Supplementary Information

Computational Insights of Steady-State and Dynamic Joule-Heated Reactors

Arnav Mittal[1], Marianthi Ierapetritou[1,2], and Dionisios G. Vlachos[1,2,*]

[1] *Department of Chemical and Biomolecular Engineering, 150 Academy St., University of Delaware, Newark, DE 19716, United States*
[2] *Delaware Energy Institute, 221 Academy St., University of Delaware, Newark, DE 19716, United States*
*Corresponding author: vlachos@udel.edu

POWER DISSIPATED DUE TO RESISTIVE HEATING

Under the assumption of a material being isotropic

$$\frac{1}{2}\varepsilon_0\varepsilon_r E \cdot E = E \cdot \frac{\partial D}{\partial t}$$

By Poynting's theorem

$$E \cdot \frac{\partial D}{\partial t} = J \cdot E$$

Given the electromagnetic constitutive relation,

$$J = \sigma E$$

one gets

$$J \cdot E = \sigma E \cdot E$$

Integrating this over the volume of the element

$$\int_V \sigma E \cdot E = \sigma |E|^2 V_s$$

$$= \sigma \left(\frac{V}{L}\right)^2 V_s = \frac{\sigma V^2 L_w L_{th}}{L}$$

$$= \left(\frac{\sigma L_w L_{th}}{L}\right) V^2 = \frac{V^2}{R}$$

$$Grasshof\ number = Gr = \frac{g\beta(T_s - T_\infty)L^3}{\vartheta^2} \quad (S1)$$

$$Reynlods\ number = Re = \frac{\rho UL}{\mu} = Interfacial\ forces\ /\ Inertial\ forces \quad (S2)$$

$$Pr = \frac{C_p\mu}{k} = momentum\ diffusivity/thermal\ diffusivity \quad (S3)$$

$$Ra = Rayleigh\ number = Gr_x Pr = \frac{\rho\beta\Delta T x^3 g}{\mu\alpha} \quad (S4)$$

g is gravity, β is the expansion coefficient, L is the length of the element, ϑ is the kinetic viscosity, ρ is the density, U is the velocity, μ is the dynamic viscosity, C_p is the heat capacity, and k is the thermal conductivity.

Experimental condition	**Experimental value**	**CFD value**
Steady state temperature at 30 V	1922 K	1790.2 K
Steady state current – 12.4 V	3.8 A	4.075 A
Steady state current – 14 V	4.61 A	4.5 A
Steady state current – 15 V	5.25 A	5.3 A
Peak of pulsing temperature – 50 V	1473 K	1479.7 K
Minimum of pulsing temperature – 50 V	773 K	716.3 K

Table S1: Experimental validation of CFD model.

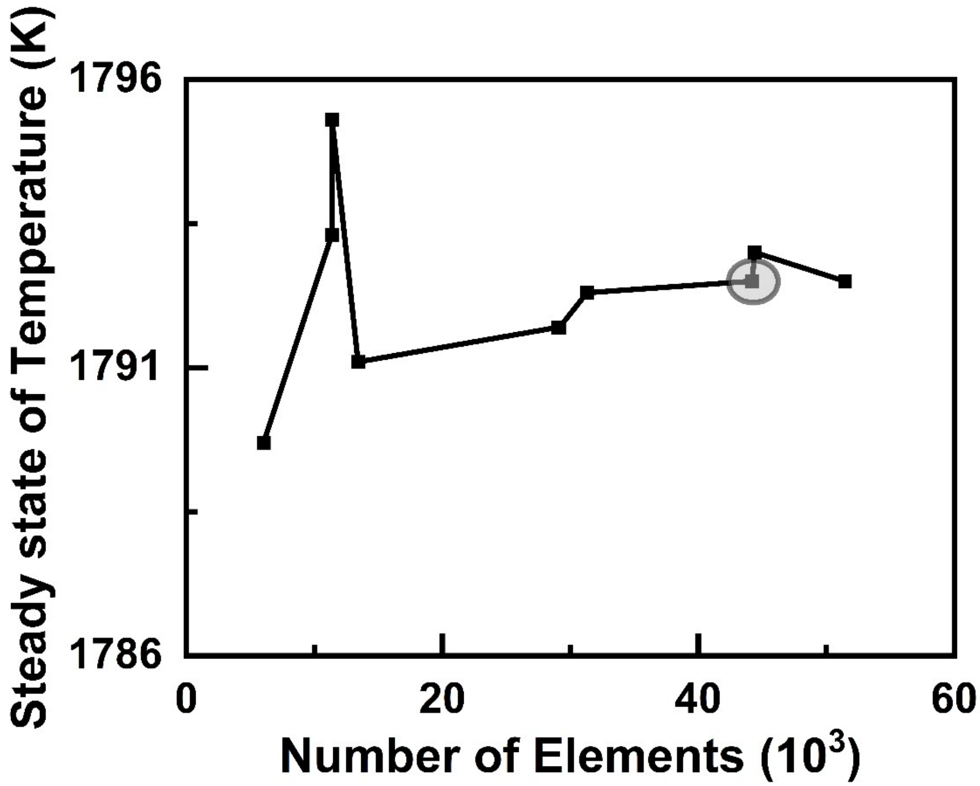

Figure S1: Mesh dependency study showing the steady state temperature for 30 V, with *He* flowing at 90 mL/min vs. the number of elements in the mesh. Highlighted mesh configuration used in this study.

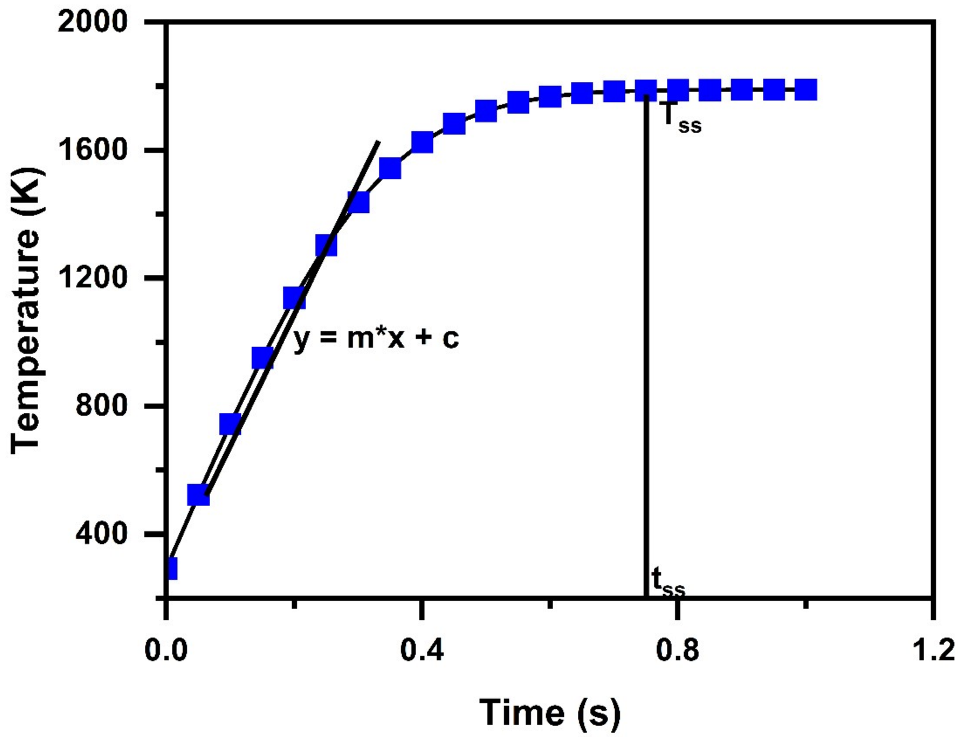

Figure S2: Temperature of the element vs. time. ($T_{e,ss}$ = temperature at steady state, t_{ss} = time to steady state, and m = R_h = temperature ramp rate). Voltage of 30 V with a *He* inlet flow of 90 mL/min.

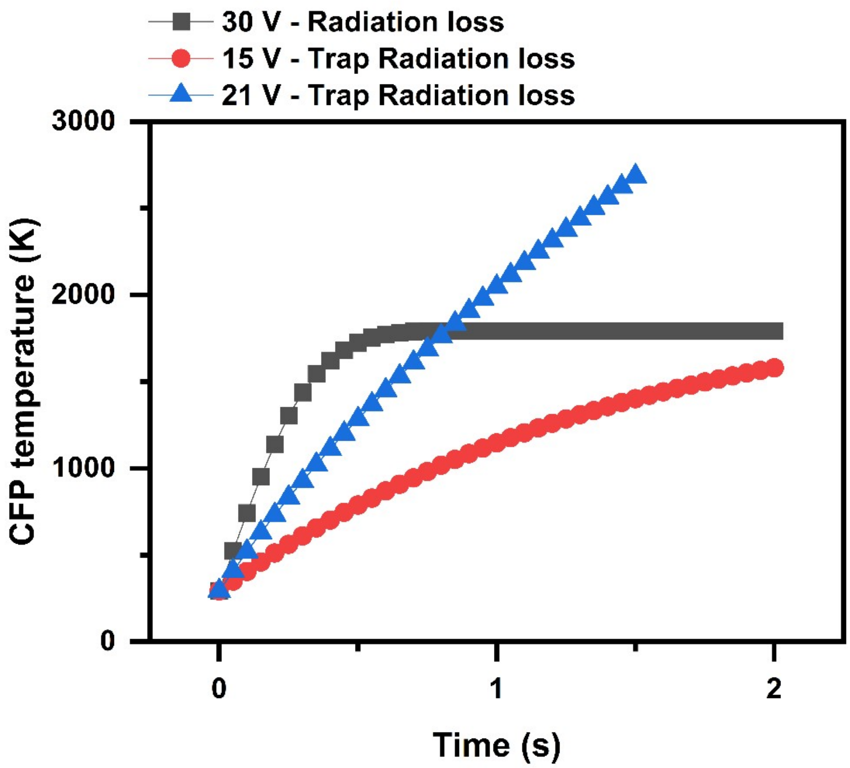

Figure S3: Impact of trapping radiation using a completely reflective surface over quartz tube on the temperature.

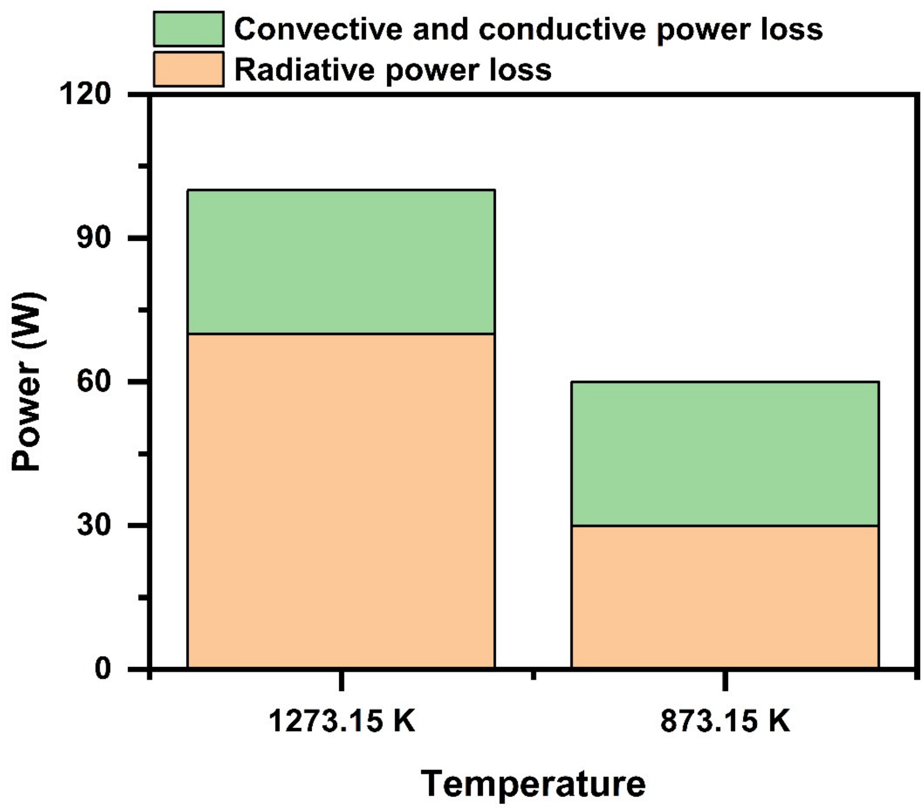

Figure S4: Power loss contributions when $(\bar{T}_{e,ss})$ = 1000 ℃ and 600 ℃, using N_2 at 90 mL/min.

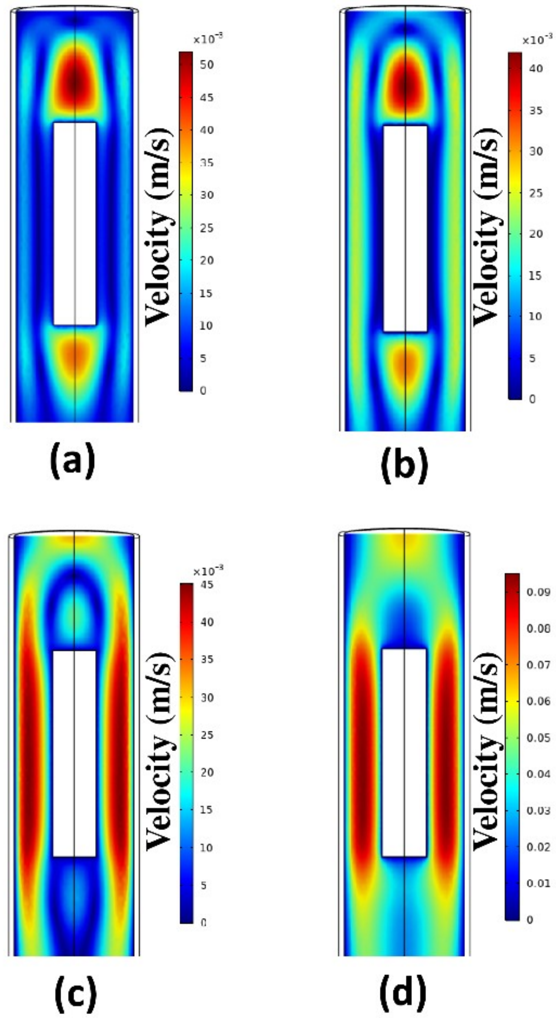

Figure S5: Flow maps of the magnitude of velocity parallel to the element at steady state for 30 V applied with *He* flowing at (a) 90 mL/min, (b) 180 mL/min, (c) 360 mL/min, and (d) 720 mL/min.

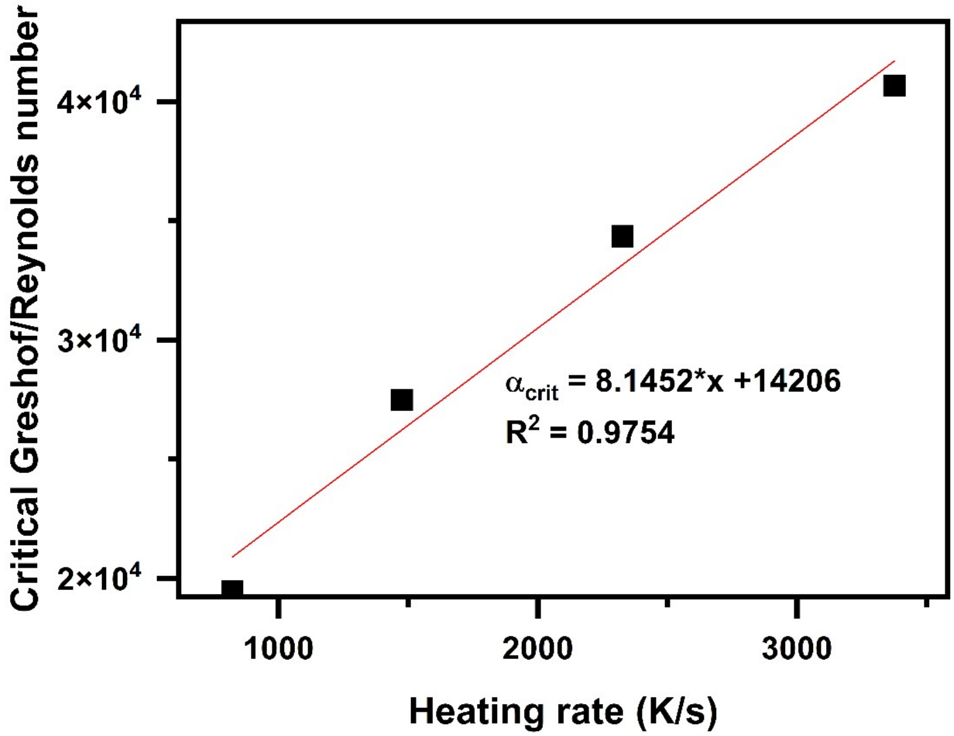

Figure S6: Variation of the critical value for reverse flow with heating rate.

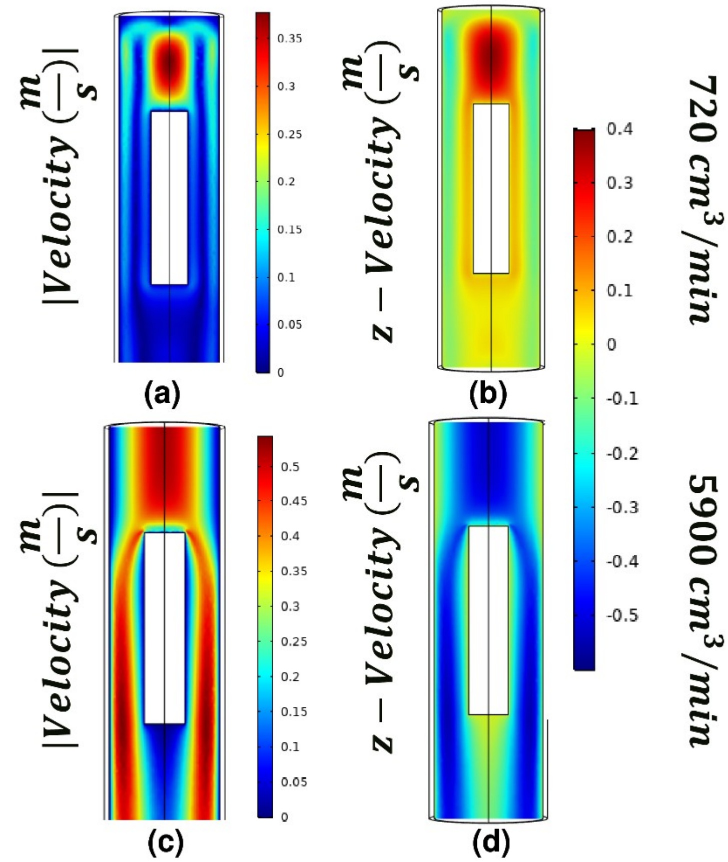

Figure S7: Flow maps of the magnitude and axial velocity for N_2 for 30 V applied to CFP. (a) Magnitude at 720 mL/min. (b) Axial velocity at 720 mL/min. (c) Magnitude at 5900 mL/min. (d) Axial component at 5900 mL/min.

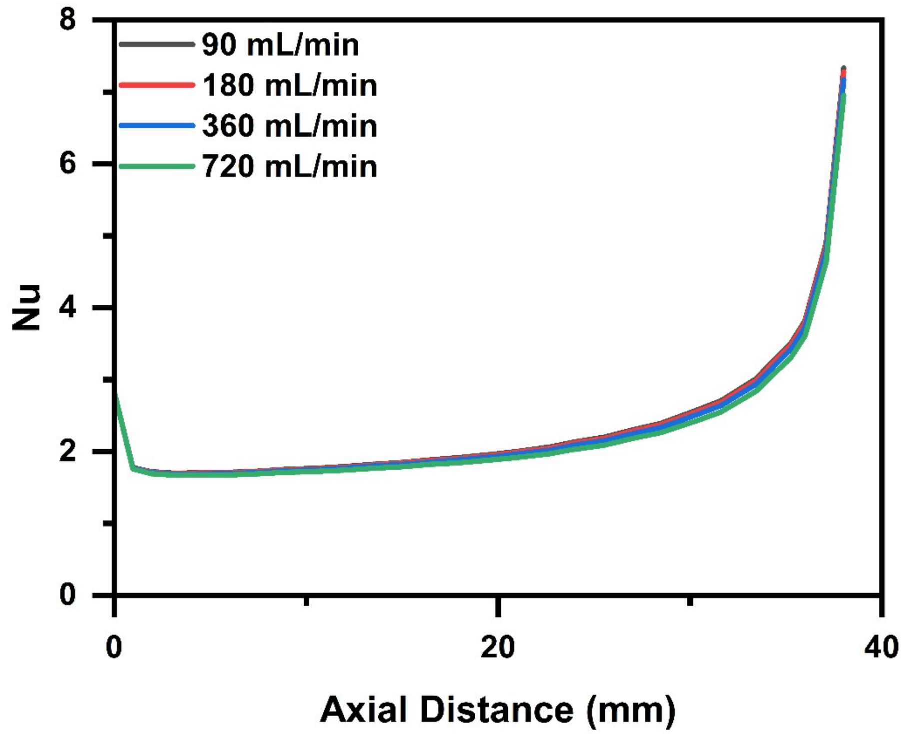

Figure S8: Impact of inlet flow rate at steady state for 30 V applied to CFP on the Nu number vs. distance from the leading edge of the heating element (assuming incompressibility).

HEATING TIME SCALE & STEADY STATE TEMPERATURE DERIVATION AT LOW TEMPERATURES (Temperature range I)

$$\frac{d\theta}{d\tau} = \alpha - \beta * (\theta - 1)$$

$$\frac{d\theta}{\alpha - \beta * (\theta - 1)} = d\tau$$

$$log\left(\frac{\alpha - \beta(\theta - 1)}{\alpha}\right) =- \beta\tau$$

$$t_H = \frac{t_0}{\beta}$$

For steady state: $\frac{d\theta}{d\tau} = 0$

$\alpha - \beta * (\theta - 1) = 0$

$\frac{\alpha}{\beta} + 1 = \theta_{ss}$

HIGH TEMPERATURES (Temperature range II-IV)

$$\frac{d\theta}{d\tau} = \alpha - \gamma * (\theta^4 - 1)$$

$$\frac{d\theta}{\alpha - \gamma * (\theta^4 - 1)} = d\tau$$

$$\frac{d\theta}{\alpha + \gamma - \gamma\theta^4} =$$

$$\frac{d\theta}{A - \gamma\theta^4} = \frac{0.5A^{-0.5}d\theta}{A^{0.5} + \gamma^{0.5}\theta^2} + \frac{0.25A^{-0.75}d\theta}{A^{0.25} + \gamma^{0.25}\theta} + \frac{0.25A^{-0.75}d\theta}{A^{0.25} - \gamma^{0.25}\theta}$$

$$= \frac{1}{4A^{0.75}\gamma^{0.25}}(log(A^{0.25} + \gamma^{0.25}\theta) - log(A^{0.25} + \gamma^{0.25}\theta) + tan^{-}(\frac{\gamma^{0.25}}{A^{0.25}}\theta))$$

$$A = \alpha + \gamma \gg \gamma, tan^{-}\left(\frac{\gamma^{0.25}}{A^{0.25}}\theta\right) \sim 0$$

Putting in limits, the RHS becomes

$$\tau(4A^{0.75}\gamma^{0.25}) = log\left(\frac{A^{0.25} + \gamma^{0.25}\theta}{A^{0.25} - \gamma^{0.25}\theta}\right) - log\left(\frac{A^{0.25} + \gamma^{0.25}}{A^{0.25} - \gamma^{0.25}}\right)$$

$$\frac{A^{0.25} + \gamma^{0.25}}{A^{0.25} - \gamma^{0.25}} exp\left(\tau\left(4A^{0.75}\gamma^{0.25}\right)\right) = \frac{A^{0.25} + \gamma^{0.25}\theta}{A^{0.25} - \gamma^{0.25}\theta}$$

The exponential term can be written as the ratio of time and heating timescale.

$$t_H = \frac{t_0}{4(\alpha+\gamma)^{\frac{3}{4}}\gamma^{\frac{1}{4}}}$$

At steady state: $\frac{d\theta}{d\tau} = 0$

$$0 = \alpha - \gamma * \left(\theta^4 - 1\right)$$

$$\left(\frac{\alpha}{\gamma} + 1\right)^{0.25} = \theta_{ss}$$

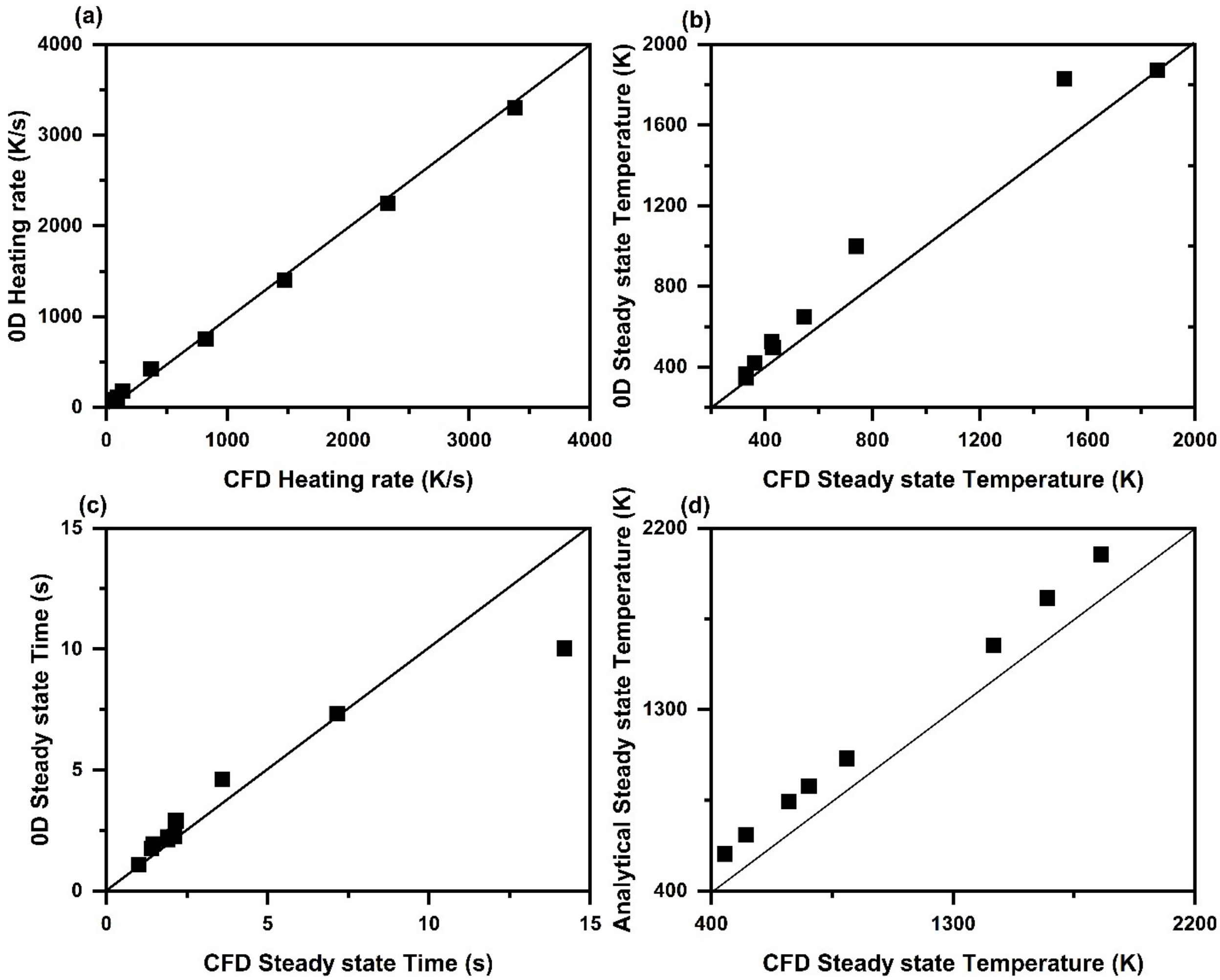

Figure S9: Parity plots of 0D predicted vs. CFD. (a) Heating rate, (b) temperature at steady state, (c) time to steady state, and (d) temperature. (Voltage applied to CFP = 2-30 V, L = 38 mm, L_{th} = 0.21 mm, L_w = 8 mm, He flowing at 90 ml/min).

COMPARISON BETWEEN GAS AND LIQUID

We consider water and Helium (He). Assuming similar setup, CFP at $T_{e,ss} = 90$ ℃, a quartz tube with r = 11 mm.

Table S2: Material properties for water and He (at 20 ℃).

	Water[48]	Helium
Flow rate	30 mL/min – 1000 mL/min[45]	90 sccm[18]
Density	1000 kg/m^3	0.16
Viscosity	0.001 Pa*s	1.95 x 10^{-5} Pa*s
Thermal conductivity	0.6 W/(m*K)	0.15 W/(m*K)
Heat capacity	4180 J/(kg*K)	5195 J/(kg*K)

Table S2: Material properties for water and He (at 20 ℃).

Based on the material properties and the flow rate, an empirical correlation is used and the results are shown below.

	Water	Helium
Reynolds number (Re)	50 - 1700	0.86
Richardson Number (Ri)	0.0001 - 0.07 < 0.1, forced convection	134 > 0.1, free convection
Empirical correlation for Nusselt number (Nu)	$0.663 Re^{0.5} Pr^{0.33}$	Eq. 12
Nu	10-53	4

Table S3: Empirical correlations used and results.

For water flow rate < 30 mL/min, Ri >0.1, charactrristic of free convection. Eq. 12 can be used for water in this case and Nu ~ 52. Pe < 0.3, hence $\frac{Nu}{Pe}$ is an order of magnitude higher as compared to gases, making β much larger for liquids.

HEATING TIME SCALE FOR THE GAS

Our results show only a minute fraction of the electric heating gets transferred to the gas. To capture the influence of physical properties on the gas outlet temperature (Figure *3*), we propose Eq. S5, to calculate t_{eh}

$$t_{eh} = \frac{\rho_g C_{p,g} V_s (T_{e,ss} - T_a)}{hA(T_{e,ss} - T_a)} \sim \frac{\rho_g C_{p,g}\, LL_{th}}{\kappa Nu_L} \sim \frac{LL_{th}}{aNu_L} \tag{S5}$$

Here a is the thermal diffusivity of the system. h can be predicted using Eq. 16, and this allows Eq. S5 to capture all important gas properties. Eq. S5 assumes that the time constant for a gas film of the same volume as the element to reach $T_{e,ss}$ (gas film is in thermal equilibrium with the element). For instance, *He* flowing with an inlet flow rate of 720 mL/min over the carbon element has t_{eh} ~ 0.03 s, which is much smaller than t_0 (~ 1 s) allowing sufficient time to heat up the gas near the element. *Balakotaiah and Ratnakar*[40] also propose a gas heating time scale from the power generated by Joule heating. Figure 3 indicates that under certain conditions, such as ours, only a fraction of the generated power is absorbed by the gas; most heat is lost directly by radiation.

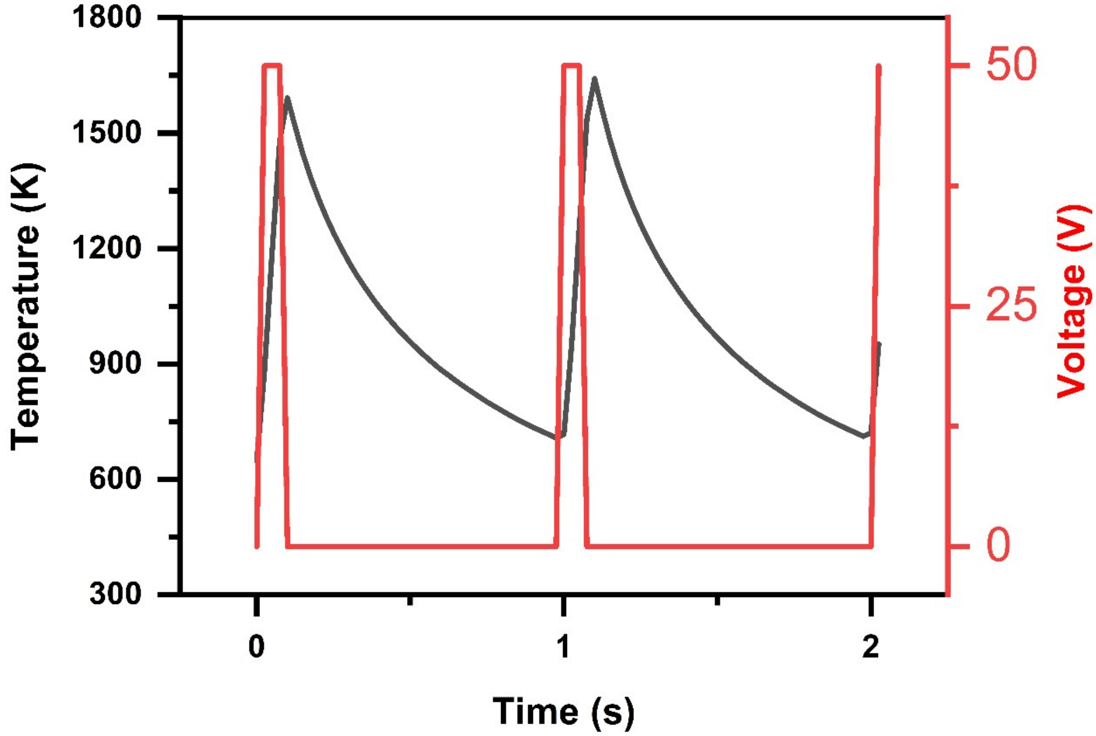

Figure S10: Temperature profile outcome for the voltage profile applied to the CFP element (50 V applied for 50 ms and 950 ms of cooling time).

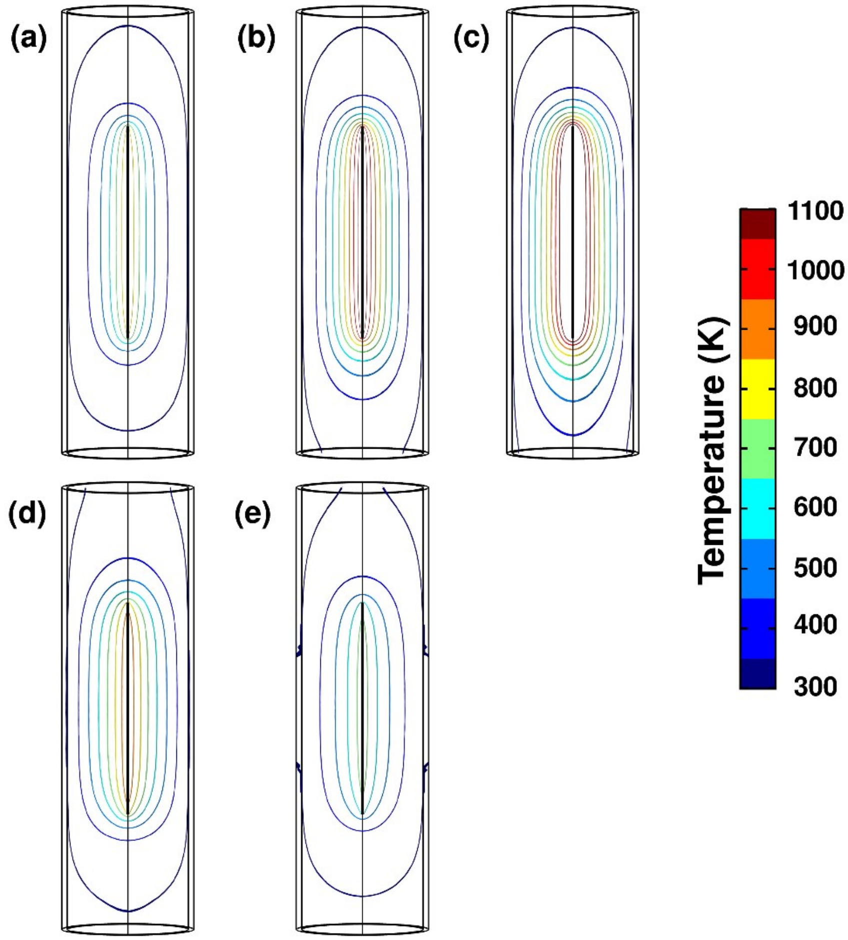

Figure S11: Temperature contours in the ZX plane at various times of one pulse of 50 V for 50 ms heating and 950 ms cooling with a *He* inlet flow rate of 90mL/min. (a) Heating starts, (b) heating in the middle, (c) heating stops, (d) middle of cooling, and (e) cooling stops.

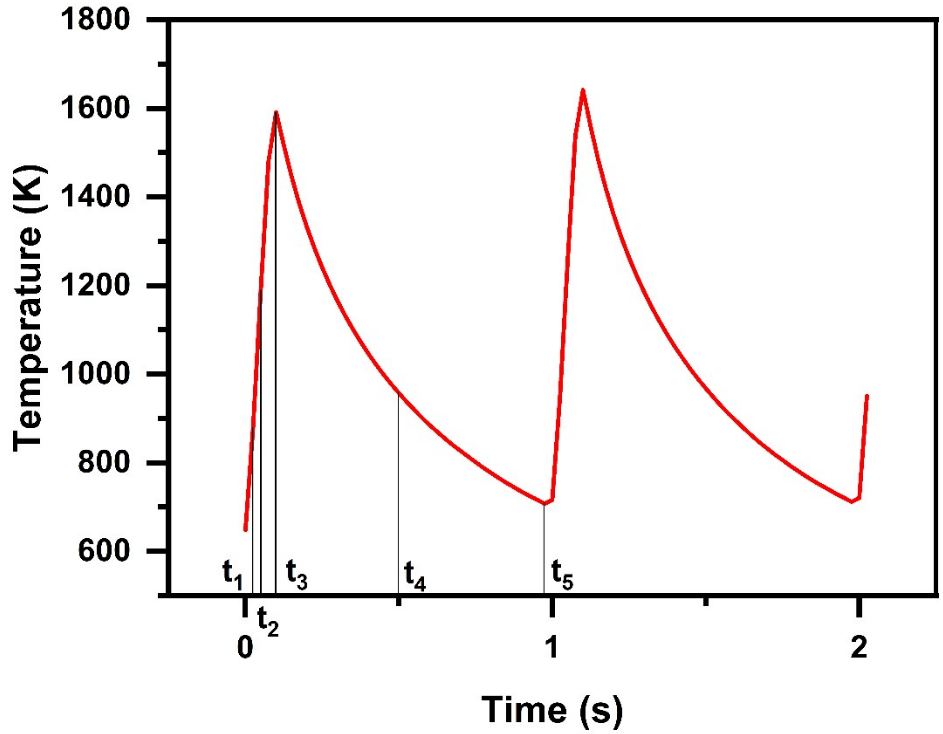

Figure S12: Temperature profile for 50 V applied for 50 ms and turned off for 950 ms. t_1 = heating starts, t_2 = heating middle, t_3 = heating stops, t_4 = cooling middle, and t_5 = cooling stops.

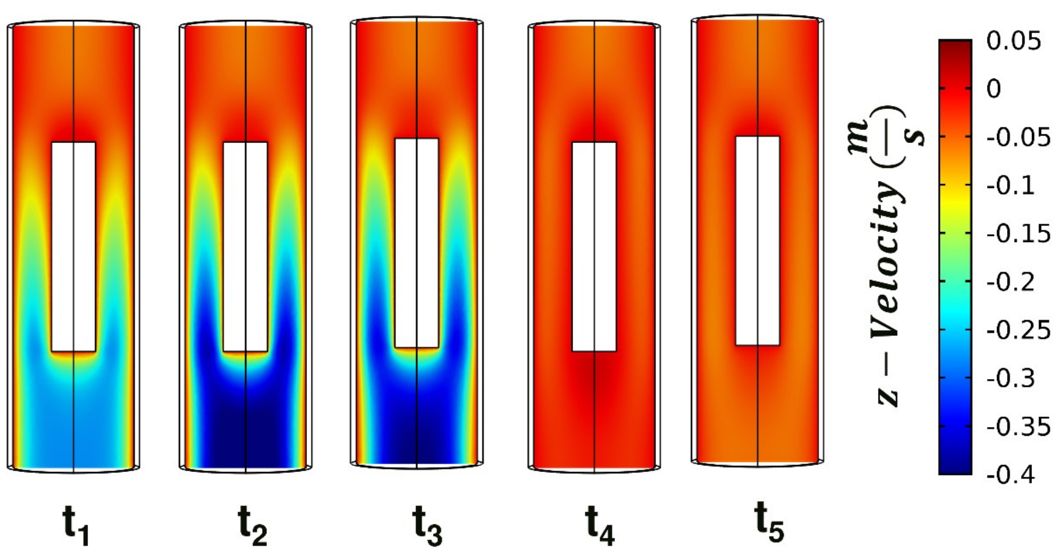

Figure S13: Axial velocity of *He* flowing in at 720 mL/min along various times of a pulse of 50 ms of 50 V applied and 950 ms of cooling.

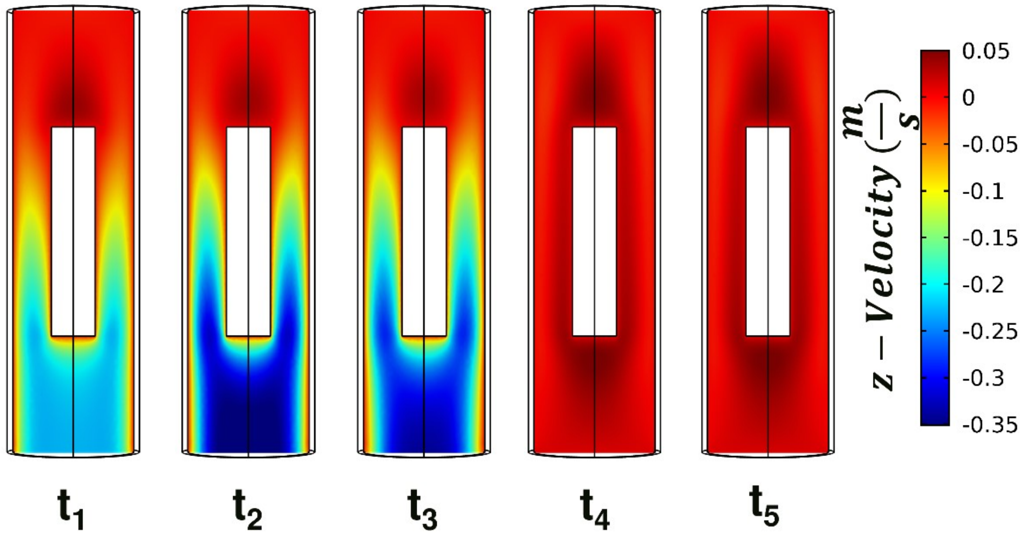

Figure S14: Axial velocity of *He* flowing in at 90 mL/min along various times of a pulse of 50 ms of 50 V applied and 950 ms of cooling.

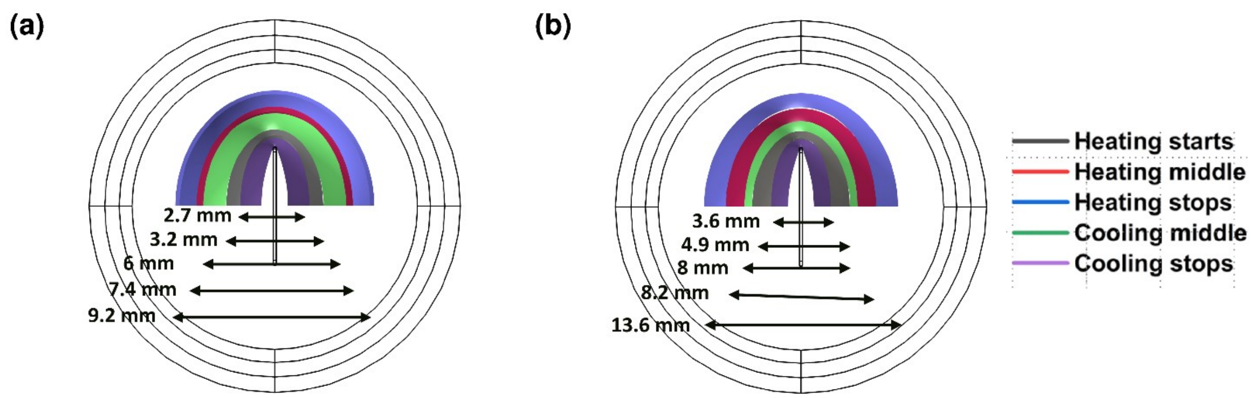

Figure S15: 600 K isothermal contours at various times of one pulse of 50 V applied for 90 ms and 950 ms of cooling with *He* flowing at (a) 90 mL/min and (b) 720 mL/min.