Appendix A: Thermocouple Radiation Correction

In a high-temperature environment, such as the exit of the hot-air duct in the current counterflow arrangement, one major source of uncertainty in the measurement of ignition temperature is the correction for radiation, closely followed by the assurance that simplifying assumptions made about the nature of the flow are accurate. In the following, the various factors impacting the fidelity of the measured temperature to the gas temperature are considered.

The first important consideration is the temperature uniformity of the thermocouple bead, such that the entire bead may be considered to be at a single temperature. This assumption is critical so that spatial variations in temperature within the thermocouple need not be considered. This is accomplished by evaluating the Biot number, Bi:

$$\mathrm{Bi} = \frac{hL_C}{k_w} = \frac{\mathrm{Nu}\, k_g}{\beta k_w}, \tag{1}$$

where h is the convective heat transfer coefficient, $L_C = \frac{d_b}{\beta}$ is a characteristic length defined as the bead volume divided by the bead surface area, β is a geometric factor (β = 6 and 4 for a spherical bead and a cylindrical bead, respectively), d_b is the bead diameter, $\mathrm{Nu} = \frac{hd_b}{k_g}$ is the Nusselt number, and k_g and k_w are the gas and wire thermal conductivities, respectively. The Nusselt number is calculated from Eqn. (2) using the correlation for a cylinder developed by Collis and Williams [1], following the suggestion of Shaddix [2] for thermocouple configurations where the ratio between bead and wire diameters d_b/d_w is less than 3:

$$\mathrm{Nu} = (0.24 + 0.56\, \mathrm{Re}_b^{0.45}) \left(\frac{T_m}{T_g}\right)^{0.17}. \tag{2}$$

In Eqn. (2), Re_b is the Reynolds number around the thermocouple bead with d_b as the length scale and T_m refers to the arithmetic mean of the gas temperature (T_g) and the thermocouple bead temperature (T_b). The gas viscosity needed for Re_b in Eqn. (2) and the gas thermal conductivity needed for Eqn. (1) are calculated from the relations of Scadron and Warshawsky [3], similar to the analysis of Fotache et al. [4]:

$$\mu = \mu_0 \left(\frac{T_g}{T_0}\right)^{0.69} \qquad k_g = k_{g,0} \left(\frac{T_g}{T_0}\right)^{0.78}, \tag{3}$$

where μ_0 and $k_{g,0}$ refer to the gas viscosity and gas thermal conductivity at $T_0 = 300\,\mathrm{K}$. Assuming a larger-than-typical value for Nusselt number of 2.5, a chromel wire conductivity of k_w = 19.24 W/m-K, and typical gas conductivities in the range k_g = 0.06–0.08 W/m-K, Biot number is of the order 10^{-3} that is << 1, indicating that the bead temperature may indeed be assumed uniform.

Under the assumption of steady-state temperature, an energy balance around the thermocouple bead may be written as the sum of conductive (subscript cond), convective (subscript conv), radiative (subscript rad), and catalytic (subscript cat) heat gain/loss:

$$m_b c_{p,b} \frac{dT}{dt} = 0 = \dot{q}_{\mathrm{cond}} + \dot{q}_{\mathrm{conv}} + \dot{q}_{\mathrm{rad}} + \dot{q}_{\mathrm{cat}}, \tag{4}$$

where m_b and $c_{p,b}$ are the bead mass and the bead specific heat, respectively. Catalytic effects may be neglected *a priori* due to the choice of a K-type chromel/alumel thermocouple, whose materials can be considered non-reactive under the present conditions [4]. However, conduction through the thermocouple wires may or may not be negligible depending on the nature of the support structure and its proximity to the thermocouple bead. To address these issues, an analysis following that of Shaddix [2] is followed, wherein conduction losses may be neglected under the criterion that the length l_w of the lead wires extending from any support structure to the thermocouple bead should obey $\frac{l_w}{l_c} > 10$, where:

$$l_c = \sqrt{\alpha \tau_{\mathrm{conv}}} \tag{5}$$

$$\tau_{\mathrm{conv}} = \frac{\rho_w c_{p,w} d_w}{4h} \tag{6}$$

$$\alpha = \frac{k_w}{\rho_w C_{p,w}}. \tag{7}$$

Here, l_c is a critical wire length, α is the thermal diffusivity of the wire, and τ_{conv} is a characteristic convective time constant for the wire. The properties used in this calculation are those of chromel wire, and are given as wire density ρ_w = 8.73 g/cm^3, wire specific heat $C_{p,w}$ = 447.6 J/kg-K, and wire diameter d_w = 0.11 mm. Using Eqns. (5)–(7) and substituting a Nusselt number for the convective transfer coefficient h, the minimum length criterion may be re-expressed as:

$$l_w \geq 10\, l_c = 10\sqrt{\frac{{d_w}^2 k_w}{4\text{Nu}\, k_g}}. \tag{8}$$

This criterion results in minimum lengths ranging from 9–20 mm for flow conditions of practical interest. As a result, the length of bare thermocouple wire protruding from the support structure is set at 20 mm.

The above simplifications result in the reduction of Eqn. (4) to a balance between radiative and convective heat transfer, with the radiative transfer a complex function of the rates of transfer between the various surfaces contained within the pressure chamber. To simplify the analysis, the configuration factor method is used, with the geometry approximated by that shown in Fig. A1. It is worth noting that the co-flow "gap" (surface 3) is a simplification of the actual geometry (*cf.* Fig. 1 in main text) and models the region between the inner wall of the air duct to the outer wall of the co-flow duct as a flat surface. The radiative energy gain/loss from the bead can therefore be written as the sum of net radiative transfer between the thermocouple bead and its surroundings. The resulting equation, after invoking Kirchoff's Law to substitute emissivities for absorptivities and using the configuration factor reciprocity relation $A_1F_{1\text{-}2} = A_2F_{2\text{-}1}$, gives the air temperature as:

$$T_g = T_b + \frac{\varepsilon_b \sigma d_b}{\mathrm{Nu}\, k_g}(T_b^4 - \varepsilon_1 F_{b-1} T_1^4 - \varepsilon_2 F_{b-2} T_2^4 - \varepsilon_3 F_{b-3} T_3^4 - \varepsilon_\infty F_{b-\infty} T_\infty^4), \tag{9}$$

where σ is the Stefan–Boltzmann constant, ε is the emissivity, the subscripts g and b refer to the gas and bead, numbers 1–3 refer to each surface given in Fig. A1, and ∞ refers to the remainder of the interior surfaces of the pressure chamber, taken here as the emissivity properties of aluminum. The configuration factors needed for Eqn. (9) are taken from Siegel and Howell [5]:

$$F_{b-1} = \frac{1}{2}\left\{1 - \frac{1}{\left[1+\left(\frac{d_1}{2(z+H)}\right)^2\right]^{1/2}}\right\} \tag{10}$$

$$F_{b-2} = \frac{1}{2}\left\{\frac{1}{\left[1+\left(\frac{d_1}{2(z+H)}\right)^2\right]^{1/2}} - \frac{1}{\left[1+\left(\frac{d_1}{2z}\right)^2\right]^{1/2}}\right\} \tag{11}$$

$$F_{b-3} = \frac{1}{2}\left[\frac{1}{\left[1+\left(\frac{d_1}{2z}\right)^2\right]^{1/2}} - \frac{1}{\left[1+\left(\frac{d_2}{2z}\right)^2\right]^{1/2}}\right] \tag{12}$$

$$F_{b-\infty} = 1 - (F_{b-1} + F_{b-2} + F_{b-3}). \tag{13}$$

Referring to Eqn. (9), it is immediately evident that the chamber temperature T_∞ as well as the surface temperature of surfaces 1–3 are necessary. While T_∞ can be reasonably approximated to the fuel-side boundary temperature as the chamber is cooled to limit its temperature rise (*cf*. Fig. 1 in main text), the surface temperature of the quartz tubes (surfaces 2 and 3 in Fig. A1) would be difficult to accurately quantify. Noticing, however, that the configuration factors for surfaces 1 and 3 for a normal operating position of z = 1 mm are 0.013 and 0.026, respectively, their contributions to the radiation correction are negligible. In addition, as surface 2 is in direct contact with heated air, it can be expected to be of similar temperature, and thus the net contribution to the radiation correction is quite small; less than 5 K for typical ignition temperatures in the present study and reasonable estimates of wall temperature. Due to these factors, the net radiative transfer between surfaces 1–3 and the thermocouple bead can be neglected and Eqn. (9) may be simplified to:

$$T_g = T_b + \frac{\varepsilon_b \sigma d_b}{\mathrm{Nu}\, k_g}\left(T_b^4 - \varepsilon_\infty F_{b-\infty} T_\infty^4\right). \quad (14)$$

Equation (14) is used to correct for radiation for all experimental data presented in this work, with bead diameter d_b = 0.15 mm, Nu and k_g computed from Eqns. (2) and (3), respectively, and $\varepsilon_b(T)$ linearly extrapolated from the chromel emissivity data of Sasaki et al. [6]. The magnitude of this correction ranges from 25 K to 60 K for the conditions investigated in the present study, with larger corrections corresponding to higher ignition temperatures.

In closing this section, it is worth emphasizing the sensitivity of the temperature measurement to both the emissivity of the thermocouple bead and the Nusselt number. These quantities rely upon significant assumptions regarding the surface condition of the thermocouple bead and the flow around the junction, respectively, and as a result the authors have made a concerted effort to constrain these values as much as possible. Nonetheless, some uncertainties remain and are worth stating explicitly given their potential impact on the interpretation of both the present data as well as the data evolved from similar experimental systems. With regards to thermocouple emissivity, Shaddix [2] stated the problem succinctly:

> Unfortunately, experimental data on the emissivity of the most common high-temperature thermocouple wires are quite scarce. Compounding this difficulty is the fact that the emissivity of a given metal is strongly dependent on the surface characteristics, including roughness, oxidation layer, and any other coating of the metal, whether intentional or not. [2]

The effect of these uncertainties can be quite large; arbitrarily increasing the emissivity from typical values of ε_b = 0.17–0.19 to ε_b = 0.3 results in temperatures 20–35 K higher than presently stated. While no discernable thermocouple "ageing" – in terms of variability in ignition temperature measurements separated in time by several weeks – has been observed, and appropriately adjusted measurements using thermocouples of varying size have resulted in highly similar ignition temperature measurements, this merely indicates that the overall methodology is reasonably robust and that the thermocouple's condition is quite stable. As a result, it is very difficult to determine whether the emissivity value used is accurate, and what range of values would constitute "reasonable" degrees of uncertainty. Since any error estimate for emissivity would be purely conjecture, the issue of uncertainty in emissivity is neglected in the temperature uncertainty estimation in Appendix B.

Nusselt number correlations also present a significant problem. While the present study follows the suggestion from Shaddix [2] to use a cylindrical correlation, other authors have followed significantly different methods such as a constant Nu = 2 assumption [7] – essentially an implicit assumption of spherical geometry – or an average of the spherical and cylindrical predictions [4]. This disparity can have a significant impact on the magnitude of the radiation correction, as clearly demonstrated in Section 3.5 of the main text. To the extent possible, this uncertainty is taken into account in this work (*cf.* Appendix B) and is the primary source of systematic uncertainty in ignition temperature measurements. However, it should be understood that the error bars provided for ignition temperatures are based upon estimated uncertainties only within the context of a presumed cylindrical Nusselt number formulation (per Shaddix [2]), and do not consider the effect of varied junction geometry.

The combined effect of uncertainties in both thermocouple emissivity and Nusselt number are potentially significant when considering that the ignition data generated from this experiment is expected to guide model development. Certainly it becomes difficult to interpret the meaning of differences between experimental and numerical data when the estimated uncertainty in experimental values is exceedingly large. However, it is worth considering that even the Sarathy mechanism [8] – which exhibits the closest agreement with experimental ignition temperatures – over-predicts experimental ignition temperatures by a minimum of 80 K. This suggests that even in the worst-case event that Nusselt number is significantly smaller than anticipated while bead emissivity is simultaneously larger, the resulting increase in the magnitude of the radiation correction would still not fully account for the differences observed between the experiment and the model. As a result, the conclusions of the present study are independent of the details of the radiation correction.

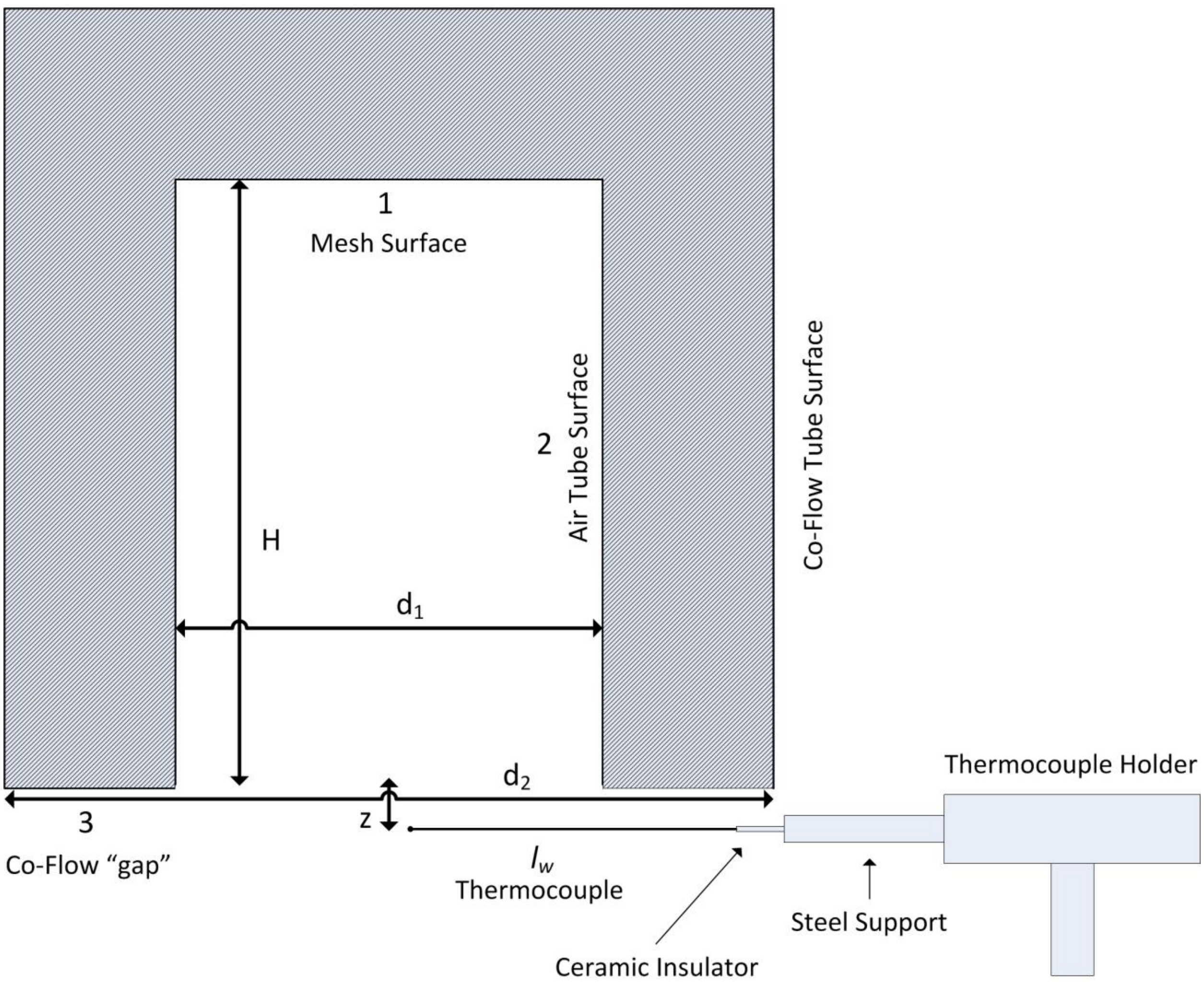

Figure A1: Simplified diagram of the air duct near the thermocouple bead, used for the determination of configuration factors for the radiation correction.

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