, 70mm (the base case), and 140mm. Because the tube temperature is a function of axial position, changing the tube length requires adjusting the temperature profile. Our method was to implement a “fixed profile” for all three lengths: in each case the temperature would decrease from 1000 K at the tip to 300 K at the base. The results are shown in Fig. 6.4. As tube length increases, the Reynolds number for which oscillations covering the full length of the tube will occur increases substantially, and with it the boundary of the upper left extinction region. This is an effect of the shallower temperature gradient encountered in longer tubes. For reactant gases traveling at a given flow speed, a longer tube allows more time for heat transfer to occur, and the wall temperature changes more slowly as the reactant gases move along the tube. This allows reactant gases to reach the ignition temperature before

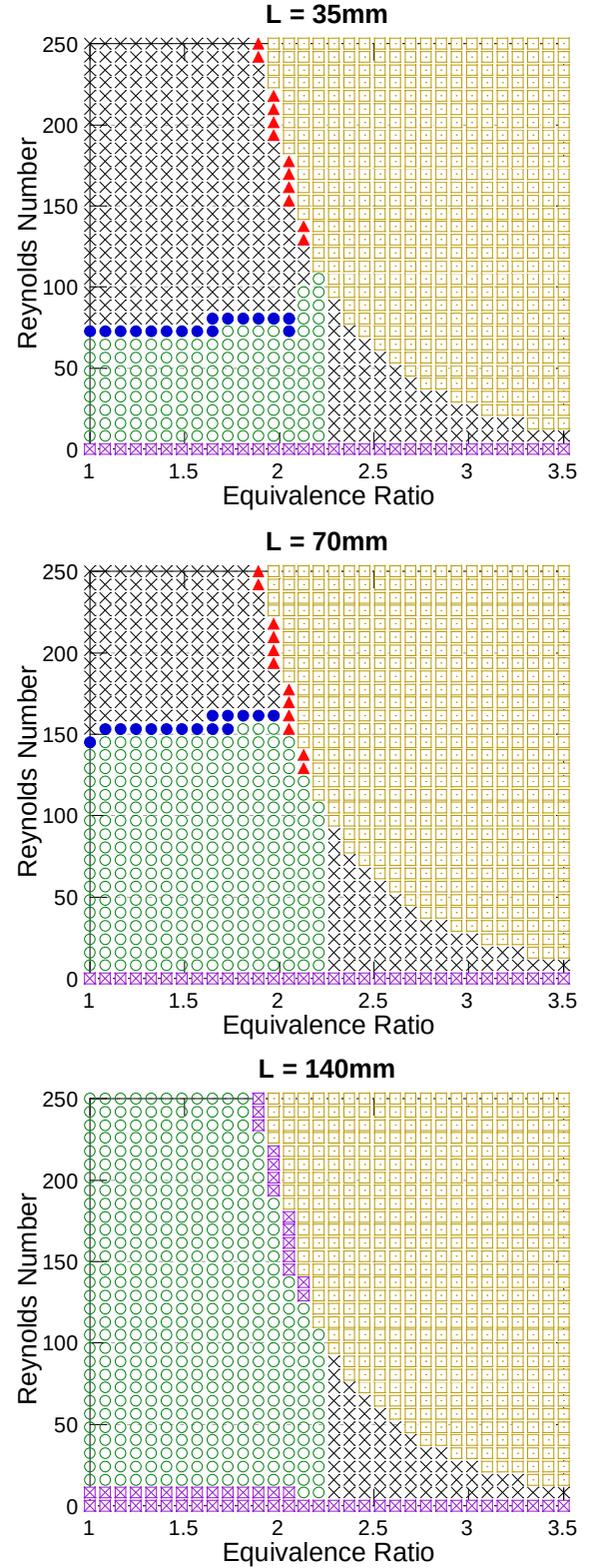


Figure 6.4. Phenomenology plots for 4 mm diameter tubes of three lengths. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

exiting the tube that might not have time to do so in a shorter tube with a steeper temperature gradient. The results demonstrate the importance of the wall temperature gradient.

6.1.2.3 Wall Temperature Profile

For our final experiment with the initial version of the model, we left the tube geometry in the default setup of 70 mm length, 4 mm diameter, and varied the wall temperature profile. In addition to the base case, three additional configurations were examined. For one, the temperature of the tip was the same as the base case, 1000 K, but temperature gradient was twice as steep, decreasing at a rate of 20 K/mm until a minimum temperature of 300 K was reached after 35 mm, and maintaining a temperature of 300 K for the lower half of the tube. A second case used an elevated peak temperature of 1700 K, but kept the default temperature gradient, so the temperature decreased at a rate of 10 K/mm to a minimum of 1000 K at the base. The third case used both the elevated tip temperature of 1700 K and the higher temperature gradient of 20 K/mm, so that the temperature reached 300 K at the base. These temperature profiles are illustrated in Fig. 6.5.

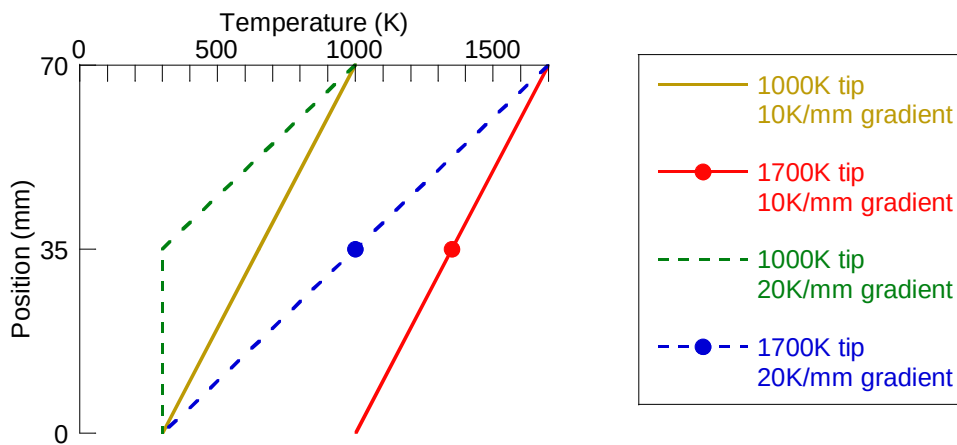


Figure 6.5. Illustration of the four wall temperature profiles used in the studies of this parameter.

Figure 6.6 shows the results of these simulations. The phenomenology map for the 1000 K tip temperature, 20 K/mm gradient is nearly identical to the 35 mm case in the tube length studies. This result is sensible, as the 1000 K tip, 20 K/mm gradient case is essentially the same as the 35 mm case with an extended region at 300 K. There will be no change in the conditions of a flame front or the reactant gases as they flow through this 300 K region, so it has no real effect on the system behavior. Here again it can be seen in the two 1000 K tip temperature plots (the bottom two panels) that a steeper gradient causes oscillations covering the full length of the tube to occur at lower Re, for the same reasons described in the tube length studies of section

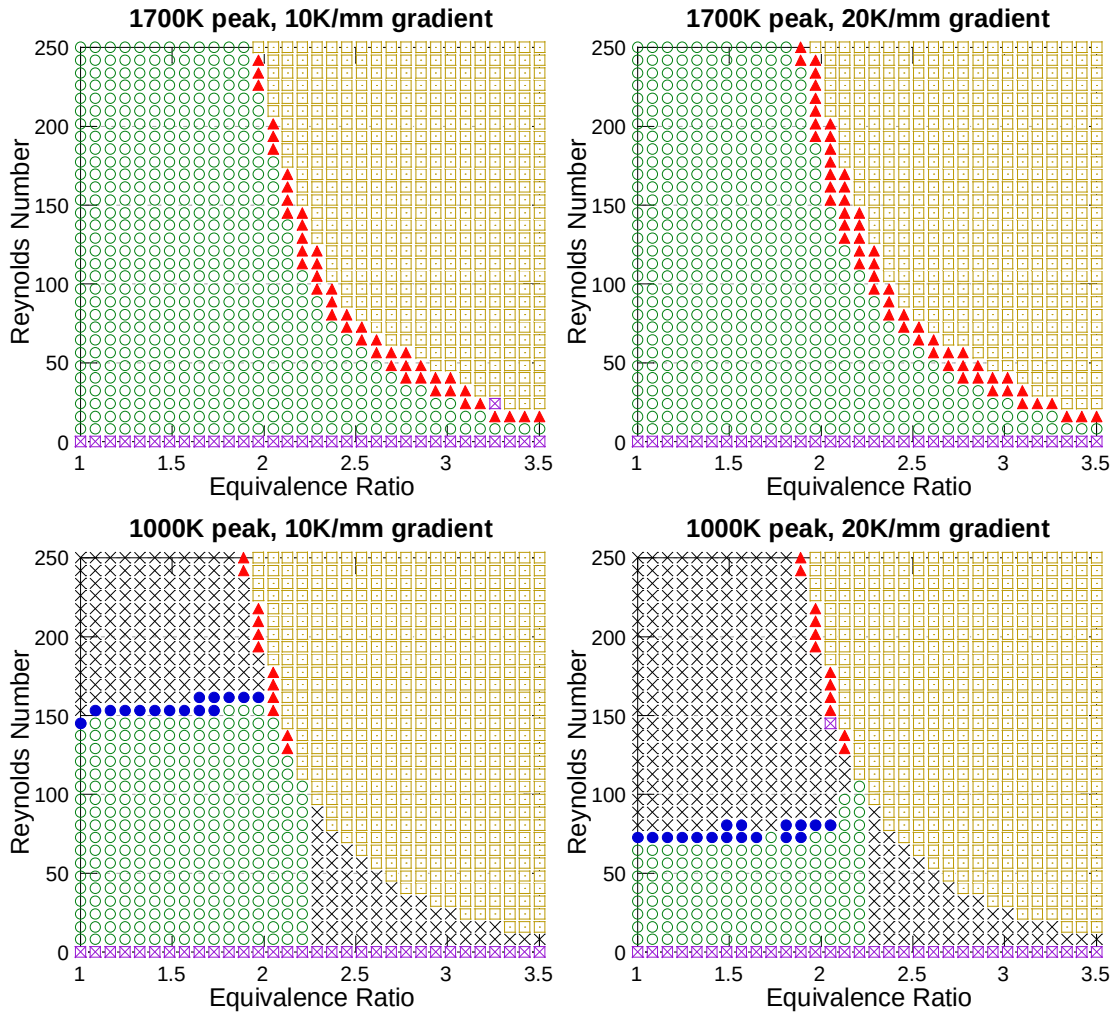


Figure 6.6. Phenomenology plots for 70 mm length, 4 mm diameter tubes with different exit temperatures and thermal gradients. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

6.1.2.2. Otherwise, things are the same. The 1700 K tip temperature plots (the top two panels), on the other hand, show a marked departure from the base case. The lower right “Extinction” region is eliminated completely. The tube is now hot enough that even extremely rich mixtures can propagate into it some distance and remain stable. Oscillations covering the full tube length and an extinction region for less rich mixtures are also eliminated from the plot, likely pushed outside the plot range. So much of the tube has a temperature above the ignition point, and the much hotter walls transfer heat to the reactant gases so quickly, that only an extremely high flow velocity could carry the reactant gases to the tube exit or beyond before ignition can occur. It should still be possible to produce oscillations covering the full tube length as well as terminal extinctions in the less rich regions, but only at Re well above the range of the plot. The configuration with the steeper gradient should produce oscillations over the full tube length at a lower Re than the configuration with the steeper gradient, as seen above.

6.2 Version 2 of the Model (Including Reactant Preheating)

As described in chapter 2, in the second version of the software, the effects of heat transfer on the fluid were modeled. As reactants flow through the tube from the inlet, they are warmed by the hotter tube wall and so have a higher temperature when they reach the flame front. The flame temperature and speed are calculated as a function of both equivalence ratio and unburned gas temperature, in order to capture the effects of the reactant preheating on the flame properties. The wall temperature distribution still remains fixed throughout the simulation, and has the same profile as in the previous version, depicted in Fig. 2.4.

6.2.1 Base Case

Figure 6.7 shows the phenomenology map for the base case as predicted by the second version of the model. The map from the first version of the model for this situation is included for reference. The basic layout of regimes for the new version is similar to that of the previous model, except for the presence of a region of unknown phenomenology in the upper middle position, in the area of $\phi = 2.5$ and $Re = 200$. In most cases, the software fails to identify the flame behavior exhibited here. However, inspection of the logs of flame position reveals that in this region, stationary flame fronts develop within the tube. The simulation code has difficulty producing (and subsequently identifying) true stationary flame fronts, because the finite element mesh of the wall creates discontinuities in the temperature distribution. Only a specific wall temperature will produce the heat loss necessary to depress the flame speed to the value of the flow speed, and that specific value may not exist in the mesh elements. As a result, this type of flame generally bounces back and forth between two adjacent elements with temperatures on either side of the value that would produce a true stationary flame, hence the “Indeterminate” characterization.

Comparison of the two cases depicted in Fig. 6.7 reveals two notable improvements when reactant preheating is considered. First, including preheating eliminates the lower right extinction region of Fig. 6.1, which is not observed in the experiments. Without preheating, extinction occurred in this region because the low flow speeds allowed the high equivalence ratio flames to propagate into the tube. Extinction occurred immediately since the low energy flames could not sustain contact with even the warmer walls of the tube, and there was not sufficient time for the reactants to reach the ignition temperature before exiting. Accounting for reactant preheating increases the effective energy content of these flames, allowing them to remain stable despite greater heat losses. Though the new model also

predicts the behavior of these flames imperfectly – blown out flames are observed experimentally in this region, rather than oscillations – the prediction of a stable flame rather than extinction is closer to the actual behavior.

The second improvement that results from consideration of reactant preheating is an expansion of the “stationary flame front” region. The previous model produced only a very

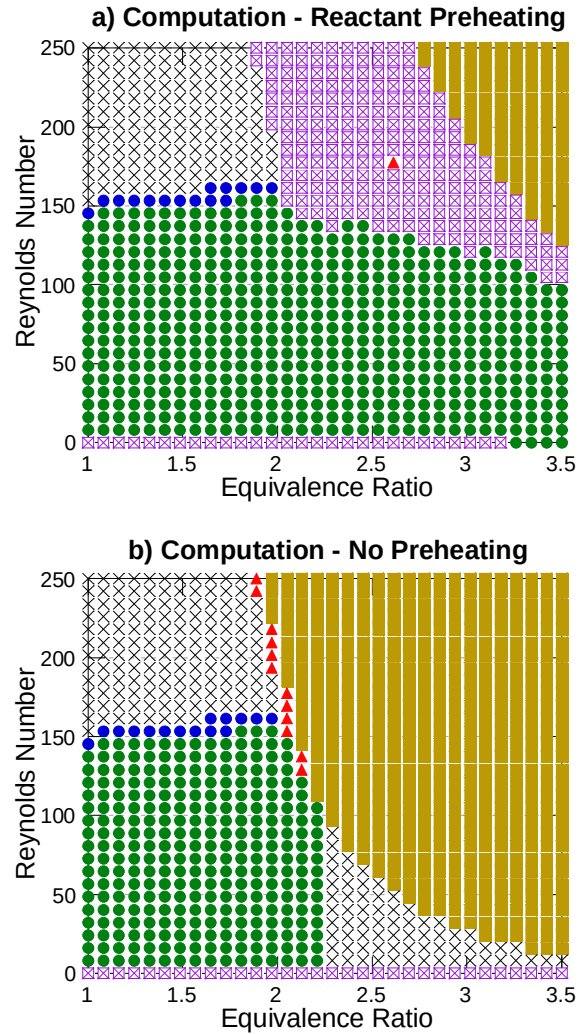


Figure 6.7. Phenomenology plots for a 70 mm length, 4mm inner diameter tube with a wall temperature decreasing linearly from 1000 K at the exit to 300 K at the inlet. a) Including reactant preheating, b) neglecting reactant preheating.. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

narrow band of stationary flame fronts, whereas experiments with quartz have shown a much larger stationary flame front region, as described in chapters 3, Fig. 3.2, and chapter 4, Fig. 4.2.a). The shape of the stationary flame front region with preheating in Fig. 6.7.a) is very similar to experimental observations. This suggests that reactant preheating may be a crucial factor in the establishment of stationary flame fronts in these ducts.

Despite the improvements, there remain some significant differences between the simulations and experiments. A major finding of the phenomenology studies was that the boundaries between behavioral regimes are almost entirely a function of equivalence ratio, and not of Reynolds number. In some cases this is still not true for the computations, most notably for oscillations with full tube length propagation, which occur only in a narrow Re band. Although the relative positions of the behavioral regimes in the simulations are accurate, the scale is not – many behaviors occur at very different Re than what was observed experimentally. During oscillations, re-ignition consistently occurred at the tube exit for the experimental tests, not within the tube as in some of the computations. The different wall temperature profiles in the experimental and computational cases are likely a factor in these discrepancies.

6.2.2 Simulations with Experimentally Measured Wall Temperature Distributions

We next inserted into the model as boundary conditions the experimental wall temperature distributions measured during the IR thermometry experiments of chapter 4. Our goal was to evaluate whether the model could produce the same steady-state behaviors as the experimental setup, when given an appropriate steady-state wall temperature distribution. For each experiment, the model was configured to use an equivalence ratio, flow speed, and wall

temperature distribution identical to what was measured in the experimental case. Figure 6.8 compares the experimentally-observed phenomenology with the predictions of the model.

The model fails to duplicate the experimental behavior in almost all cases. The only successes occur at high equivalence ratios, where the model produces blown out flames in agreement with the experiments. For lower equivalence ratios – including other conditions that should produce blown out flames – only extinction occurs.

Achieving blown out flames for high equivalence ratios is a modest success. As noted above, the default temperature profile used in the initial tests produced oscillations for these same equivalence ratios and Reynolds numbers. These tests demonstrate that with the proper thermal boundary conditions, the model has the potential to produce the correct flame behavior.

The failure to produce oscillating and stationary flame fronts, on the other hand, shows that problems remain in our implementation. The flame fronts succeeded in propagating into the tube in these cases, but failed to achieve the experimentally observed behavior. Cases that should have produced stationary flame fronts generally failed because the flame front did not

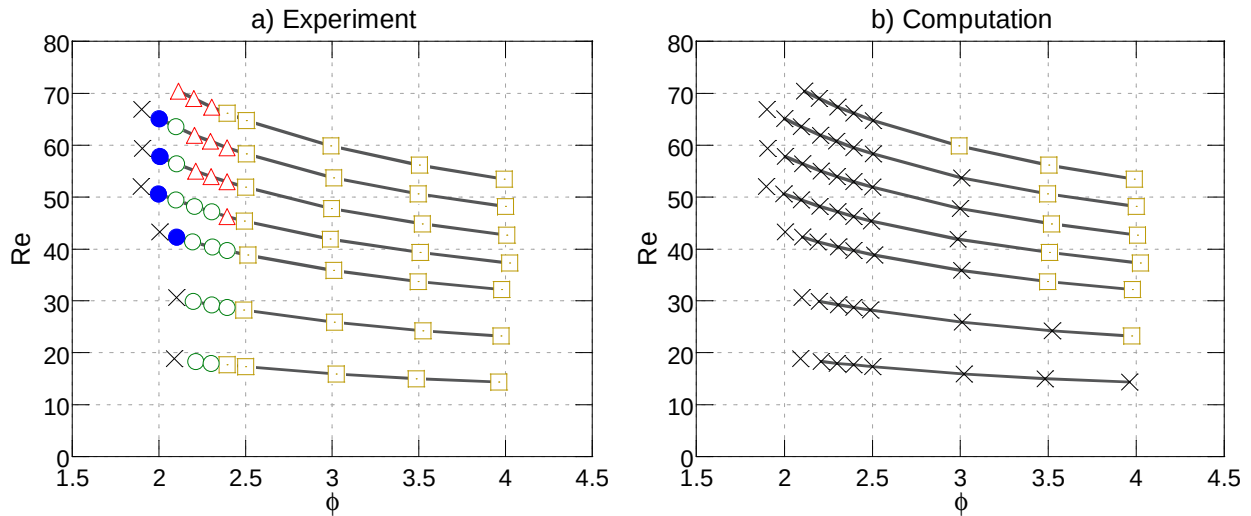


Figure 6.8. Experimental and computational plots of flame phenomenology. Lines connect points with equivalent fuel flow rates. The experimentally measured wall temperature distribution at each point is used as the wall temperature distribution in the simulation. Colors correspond to the behavioral regimes defined in Fig. 6.1.

come to rest. Instead, the flame front propagated past what should have been the resting depth, and extinguished upon reaching a cooler portion of the wall or the base of the tube. This suggests that the method of flame speed calculation within the model may be flawed.

Oscillations could not be established because the flame front failed to re-ignite after extinguishing. A possible explanation is that the ignition criterion used in the model is insufficient. As discussed in chapter 2, ignition of reactant gases becomes possible (but not certain) when they exceed the ignition temperature for methane. However, examination of the input wall temperature distributions reveals that the temperature of the tube exit exceeds the model's ignition temperature in only a few cases. This raises two possibilities. One, that the ignition criterion is not adequately sophisticated. In the physical system, re-ignition may not occur through the simple on-off mechanism in the code, but rather through a more gradual process. Catalytic action may also be a factor. The second possibility is that the diffusion flame that exists at the tube exit during oscillations does not actually extinguish at the same time as the moving flame front, as high-speed video recordings would suggest. Instead, it may merely change to a non-luminous state from which it can still help to ignite reactant gases. As discussed in chapter 5, this second possibility seems implausible. The more likely explanation is that the criteria for and process of re-ignition are more complex than what is included in the model.

In closure, the substantial discrepancies between modeling and experiments indicates that flame dynamics in small-scale hardware is a problem that has to be treated with Direct Numerical Simulation of the chemistry and the Fluid Mechanics involved in a manner that will probably challenge the current limits of reactive flow computation, as discussed in detail in the following section.

6.3 Version 3 of the Model (Including Solution of the Wall Temperature Distribution)

As described in chapter 2, the final version of the software added the ability to model the wall heat transfer and its effects on the wall temperature distribution. An initial wall temperature distribution is given to the wall at the beginning of simulation, but during execution the temperature of each point along the wall changes according to the heat transfer it experiences. This functionality is in addition to the fluid temperature modeling that was added in version 2.

6.3.1 Base Case

Initial simulations were performed for a tube with geometry similar the one used in the experiments, using 70 mm length, 4 mm ID, 6 mm OD, and the material properties of quartz. The results of the simulations for this case are shown in Fig 6.9. As discussed above, the software has difficulty identifying stationary flames and instead labels them as unknowns. As in previous versions of the software, the general layout of the regimes is similar to the experiments, with blowouts at high ϕ , extinction at low ϕ , and oscillating and stationary flame fronts in between. Also as in previous versions of the software, stationary flame fronts are observed at higher Re than oscillating flames. However, many discrepancies linger between the experimental and computational results. The boundaries between regimes are still a function of both ϕ and Re in the computations, whereas in the experiments ϕ was the primary determinant of behavior. The computational predictions of the boundaries are still very different from what was observed experimentally. Perhaps most problematic, the computational case is much more prone to extinction.

The results show that the initial wall conditions have some effect on the results, though not as much as expected from the experimental studies. Experimentally, producing stable flames for some of the lower equivalence ratios requires preheating, usually in the form of starting at a higher ϕ and gradually decreasing until the desired value is reached. In the computations, however, preheating is observed to have more of an effect at higher ϕ . With a preheated wall, a few more oscillating flames are observed at high ϕ and low Re, and two more stationary flames

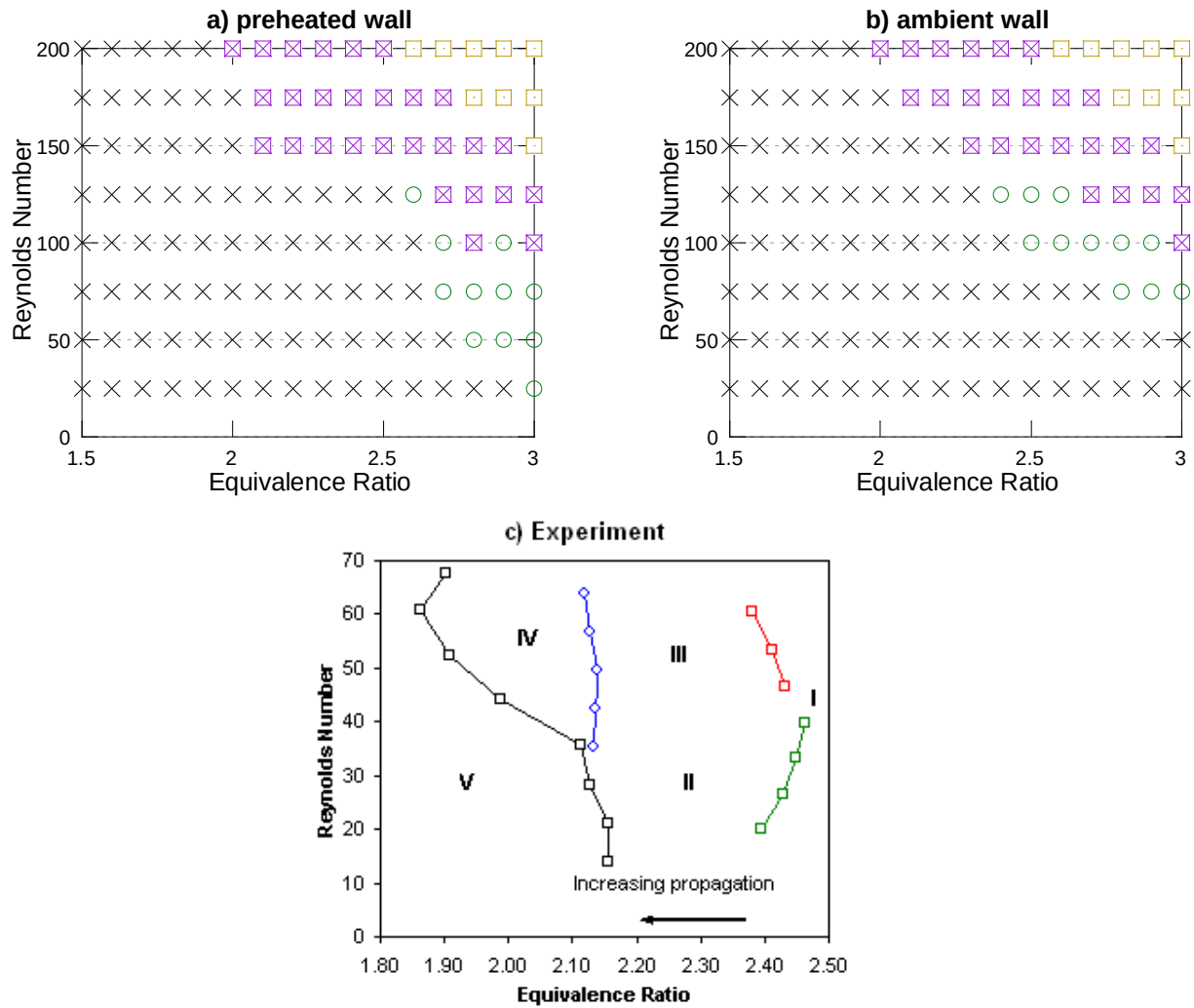


Figure 6.9. Phenomenology plots for a 70 mm length, 4 mm ID, 6 mm OD tube with dynamically varying wall temperature. The preheated wall case uses an initial wall temperature distribution that decreases linearly from 1000 K at the tip to 300 K at the base. The ambient wall case uses an initial wall temperature of 300 K throughout. A map of the experimental case is included for comparison. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

at lower ϕ . For the ambient wall, these cases produce extinction. The use of a preheated wall provides initial stability to the system in these cases, until such time as the flame heats the wall sufficiently to sustain itself. However, preheating does not always seem to improve stability. Around $\phi = 2.5$, the initially ambient wall produces several oscillating cases that go to extinction with a preheated wall. This may occur because a preheated wall allows an oscillating flame to propagate farther into the tube. In some cases this may cause the flame to overextend itself by distributing enthalpy over a larger area in a fashion that it is unable to sustain; confined to a smaller area by colder walls, the energy output of the flame may be able to heat the wall enough to remain stable.

It is a notable achievement that the code succeeded in modeling the major flame behaviors without needing to impose a fixed wall temperature distribution specifically designed to make them possible. Even with dynamic solution of the wall temperature and requiring the enthalpy content of the wall to be entirely supplied by the flame, the major flame behaviors were reproduced, including oscillations. This is not to be understated. However, the addition of wall temperature solution did not resolve all of the remaining issues with the model, contrary to previous speculations. This raises the question of what is lacking or inaccurate in the model as it stands. Three major areas in which the model may be lacking are heat transfer, chemistry, and fluid mechanics.

In an oscillating flame, the balance of various aspects of heat transfer is crucial. The duct must be warm enough to keep the flame stable, the flame must transfer enough energy to warm the duct and counter convective and radiative losses, the duct must retain enough energy to re-ignite the reactants after extinction, and so on. The proper balance allows the flame to give the duct a specific temperature distribution that allows it to propagate, extinguish, and re-ignite in a

sustainable fashion. Achieving the exact same behavior computationally requires correctly modeling each mode of heat transfer in order to achieve the same tenuous balance. A slight discrepancy in even one component of the heat transfer could disrupt this balance, producing different behavior. Although the model succeeds in producing all flame behaviors under some circumstances, it is unlikely that each aspect is modeled with exact accuracy. This is likely a factor in the discrepancies.

To limit the computational intensity, the model has always implemented instantaneous, one-step chemistry during simulations. To improve the accuracy, flame properties are calculated ahead of time with full chemistry using the GRI 3.0 mechanism. However, the properties are calculated for an equilibrium state. This undoubtedly results in less realistic property calculations for the flame, most likely in the form of overestimation of the flame temperature and speed. An overestimated flame temperature can result in greater heat transfer to the wall in a localized area, which in turn can produce larger heat losses than what should occur, compounding the error. An overestimated flame speed will cause the flame to propagate when it should not. The result of this will be less blown out flames and more extinctions, two of the exact issues in our computational results. Better modeling of the chemistry would improve the simulations, though the cost in computational intensity may be prohibitive.

Because of the evidence for a thermal driving mechanism underlying flame oscillations, the model was designed to use only limited fluid mechanics and a plug flow model. In a turbulent system this might be sufficient, but the flow in this system is laminar, as evidenced by the values of the Reynolds number. This has two ramifications. First, the flow speed for a fully developed laminar flow is not uniform, so calculating the propagation speed as the difference between the flame speed and the average flow speed may be invalid. Because of this, the model

may predict upstream propagation of the flame when the flame should actually remain stationary or propagate downstream. This may be part of the reason why the model predicts blown out flames only at high ϕ and Re. Second, the temperature distribution for a laminar flow through a warmer duct is not uniform, as in the plug flow model. The temperature near the walls will be greater than the mean temperature. In the model, the plug flow model requires the mean temperature of the fluid to reach the ignition temperature before re-ignition can occur. In the actual system, ignition could begin when only the fluid near the wall is at the ignition temperature, allowing re-ignition to occur with less energy input and a lower mean fluid temperature than in the model. Because of this, the model will predict extinction in cases that should produce re-ignition and oscillating flames. This issue likely contributes to the overly large extinction region in the computational results. Although fluid mechanics may not be the driving force behind oscillations, a more accurate model of fully developed laminar flow might help to bring the computational predictions more in line with experimental observations by enlarging the stable flame regimes. Such an undertaking is a non-trivial problem, and lies beyond the scope of this research. One approach would be to change the model of the fluid region from one-dimensional to two-dimensional, and to model viscous effects. However, this would drastically increase the computational intensity, almost certainly beyond acceptable levels. In addition, a two-dimensional fluid region would be difficult to reconcile with existing models for the chemistry and flame propagation, almost certainly requiring expanded models for these regions as well. As a less computationally intensive alternative, the one-dimensional design could be retained, and the average fluid speed and temperature already in the code could be used as starting values to derive appropriate parabolic distributions. However, this still leaves the question of how to calculate the net flame propagation speed with a non-uniform fluid velocity

distribution, and necessitates alterations to the method of computing the heat transfer between the wall, fluid, and flame.

6.3.2 Effects of Heat Transfer

The software was next used to perform a parametric study of several aspects of heat transfer in the system, including the heat loss from the propagating flame to the wall, the heat transfer from the diffusion flame to the tip of the tube, and the thermal properties of the duct wall.

6.3.2.1 Heat Loss from the Propagating Flame Front

In the base case, heat loss from the propagating flame front to the tube wall is modeled as occurring with a constant Nusselt number of 4.36, a value for laminar flow. For this study, simulations were conducted with a Nusselt number of 8.72, artificially doubling the heat loss from the flame front. This was expected to have two major effects. One was to increase the size of the blowout region, as greater heat losses would reduce the flame speed and make propagation into the tube more difficult. The other was to increase the extinction region, as greater heat losses should destabilize the flame. The resulting phenomenology maps for this situation as compared to the base case are presented in Fig. 6.10.

For the preheated case, both of the expected effects appear to have occurred. With higher heat losses, extinction happens at much higher ϕ . Blown out flames also occur in a few more instances with greater heat losses. One unexpected effect was an increase in stable oscillating cases at low Re . It was noted in the previous section that preheating in the base case causes extinction for some cases that produce stable oscillations with a non-preheated wall by

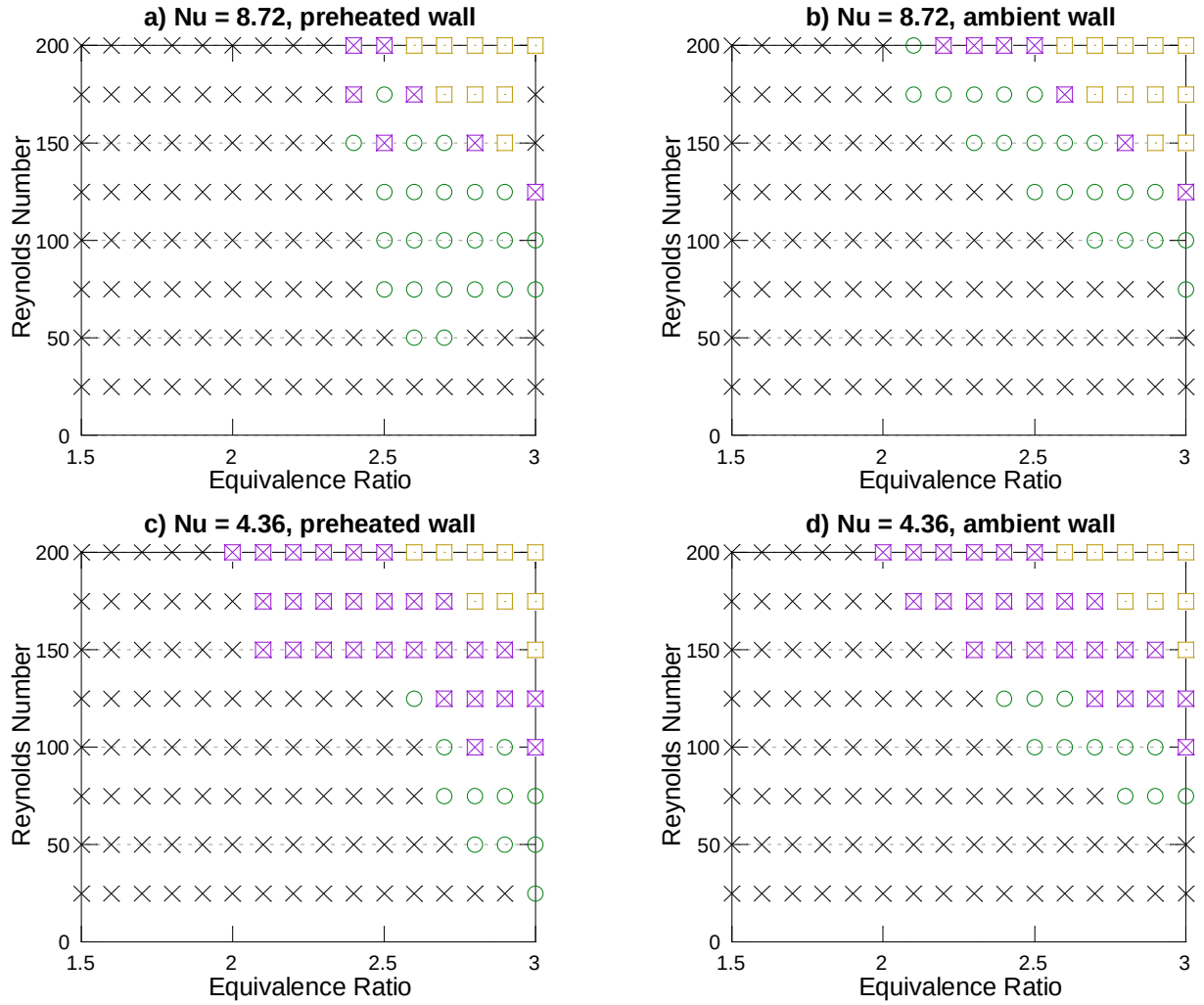


Figure 6.10. Phenomenology maps showing the effects of an elevated Nusselt number for the heat transfer from the propagating flame front. The lower row, panels c) and d), correspond to the base case depicted in Fig. 6.9. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

promoting excessive propagation into the tube. Larger heat losses seem to counteract this effect by limiting the penetration depth of flame fronts and keeping them nearer the exit, where they may be able to heat the wall to a sufficient temperature for stable oscillations to occur.

In the ambient wall case, both expected are also observed, but only to a small extent. With higher heat losses the blowout region is slightly enlarged and there are a few more extinction cases. The more noticeable effect is that with higher heat losses, cases that previously produced stationary flame fronts instead form oscillations. This is a result of the higher heat

losses causing extinction at higher temperatures, and illustrates the importance of heat losses in the formation of stationary and oscillating flames.

6.3.2.2 Heat Transfer from the Diffusion Flame

The diffusion flame is a crucial element in oscillatory phenomena, as it warms the tube exit to a temperature sufficient for re-ignition to occur. Higher heat transfer from the diffusion flame to the tube should improve oscillation stability by promoting re-ignition. To test this hypothesis, simulations were performed in which the heat transfer from the diffusion flame to the wall was artificially doubled in the computation. The phenomenology maps for these simulations are presented and compared to the base case in Fig. 6.11.

With preheating, higher heat transfer from the diffusion flame produces the expected improvement in stability. Many new oscillating cases are observed. The effects are mostly limited to low Re , however. Higher Re values produce stationary flames in both the base model and the one with enhanced heat transfer. These stationary flames will come to rest at locations with particular wall temperatures. With higher heat transfer from the diffusion flame, the wall temperature at each point will be increased by some amount, so the wall temperatures that will bring the flame front to rest will be lower in the tube. As such, the resting depth of the flame fronts will be lower with higher heat transfer, but the general phenomenology will be the same.

Without preheating, the higher heat transfer appears to have little effect on the flame phenomenology. The only notable change is that some blown out flames instead become stationary flame fronts resting just inside of the tube. This represents a slight shift in the boundary of the stationary flame front regime, and is an expected consequence of elevating the temperature near the exit. The same effect is seen with preheating as well. However, the

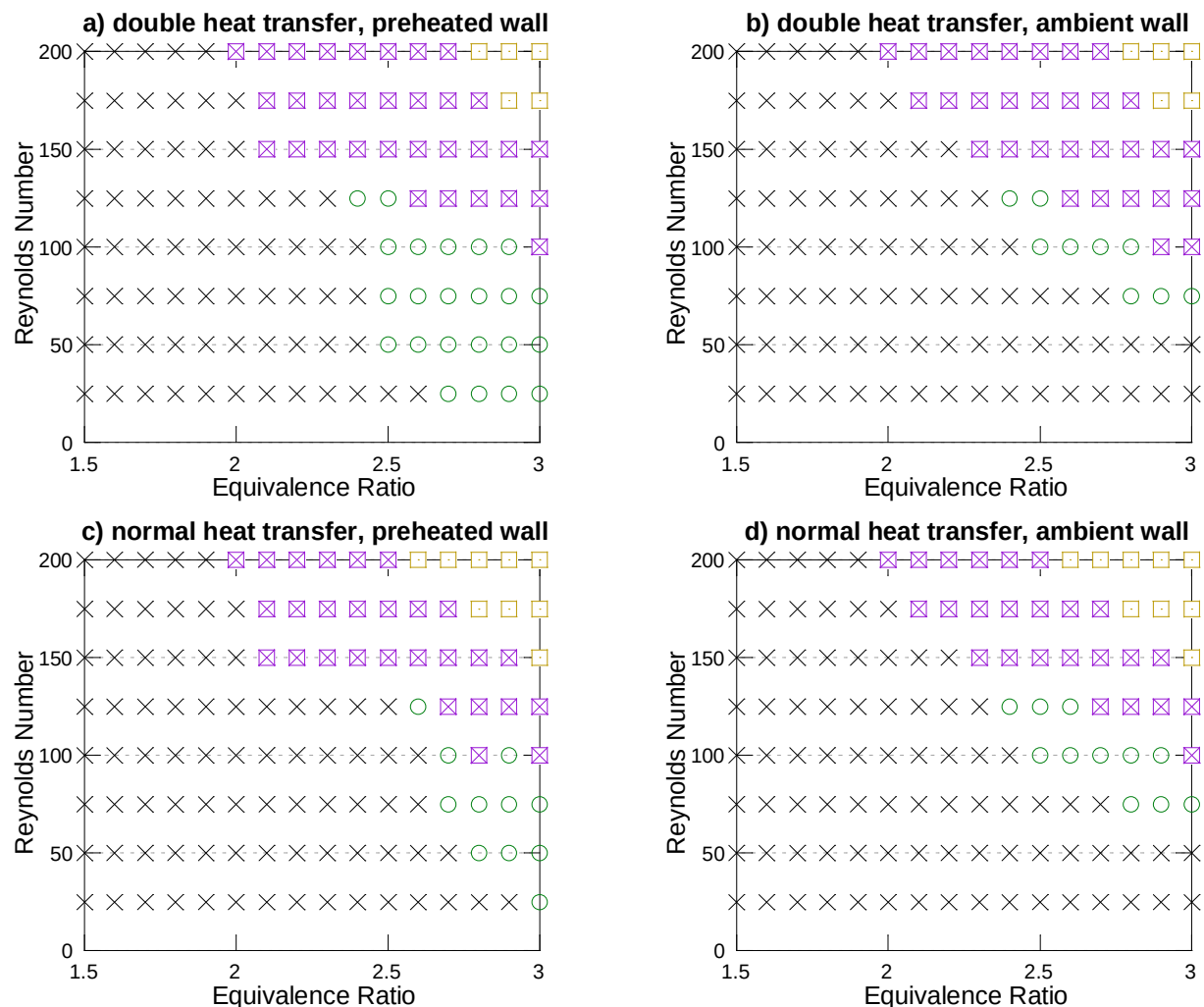


Figure 6.11. Phenomenology maps showing the effects of an elevated heat transfer from the diffusion flame to the tube tip. The lower row, panels c) and d), correspond to the base case depicted in Fig. 6.9. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

expected increase in the size of the oscillating regime was not observed without preheating. This may be because in the cases that might benefit from the greater heat transfer, extinction occurs before the diffusion flame has a chance to adequately warm the tube. Preheating the wall accelerates this warming process, allowing stable oscillations to develop.

6.3.2.3 Duct Material Properties

The importance of the duct material properties on the final temperature distribution and associated phenomenology was described in detail in chapters 3 and 4. In order to see if the model could predict similar effects, different material settings were used. To simulate the type of tube most commonly used in the experiments, the material properties of quartz were used in the base case (thermal conductivity 1.38 W/m-K, density 2220 kg/m³, specific heat 745 J/kg-K, thermal diffusivity 0.834e-6 m²/s). For this experiment, a steel tube was also simulated (thermal conductivity 15.1 W/m-K, density 8055 kg/m³, specific heat 480 J/kg-K, thermal diffusivity 3.91e-6 m²/s). The same emissivity of 0.75 was retained, so the primary difference is in the conductive heat transfer. The results of these simulations are presented in Fig. 6.12.

Similar patterns are observed in both the preheated and ambient cases. At high ϕ and low Re, fewer oscillations and more extinctions were observed in the steel tube compared to the quartz tube. This decrease in stability with higher thermal conductivity is the same effect that was observed experimentally. However, there is no difference in stability at higher Re, where stationary flame fronts occur. One other difference is in the boundary of the blown out flame regime. With the steel tube, this boundary shifts to the right, so some cases that produce blown out flames in the quartz tube instead produce stationary flame fronts in the steel tube. This is likely an effect of reactant preheating. In the steel tube, the higher thermal conductivity results in relatively shallow temperature gradient along the tube, whereas with quartz the elevated temperature regions are localized and the temperature gradients can be very steep. Because of this, in the steel tube reactants can be heated more gradually as they flow through the tube, potentially achieving a higher temperature by the exit. This boosts the flame speed, allowing propagation into the tube and the formation of a stationary flame front.

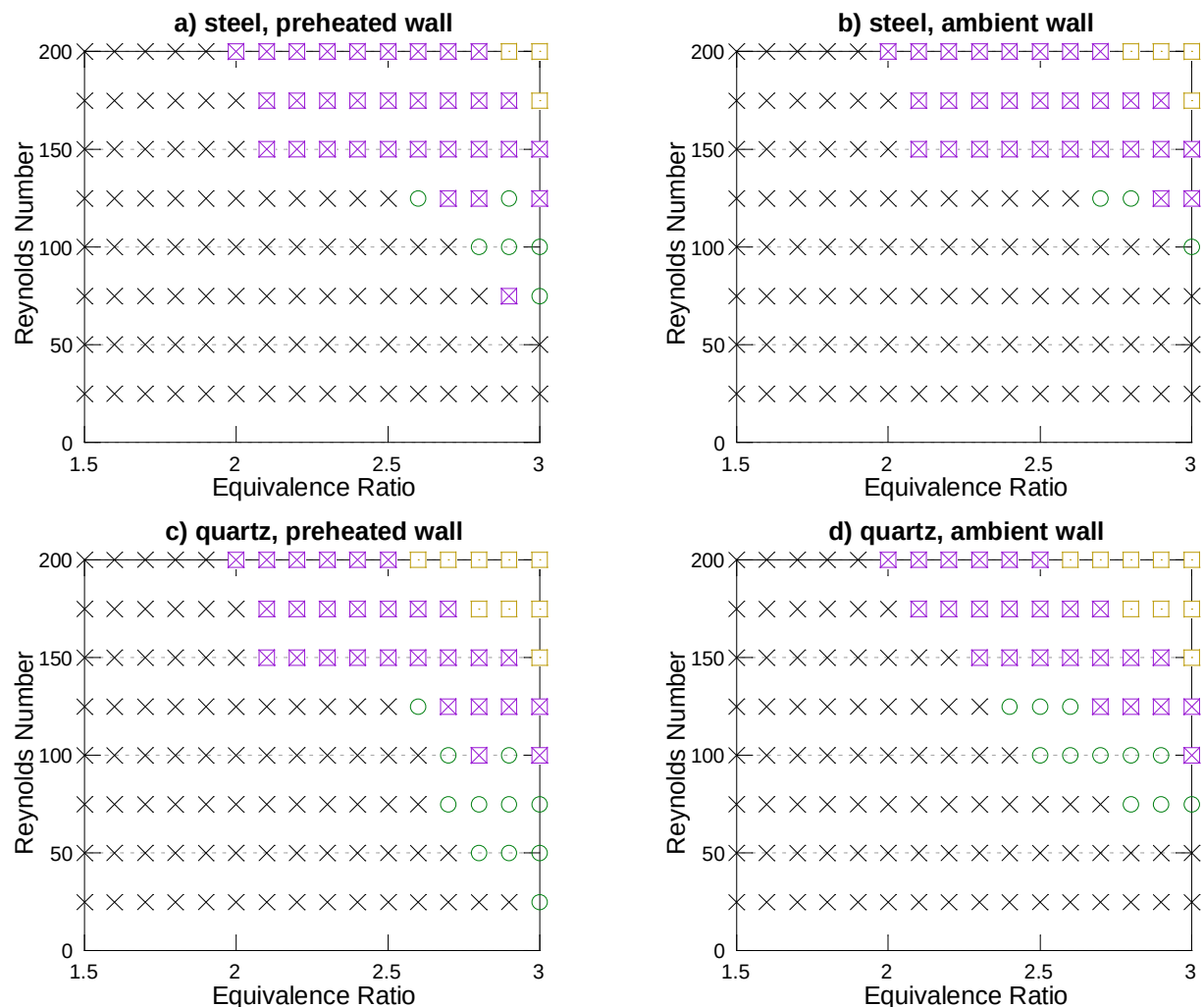


Figure 6.12. Phenomenology maps showing the effects of different material properties for the duct. The lower row, panels c) and d), correspond to the base case depicted in Fig. 6.9. Symbols correspond to the behavioral regimes defined in Fig. 6.1.

Performing simulations with the ducts having the material properties of quartz and steel also provides an opportunity to evaluate the computational predictions of the wall temperature distributions, and to compare them with the experimental results of chapter 4. As in the experiments, the computational wall temperature distributions follow varying patterns depending on the flame phenomenology. Representative results of the computations of the temperature distribution on the tube wall for each flame behavior (blowout, stationary flame front, oscillation) are presented in Fig. 6.13.

Several similarities are observed between the trends in the computational results and the experimental results of Figs. 4.3 and 4.4. The highest temperatures are observed at the duct exit, near the diffusion flame, and the temperature declines farther from the exit. Blown out flames primarily heat the exit, and produce little temperature elevation farther from the exit. Stationary flames produce two temperature peaks: one at the duct exit (near the diffusion flame), and a second one at the location of the stationary flame front. The experimentally observed effects of the duct material properties are also captured in the simulations. The quartz duct produces higher peak temperatures than the steel duct, but with temperature elevations that are localized and with steeper gradients along the wall in the temperature distribution. By contrast, the steel tube has lower peak temperatures, but the changes in temperature are more gradual so that temperature elevations are observed over an effectively larger area. Again, these are the expected results of the differences in thermal conductivity and thermal diffusivity for the two materials. The similarities between the computational and experimental results suggest that the general modeling of heat transfer is physically reasonable.

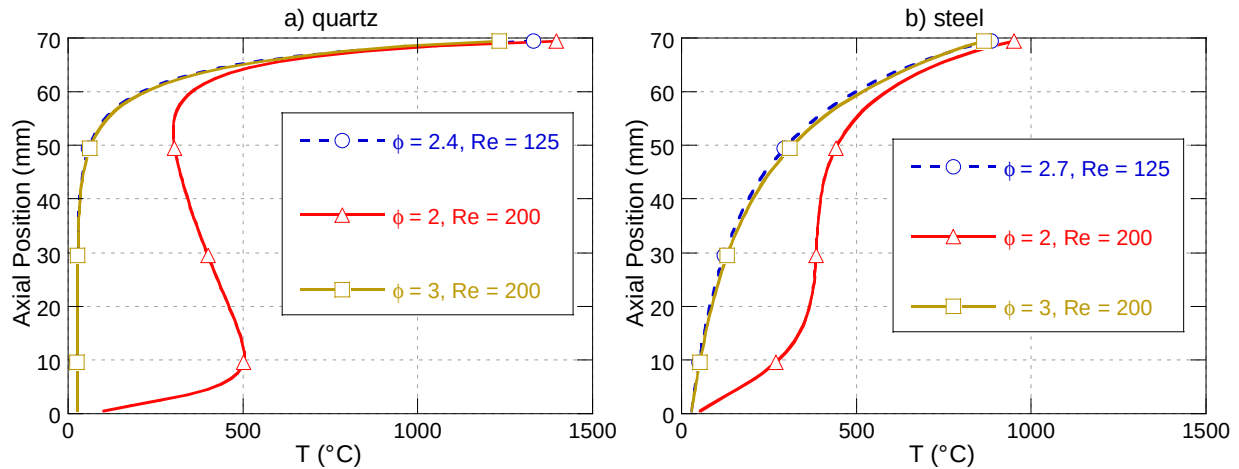


Figure 6.13. Sample computational wall temperature distributions showing the characteristic patterns for each flame behavior (blowout, stationary flame, oscillation). Symbols correspond to the behavioral regimes defined in Fig. 6.1. The blowout and oscillating flame curves very nearly overlap.

However, there are also some important differences. The temperatures near the duct exit are somewhat higher in the computational case, suggesting that either the temperature of the diffusion flame or the heat transfer from the diffusion flame to the tube may be over-estimated. During stationary flames, the larger temperature peak occurs at the in-tube flame front during experiments, but at the tube exit in the simulations. This is due in part to the reason above. In the computations, oscillations produce almost no discernible effect on the wall temperature distribution, which is drastically different from the experimental observations. Some of this may be due to the short propagation distances of the computational oscillating flames. However, it also suggests that the model under-estimates the energy supplied to the wall by the in-tube flame front. The fact that for stationary flame fronts the maximum temperature occurs at the tube exit in the computations, and not at the location of the stationary front as in the experimental observations, support this explanation. The results of 6.3.2.1 show that simply increasing the heat losses from the flame to the wall is unlikely to be sufficient to correct this deficiency. Proper modeling of the energy transfer from the in-tube flame to the wall through other modes of heat transfer, such as computing the two-dimensional flow field as well as including radiation, may be necessary. Resolving this issue may make it possible to reduce the computed heat transfer from the diffusion flame to the tube while at the same time accurately modeling the observed flame phenomenology. That could bring the exit temperatures in agreement with the experimental results.

Chapter 7

Summary, Conclusions, and Recommendations for Future Work

7.1 Flame Phenomenology Studies

Experimental maps of the flame phenomenology over a range of equivalence ratios and Reynolds numbers were determined for both methane-oxygen and propane oxygen flames. Experiments were conducted with multiple tube lengths, and tubes of different materials were tested in order to determine whether a low thermal conductivity in the tube is necessary for oscillations to occur.

Self-sustaining oscillations can be established for rich methane-oxygen and propane-oxygen mixtures in ducts of a few mm in diameter. This is an additional regime of operation that complicates the simple analysis of flame stabilization in large-scale ducts, which is based on the notion of the quenching diameter. The same basic behavioral regimes were seen for both fuels: extinction, stationary flame fronts, oscillations, and blown out flames. The boundaries between regimes are almost exclusively a function of equivalence ratio for methane flames, whereas for propane the Reynolds number is also influential and has a greater impact than equivalence ratio. Increasing the tube length from 35 to 70 mm, and by extension the area for thermal interaction, decreased the size of the stable oscillating regime for both methane and propane flames and made extinction more likely. This trend continued for methane as the tube length was increased from 35 to 70 mm, however for propane a slight increase in the extent of the stable regimes was observed. Increasing tube length also significantly affected the behavior of the propane flames,

allowing the establishment of apparently stationary flame fronts that were unseen in the shorter tubes. The results of these experiments support the idea that oscillatory phenomena in methane flames are thermally driven, however for propane flames fluid mechanics may play an important role as well. Tests with different materials demonstrated that only tubes of low thermal conductivity could sustain stable oscillations.

7.2 Infrared Thermometry Studies

Measurements of the wall temperature distribution of the duct were obtained via infrared thermometry. Quartz and steel tubes were used in order to examine the effects of the duct thermal properties on the resulting temperature distribution. The average temperature elevation, standard deviation in the temperature along the duct, and net added enthalpy were calculated in order to compare the magnitude and uniformity of the energy transfer to the duct for different flame behaviors.

Oscillating and stationary flame fronts within a tube were found to be significantly more effective in heating the duct walls than a blown out flame at the duct exit, for combustion in narrow ducts of circular cross-section. These combustion modes elevate the temperature of a larger portion of the duct walls to a higher temperature throughout. For a steel tube, an up to fourfold increase in the average wall temperature elevation was observed for oscillating and stationary flame fronts compared to blown out flames, while in a quartz tube the average wall temperature elevation was as much as eight times as great. Although in many instances higher average temperature elevations were observed for the quartz tube, the net energy content of the duct was much higher in the steel case due to the different material properties. For a fixed flow rate of fuel, decreasing the equivalence ratio increased the energy transfer to the wall by

increasing the penetration depth of the flame. Good uniformity of the wall temperature distribution was established for oscillating flames that propagate over the full length of the duct. In the quartz tube, this type of flame produced wall temperatures that were essentially uniform over the duct wall. This is particularly important in the context of micro-combustors that are to be coupled with direct energy conversion modules, such as thermoelectric generators and thermophotovoltaics, as these devices are often effective only in narrow temperature ranges.

For all combustion modes, the highest wall temperatures were observed at the exit of the tube. This high temperature region is the source of energy for re-ignition in oscillating flame fronts. Blown out flames were found to elevate the wall temperature only in the region near the tube exit. Oscillating and stationary flame fronts were capable of elevating the wall temperature farther from the tube exit.

Material properties, in particular the duct thermal conductivity, were found to have a major impact on the wall temperature profile and the flame phenomenology. In the case of fixed-position flames, the higher conductivity of the steel duct led to the development of relatively longer, but less warm, elevated temperature regions in the duct walls. The low thermal conductivity of the quartz duct caused more localized, higher temperature regions to develop with nonmoving flames, but also allowed the majority of the duct to take on a nearly uniform temperature in cases of oscillations with sufficient penetration depth. Stable oscillations could be established at lower equivalence ratios in the quartz tube, most likely because the more localized heating caused the tube exit to have a higher temperature, which facilitates re-ignition.

7.3 Emissions Studies

Samples of the exhaust gases were analyzed using Gas Chromatography and Mass Spectrometry. The percentage of fuel consumed in the flame and remaining in the exhaust were determined by comparing the methane signal for reacting and non-reacting mixtures. The major compounds produced by the rich flames within the duct were determined by analyzing samples taken from the partially burned mixture that exists between a stationary flame front in the duct and the diffusion flame at the duct exit.

Almost fuel consumption was found to occur for all flame phenomenologies, including oscillating flames. There were no significant differences in the consumption percentage as a result of flow rate or flame behavior. This was counter to expectations, as the extinction-ignition cycle of oscillations would appear to create gaps in combustion. A possible explanation is that unburned products do escape from the tube while the flame is extinguished, but that after re-ignition the diffusion flame front propagates downstream and consumes them. Carbon monoxide was found to be the major species produced by the rich flame front that exists within the tube during stationary flame fronts and oscillations. This suggests that hydrogen is another major product of the rich flame. The implication of this is that the novel flames under study could potentially be used to provide a fuel cell with both fuel and the heat required to reach its operating temperature.

7.4 Computational Studies

Three versions of a computational model were developed. Each employed simple, high-level mechanisms wherever possible, including instantaneous one-step chemistry, plug flow, and constant Nusselt number heat transfer. The first version used fixed temperatures for the tube

wall and the incoming fluid. In the second version, the heating of the incoming reactants by the hotter wall was modeled, as well as the elevations in flame temperature and speed that occur as a result of reactant preheating. The third version modeled the heat transfer within the wall and between the wall, flame, and surroundings, which allowed the wall temperature distribution to change dynamically during a simulation.

The computational model succeeded in capturing the major flame behaviors observed in rich methane-oxygen combustion in mesoscale tubes: blowout, stationary flame front within the duct, oscillations with varying propagation ranges, and extinction. With fixed wall and fluid temperatures, oscillations were observed to occur only for equivalence ratios below threshold values, determined by the local heat transfer. The threshold equivalence ratios for the simulations were close to those observed in past experimental studies, but slightly lower. This was suspected to be an indication of higher wall temperatures in the physical system. The tube diameter was found to influence oscillation stability through its effects on the rate of heat transfer between the flame front and wall. Increasing the tube diameter decreased losses from the flame front and improved oscillation stability, seen as an increase in the upper limit of equivalence ratios for which stable oscillations could be established. Simulations in which the tube length and wall temperature profile were varied demonstrated that the temperature gradient, and not simply the magnitude, has a major effect on the location of re-ignition during oscillations. A shallower temperature gradient along the tube length was found to improve oscillation stability, namely by increasing the range of Reynolds numbers for which stable oscillations could be established. Increasing the wall temperature near the tube exit enabled richer flames with lower heat generation to propagate into the tube and oscillate. This was seen as an increase in the range of equivalence ratios for which oscillations occurred.

Modeling the temperature variation in the reactants and the effects of reactant preheating on the flame properties produced two major improvements to the predictions of flame phenomenology. These were the elimination of an erroneous extinction region at high equivalence ratios and low Reynolds numbers, and greatly expanded regions of stationary flame fronts. The shape of the stationary flame front region is similar to what was observed experimentally, suggesting that reactant preheating may be a crucial factor in the development of this phenomenon. Simulations using experimentally measured wall temperature distributions failed to produce phenomenology that matched experimental observations in the vast majority of cases. Failure to reproduce oscillations was thought to be due to improper ignition mechanisms. Some success was achieved in duplicating blown out flames at high equivalence ratios.

Modeling the wall heat transfer and dynamically solving for the wall temperature distribution was the final step. When these functions were added, the model continued to capture all flame behaviors and correctly predict the relative locations of behavioral regimes. However, the boundaries between behavioral regimes were still not predicted correctly, and the model showed much greater tendency toward extinction. Although the simple, high-level mechanisms used throughout the model were capable of producing all of the flame behaviors, more detailed mechanisms may be necessary in order to correctly position the regime boundaries. Higher heat losses from the propagating flame front were associated with an increase in extinction, as well as more instances of blown out flames as opposed to flames that propagate into the tube. Higher heat transfer from the diffusion flame to the tip of the tube allowed the flame to more easily propagate into the tube and promoted re-ignition. This resulted in an increase in the size of the stationary flame front and oscillating flame regimes. Giving the tube the material properties of steel, as opposed to the default quartz properties, produced similar effects to what was observed

experimentally: in increase in the size of the extinction regime, and a less steep temperature gradient within the wall.

7.5 Summary

As they relate to the major objectives defined in chapter 1, our main findings were as follows:

- The investigations pursued here suggest that oscillations are primarily driven by thermal factors: extinction of the flame front occurs due to heat loss from the flame front to the colder walls farther from the exit, and re-ignition occurs as a result of heat transfer from the hotter walls near the exit to the reactant gases. A diffusion flame warms the wall near the exit, producing this temperature gradient. The studies conducted here do not suggest that fluid mechanics are involved in this process of oscillation, but these studies also cannot prove that fluid mechanics are unimportant, because hydrodynamics is not analyzed in a detailed manner.
- For methane-oxygen flames, the flame behavior is primarily a function of equivalence ratio. For propane-oxygen flames, Reynolds number is also a major factor. Oscillating flames are most stable in short ducts, and become more prone to extinction as the duct length increases. A duct with low thermal conductivity is necessary to produce stable oscillations, at least when no external heating is provided.
- During oscillations, a high temperature region exists in the tube wall near the exit that is capable of providing the heat needed for re-ignition. Oscillating flames are capable of producing a nearly uniform temperature throughout the duct.

- Analysis of the exhaust products indicates that oscillations do not have increased leakage of fuel or intermediate species compared to other flame behaviors. Fuel consumption percentages for oscillating flames and steady flames are comparable.
- A computational model using high-level mechanisms succeeded in capturing all experimentally observed flame behaviors, including oscillations. The relative positions of the different behavioral regimes were correctly predicted. However, more detailed mechanisms may be necessary in order to accurately predict the locations of the boundaries between behavioral regimes.

These findings suggest that oscillating flames are well suited for use in small-scale combustion driven energy systems. They are very stable at small scales, have the unique ability to produce nearly uniform duct temperature distributions, and are not less fuel-efficient or more polluting than other flames. In addition, the rich flame fronts within the tube appear to produce a CO-H₂ mixture that could be used by fuel cells.

7.6 Recommendations for Future Work

The studies described in this thesis have contributed to fundamental knowledge of the nature of oscillating flames. However, there is still much to learn, and some new questions have been raised by this work. The following are suggestions for further avenues of inquiry.

The primary focus of this thesis was on methane-oxygen mixtures. Investigations of the phenomenology revealed that propane-oxygen mixtures were also capable of producing self-sustaining oscillations. These were somewhat different than in the methane case; for example, while the methane phenomenology was primarily dependent on equivalence ratio, Reynolds

number was also an important factor in the propane case. Expanded studies of propane-oxygen flames, perhaps with experiments similar to those conducted here, would be enlightening. Since it is known that oscillations are not a phenomenon specific to methane flames, examination of other fuels might also be useful. Using hydrogen and butane would allow for study of gases with masses less than methane and greater than propane. Bearing in mind that practical meso- and micro-combustion systems are expected to use liquid fuels, tests with atomized liquid fuels such as methanol and ethanol would also be of value, first and foremost to see if oscillations could be established under such conditions.

Though the results of this thesis suggest that flame oscillations are a phenomenon driven by thermal factors, this does not mean that fluid mechanics will have no impact whatsoever. As noted in the discussion of the computational work, the nature of the velocity and temperature profiles for fully-developed laminar flow may be a crucial factor, perhaps influencing such things as whether a portion of the fluid can become hot enough to ignite, or whether a flame with a given speed will be able to propagate upstream. Nearer the inlet of the tube, the flow may not be fully developed at all, which will produce different velocity and temperature profiles that could influence the heat transfer and flame propagation. Moreover, the presence of oscillating or stationary flame fronts within the duct could have a dramatic effect on the flow. Studies of the flow patterns within the duct and near the exit would help to shed light on this aspect. Techniques such as particle image velocimetry might be suitable for this purpose.

There is significant room for development of the computational model. The results of these studies suggest that more sophisticated mechanisms for aspects such as the fluid mechanics and chemistry may be needed to produce phenomenology maps with more accurate regime

boundaries. These aspects can be added individually in order to determine their effects on the phenomenology.

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Appendix A

Nomenclature for Computational Work

A	Area	T_a	Activation temperature
C_p	Constant-pressure specific heat	T_{ad}	Adiabatic flame temperature
D_{tube}	Tube diameter	T_f	Flame temperature
E_a	Activation energy	T_{fluid}	Fluid temperature
f	Flame speed	T_u	Unburned gas temperature
f_{ad}	Adiabatic flame speed	T_{wall}	Wall temperature
h	Heat transfer coefficient	U_{in}	Inlet velocity
k	Thermal conductivity	V	Volume
L	Volumetric heat loss rate	y	Mole fraction
$\tilde{L}_v$	Non-dimensional aggregate volumetric heat loss rate	Y_u	Fraction of inlet mixture that is reacting fuel
Nu_D	Nusselt number	z	Axial position
q	Heat flux	ρ	Density
q_c	Heat of combustion	$\sim$	Denotes non-dimensional value
R_u	Universal gas constant	$-$	Denotes upstream (preheat zone)
t	Time	$+$	Denotes downstream (cooling zone)

Author's Biography

Christopher Evans was born in South Carolina on August 10, 1981, and lived in the state throughout his youth. He began his studies in mechanical engineering at Carnegie Mellon University in 1999. In 2003 he received his BS and went on to pursue graduate work at the University of California at Berkeley, where he completed his MS in 2005. The same year, he enrolled in the Department of Mechanical and Industrial Engineering at the University of Illinois at Urbana-Champaign to continue his graduate work.

He joined the combustion research group of Professor Dimitrios C. Kyritsis and began research into mesoscale combustion and oscillating flames. His work on this topic has led to the publication of 1 journal article and 4 conference proceedings. On several occasions he also served as a teaching assistant for the department's undergraduate fluid mechanics course.