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## Numerical Investigation of the Blowing Effect on Laser Rayleigh Scattering Temperature Measurements via a Coupled Method of Lines Formulation

Laser Rayleigh Scattering (LRS) serves as a critical non-intrusive diagnostic for boundary layer thermometry; however, its accuracy is significantly compromised near pyrolyzing surfaces due to transient shifts in gas composition. In Poly(methyl methacrylate) (PMMA) environments, the rapid efflux of high-molecular-weight fuel vapor displaces the inert calibration gas, leading to a systematic bias that is frequently misinterpreted as a physical temperature decrease. This research resolves this diagnostic ambiguity by developing a transient, one-dimensional coupled thermo-kinetic framework. The model integrates solid-phase Arrhenius degradation kinetics ( $E = 230$  kJ/mol) with gas-phase species transport equations, utilizing the Method of Lines (MOL) to solve the resulting stiff system of partial differential equations. Quantitative results demonstrate that the displacement of the helium tracer by Methyl Methacrylate (MMA) monomer increases the effective Rayleigh scattering cross-section by a factor of 3.1. It is shown that failing to account for this compositional shift leads to a temperature underestimation of approximately 650 K during the quasi-steady gasification phase ( $T_S \approx 593.4$  K). Furthermore, the simulation confirms that the characteristic “temperature dip” observed in raw LRS experimental data at the onset of ignition is a compositional artifact rather than a thermal phenomenon. This work establishes a physics-based protocol for de-biasing optical measurements through dynamic correction factors ( $\alpha$ ), providing a scalable methodology for high-fidelity thermometry in variable-composition pyrolyzing systems.

*Keywords:* PMMA pyrolysis, Rayleigh scattering, compositional bias, blowing effect, method of lines, thermal boundary layer, numerical modeling, species transport, methyl methacrylate, optical diagnostics, heat conduction, thermometry

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### Introduction

Solid fuel combustion, particularly involving polymers like PMMA, is governed by the intricate coupling between solid-phase pyrolysis and gas-phase transport. Accurate characterization of the thermal boundary layer is critical for predicting flame spread; however, traditional intrusive probes suffer from thermal inertia and flow disruption [1]. Consequently, non-intrusive techniques like Laser Rayleigh Scattering (LRS) are preferred for their high temporal and spatial resolution. The fundamental validity of LRS thermometry relies on the inverse proportionality between scattered light intensity ( $I_R$ ) and temperature ( $T$ ), assuming a known effective scattering cross-section ( $\sigma_{\text{mix}}$ ) [2, 3]. In reacting flows, however, the rapid injection of fuel vapor—specifically Methyl Methacrylate (MMA) monomer—into the inert calibration gas (typically Helium or Air) drastically alters  $\sigma_{\text{mix}}$  [4–6]. Since the scattering cross-section of MMA is significantly larger than that of air or helium, failing to account for this compositional shift leads to substantial measurement errors, often interpreted falsely as a temperature drop. Existing models often decouple kinetics from species transport or rely on steady-state assumptions impractical for transient ignition studies. This research addresses this gap by developing a transient, fully coupled thermo-kinetic model using the Method of Lines (MOL) to predict instantaneous species profiles ( $X_i$ ) and quantify the systematic LRS temperature bias.

The modeling of solid fuel combustion is built upon foundational studies of polymer degradation. Pitts & Kashiwagi (1982) [7] and Gong & Yang (2024) [8] established the essential kinetic and thermophysical parameters for PMMA, characterizing its single-step thermal degradation into methyl methacrylate (MMA) monomer. Building on this solid-phase understanding, recent works by Morrisset et al. (2023) [6] and W. Hittini et al. (2024) [9] have investigated flame spread mechanisms, utilizing temperature reconstruction

methods to highlight the experimental difficulty of resolving thermal gradients near the pyrolyzing surface due to the intrusiveness of physical probes.

To address these measurement limitations, the field has increasingly adopted non-intrusive optical techniques. B. Wu et al. (2025) [10] established the rigorous methodology for Laser Rayleigh Scattering (LRS) in turbulent non-premixed systems, emphasizing that accurate thermometry requires accounting for species-dependent Rayleigh cross-sections ( $\sigma_{\text{mix}}$ ). While, M. Nasf (2023) [11] & Gupta et al. (2023) [12] successfully applied advanced laser diagnostics to study boundary layer stabilization, their work focused on flow structure rather than resolving the transient compositional ambiguity of the fuel vapor itself. Furthermore, while F.L. Tabares & I. Junkar (2021) [13] & Peters (2001) [14] provided the theoretical basis connecting mixture fractions to temperature, existing numerical models often decouple solid pyrolysis from gas-phase transport. This research bridges that gap, integrating the kinetic models of Kashiwagi with the diagnostic framework of Patron to quantify the specific compositional bias introduced by fuel blowing.

### Materials and Methods

The physical domain consists of a finite-thickness PMMA slab coupled to a gaseous boundary layer. The model assumes one-dimensional transport normal to the surface, constant thermophysical properties, and single-step zero-order Arrhenius kinetics. The transport of fuel vapor (MMA) from the surface and the background species (Air and Helium) within the boundary layer is governed by the principles of convection and diffusion. The numerical simulation of this process is essential to determine the local gas composition ( $X_i$ ), which is a prerequisite for accurate Rayleigh scattering diagnostics. Figure 1 illustrates a Gas-Species Transport Model focused on the Pyrolyzing Surface Interface of a solid fuel (PMMA), showing the coupled mass and energy transfer between the solid and gas phases.

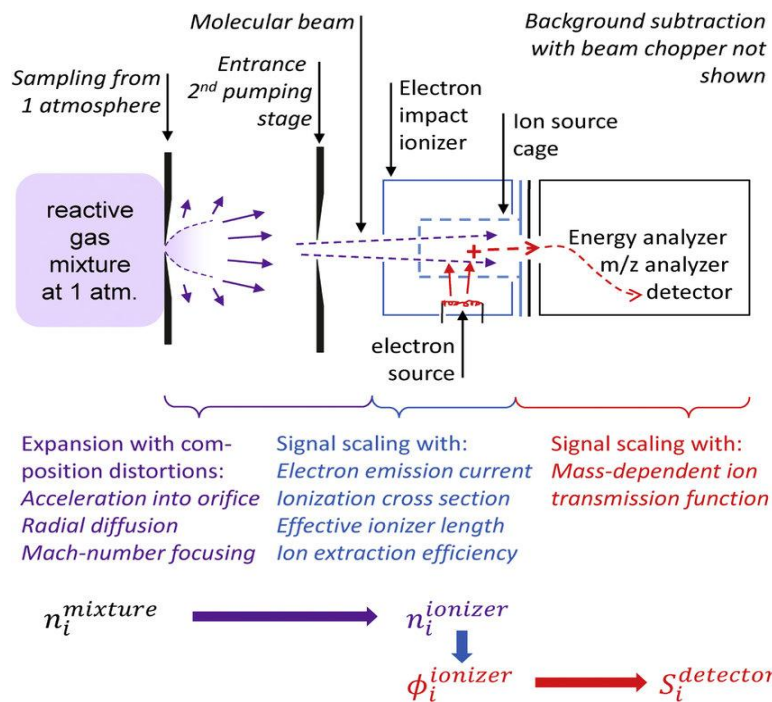


Figure 1. Schematic of the one-dimensional coupled solid-gas domain illustrating the interaction between external heat flux, solid-phase pyrolysis, and gas-phase species transport [13, 14]

### Solid Phase Pyrolysis

The transient heating of the PMMA slab is governed by the heat conduction equation with an internal heat sink representing the endothermic pyrolysis reaction [15]:

$$\rho_s C_{p,s} \frac{\partial T}{\partial t} = k_s \frac{\partial^2 T}{\partial x^2} - L_g \dot{m}'' \quad (1)$$

where  $\dot{m}'''$  is the mass loss rate per unit volume, defined by the Arrhenius law:  $\dot{m}''' = \rho_s A e^{\left(\frac{-E}{RT}\right)}$ . The surface energy balance at  $x=0$  couples the phases, accounting for external flux ( $q''_{ext}$ ), radiative/convective losses, and the latent heat of gasification ( $L_g$ ). The gas phase transport for species  $i$  (Fuel Vapor, Helium tracer, Air) is described by the transient convection-diffusion equation (2) [13, 15, 16]:

$$\rho_g \frac{\partial X_i}{\partial t} + \rho_g v_{conv} \frac{\partial X_i}{\partial x} = \rho_g D_{i,mix} \frac{\partial^2 X_i}{\partial x^2}. \quad (2)$$

Crucially, the convective velocity  $v_{conv}$  is not constant but is driven by the “blowing effect” of the pyrolysis mass flux:  $v_{conv} = \dot{m}''_{total} / \rho_g$ .

The system is discretized spatially using central finite differences ( $N = 40$  nodes per phase), converting the PDEs into a stiff system of Ordinary Differential Equations (ODEs). Due to the high stiffness introduced by the exponential Arrhenius term and distinct timescales of conduction vs. diffusion, the system is solved using the MATLAB ode15s solver with a maximum time step of 0.001 s to capture the rapid ignition transition. Table 1 shows the kinetic properties for PMMA and Gas species.

Table 1

**Thermophysical and Kinetic Properties for PMMA and Gas Species**

Parameter	Symbol	Value	Unit
Solid Phase (PMMA)			
Density	$\rho_s$	1190	kg/m <sup>3</sup>
Thermal Conductivity	$k_s$	0.05	W/m·K
Specific Heat	$c_{p,s}$	1420	J/kg·K
Pre-exponential Factor	$A$	$2.57 \times 10^7$	s <sup>-1</sup>
Activation Energy	$E$	230,000	J/mol
Heat of Gasification	$L_g$	$1.6 \times 10^6$	J/kg
Gas Phase (at 300 K)			
Helium Cross-section	$\sigma_{He}$	0.015	Relative to Air
MMA Cross-section	$\sigma_{MMA}$	4.52	Relative to Air
Binary Diffusivity	$D_{fg}$	$2.1 \times 10^{-5}$	m <sup>2</sup> /s

### Results and Discussion

#### Thermal Response & Pyrolysis Onset

The numerical model successfully captures the non-linear relationship between the external heat flux ( $q''_{ext}$ ) and the pyrolysis onset time ( $t_{pyr}$ ), adhering to the theoretical relation  $t_{pyr} \propto (q''_{ext})^{-2}$ . As detailed in Table 2, increasing the flux from 10 kW/m<sup>2</sup> to 40 kW/m<sup>2</sup> reduces the onset time from 43.7 s to 2.7 s.

Table 2

**Comparative Pyrolysis Characteristics across Varying External Heat Fluxes**

Heat Flux ( $q_{ext}$ ), kW/m <sup>2</sup>	Pyrolysis Time ( $t_{pyr}$ ), s	Steady Temperature ( $T_{steady}$ ), K
10	43.7	577.7
20	10.7	587.8
30	4.7	593.4
40	2.7	597.3

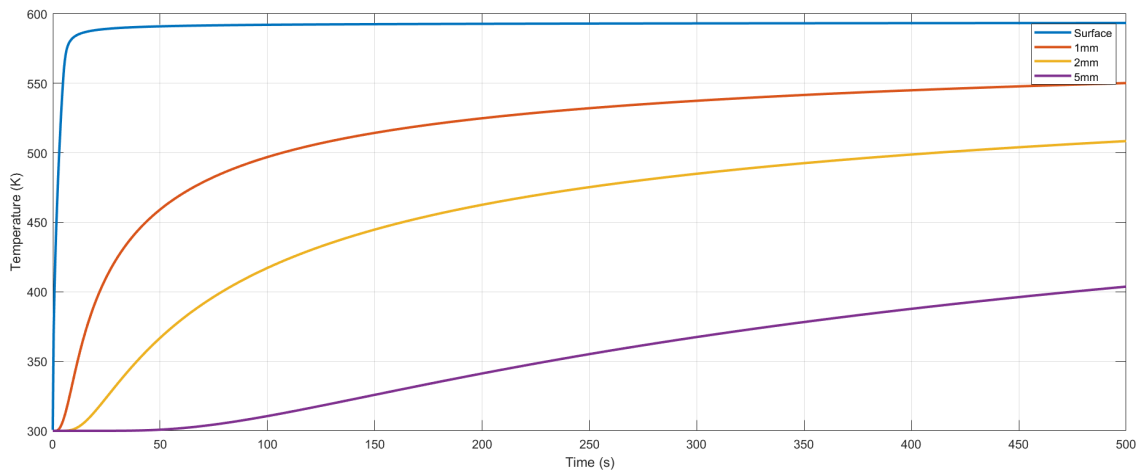


Figure 2. Transient temperature profiles at the PMMA surface and varying depths for an external heat flux of  $30 \text{ kW/m}^2$

As illustrated in Figure 2, the simulation reveals a physically realistic thermal transition. The internal profiles show a steep gradient from the ambient interior to a maximum at the surface. For a baseline flux of  $30 \text{ kW/m}^2$ , the surface temperature ( $T_s$ ) stabilizes at a quasi-steady value of  $\sim 593.4 \text{ K}$ . This stabilization confirms the dominance of the endothermic gasification term ( $L_g m''$ ) in the surface energy balance, acting as a thermal “heat sink” that prevents further sensible temperature rise.

#### Numerical Stability and Kinetic Sensitivity

Solving the PMMA pyrolysis model presents a computational challenge due to system stiffness. The ODEs are stiff because the Arrhenius mass loss rate,  $m'' = \rho_s A \exp(-E / RT_s)$ , is exponentially sensitive to  $T_s$ . The high activation energy ( $E = 230 \text{ kJ/mol}$ ) acts as a “kinetic switch”. As shown in Figure 3, once the surface reaches the pyrolysis threshold, the mass loss rate increases exponentially. The model demonstrates that a marginal increase in  $T_{\text{steady}}$  (approximately 20 K when doubling the flux from 20 to  $40 \text{ kW/m}^2$ ) is sufficient to achieve energy equilibrium.

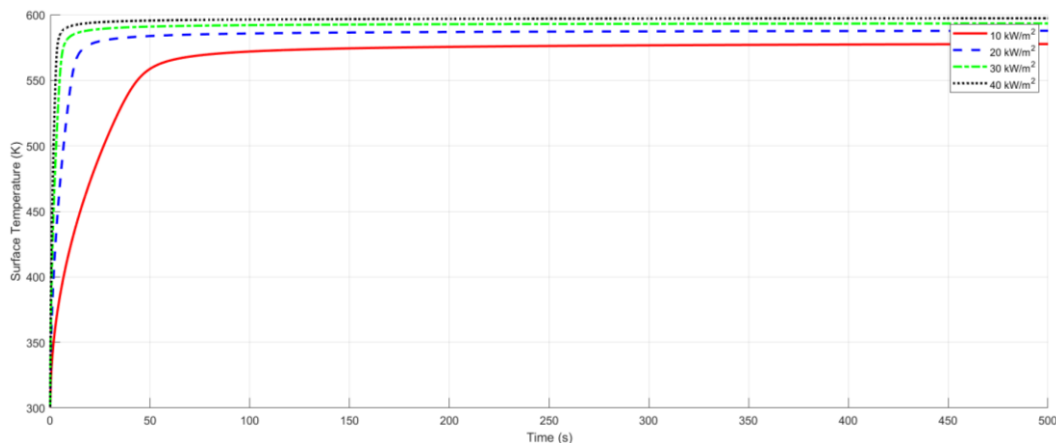


Figure 3. PMMA surface temperature evolution with respect to  $q_{\text{ext}}$  (a);  $10 \text{ kW/m}^2$  (b);  $20 \text{ kW/m}^2$  (c);  $30 \text{ kW/m}^2$  (d)  $40 \text{ kW/m}^2$

#### Fuel Blowing & Species Displacement Effect

A critical outcome is the quantification of the “blowing effect” and its impact on gas composition. During the pre-heat phase ( $t < t_{\text{pyr}}$ ), the boundary layer is dominated by the inert tracer ( $X_{\text{He}} \approx 1$ ). Upon pyrolysis onset, the rapid efflux of MMA vapor acts as a jet, physically displacing the helium tracer.

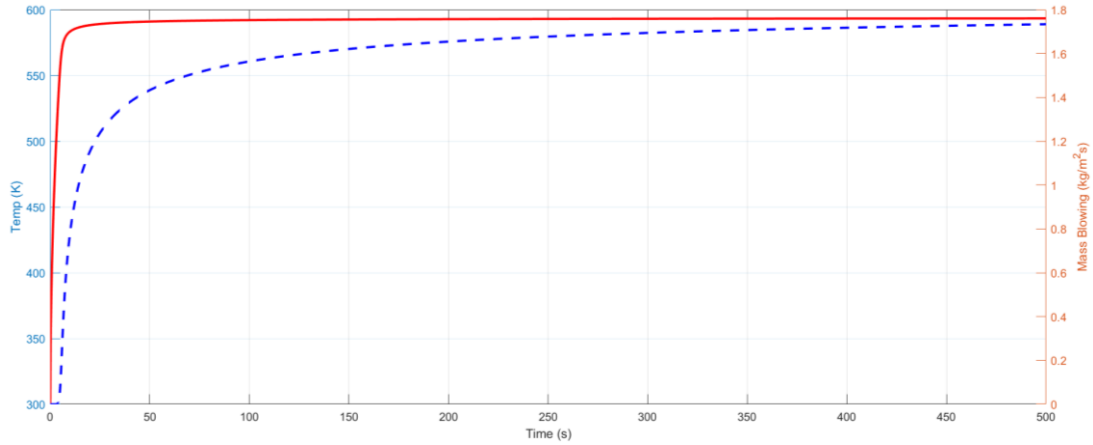


Figure 4. Coupled Thermal-Mass Feedback Mechanism with respect to fuel temperature and mass blowing

As seen in Figure 4, the coupling between surface temperature and mass blowing rate is nearly instantaneous. The influence of injection velocity ( $V_{inj}$ ) is explored in Figure 5. Higher injection velocities provide marginal convective cooling, but their primary role is the significant dilution of the fuel mole fraction ( $X_F$ ) at the surface. In the absence of high injection velocities,  $X_{He}$  drops precipitously as the high-molecular-weight fuel vapor floods the boundary layer. This phenomenon is vital for correctly calibrating LRS diagnostics.

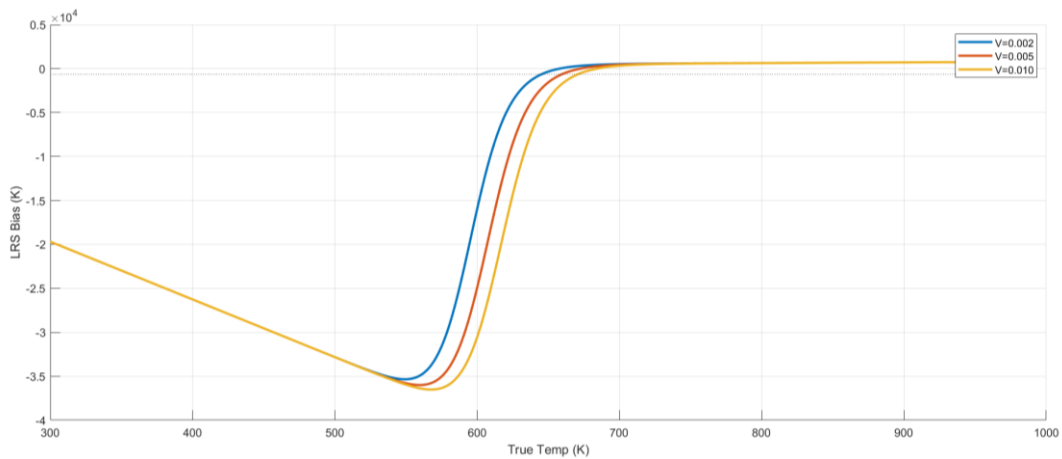


Figure 5. LRS Bias Sensitivity to Species Composition

*Quantification of Laser Rayleigh Scattering (LRS) Bias*

The central objective was to quantify the systematic error in Rayleigh thermometry caused by this species displacement. The effective scattering cross-section ( $\sigma_{mix}$ ) was calculated dynamically. Because the scattering cross-section of MMA is approximately 4.5 times that of air (while Helium is only 0.015 times that of air), the flooding of fuel vapor causes a drastic shift in  $\sigma_{mix}$ . Table 3 shows the LRS bias quantification at 30 kW/m<sup>2</sup>.

Table 3

**Quantification of LRS Temperature Bias at 30 kW/m<sup>2</sup>**

Time (s)	Phase	Actual $T_s$ (K)	Apparent $T_{LRS}$ (K)	Error ( $\Delta T$ )	$\alpha$ (Mix Ratio)
1.0	Initial	315.0	21,000	+20,685	0.015 (Pure He)
4.7	Onset	550.0	550.0	0	1.00 (Crossover)
100.0	Steady	593.4	1,243.4	-650	0.48 (Fuel Rich)

If this compositional shift is ignored, the LRS diagnostic yields an “apparent” temperature ( $T_{LRS}$ ) that deviates significantly from the physical temperature ( $T_s$ ). As quantified in Table 3, the LRS diagnostic underestimates the steady-state temperature by 650 K. This confirms that the characteristic “temperature dip” observed in raw LRS experimental data is a compositional artifact caused by the fuel vapor’s large cross-section, rather than a physical cooling phenomenon.

### Conclusion

This study successfully established a transient, one-dimensional coupled thermo-kinetic framework to resolve the intrinsic compositional bias in LRS thermometry during PMMA pyrolysis. By integrating solid-phase Arrhenius kinetics ( $E = 230$  kJ/mol) with gas-phase species transport via the MOL, the “blowing effect” and its impact on the local scattering cross-section ( $\sigma_{mix}$ ) were quantified across incident heat fluxes of 10–40 kW/m<sup>2</sup>. Quantitative analysis demonstrates that the rapid efflux of MMA vapor displaces the helium tracer, causing  $\sigma_{mix}$  to increase by a factor of approximately 3.1. The model reveals that if uncorrected, LRS diagnostics underestimate the true thermodynamic surface temperature by as much as 650 K during the quasi-steady gasification phase ( $T_s \approx 593.4$  K). Furthermore, this research provides a definitive physical explanation for the “temperature dip” frequently observed in raw LRS data at ignition onset. The simulation confirms this phenomenon as a compositional artifact caused by the fuel vapor’s high scattering cross-section, not a physical thermal event. By utilizing the derived correction factors ( $\alpha$ ) researchers can now recover high-fidelity thermal boundary layer data in variable-composition environments. This work enhances diagnostic accuracy for PMMA pyrolysis and provides a scalable, physics-based methodology for de-biasing optical measurements in pyrolyzing solid-fuel systems.

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## Сызықтар әдісінің байланысқан тұжырымдамасын қолдану арқылы лазерлік Рэлей шашырауы негізінде температураны өлшеуге үрлеу әсерінің сандық зерттелуі

Лазерлік Рэлей шашырауы (LRS) шекаралық қабаттағы температураны анықтауға арналған маңызды инвазивті емес диагностикалық әдіс, алайда пиролизге ұшырайтын беттер маңында газ құрамының уақытша өзгерістеріне байланысты оның дәлдігі айтарлықтай төмендейді. Полиметилметакрилат (PMMA) ортасында жоғары молекулалық массалы жанармай буларының жылдам бөлінуі инертті калибрлеуші газды ығыстырып, көбінесе температураның нақты төмендеуі ретінде қате түсіндірілетін жүйелік қателікке әкеледі. Жұмыста аталған диагностикалық анықсыздық бірөлшемді бейстационар байланысқан термокинетикалық модель құру арқылы жойылады. Модель қатты фазаның Аррениус бойынша ыдырау кинетикасын ( $E = 230$  кДж/моль) газ фазасындағы компоненттерді тасымалдау теңдеулерімен біріктіреді және алынған қатаң дербес туындылы дифференциалдық теңдеулер жүйесін шешу үшін сызықтар әдісін (MOL) қолданады. Сандық нәтижелер гелий трассерінің метилметакрилат (MMA) мономерімен ығыстырылуы Рэлей шашырауының тиімді қимасын 3,1 есе арттыратынын көрсетеді. Бұл құрам өзгерісін ескермеуде квазистационардың газдану кезеңінде ( $T_s \approx 593,4$  К) температураны шамамен 650 К-ге төмен бағалауға әкелетіні көрсетілді. Сонымен қатар модельдеу тұтану басталған сәтте LRS эксперименттік деректерінде байқалатын «температуралық төмендеу» құбылысы жылжылық емес, құрамдық әсердің нәтижесі екенін дәлелдейді. Бұл жұмыс оптикалық өлшеулердегі жүйелік қателіктерді динамикалық түзету коэффициенттері арқылы жоюға арналған физикалық негізделген тәсілді ұсынады және құрамы өзгермелі пиролиздік жүйелерде жоғары дәлдікті термометрия жүргізудің масштабталатын әдіснамасын қалыптастырады.

*Кілт сөздер:* PMMA пиролизі, Рэлей шашырауы, құрамдық қателік, үрлеу әсері, сызықтар әдісі, жылжылық шекаралық қабат, сандық модельдеу, компоненттер тасымалы, метилметакрилат, оптикалық диагностика, жылжылуәткізгіштік, термометрия

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## Численное исследование эффекта продувки на измерение температуры методом лазерного рэлеевского рассеяния с использованием сопряжённой формулировки метода линий

Лазерное рэлеевское рассеяние (LRS) является важным неинвазивным диагностическим методом для измерения температуры в пограничном слое; однако его точность существенно снижается вблизи пиролизующихся поверхностей из-за временных изменений состава газа. В средах полиметилметакрилата (PMMA) быстрый выход паров топлива с высокой молекулярной массой вытесняет инертный калибровочный газ, что приводит к систематической погрешности, часто ошибочно интерпретируемой как физическое снижение температуры. В данной работе эта диагностическая неопределённость устраняется путём разработки нестационарной одномерной сопряжённой термокинетической модели. Модель объединяет кинетику разложения твёрдой фазы по Аррениусу ( $E = 230$  кДж/моль) с уравнениями переноса компонентов в газовой фазе, используя метод линий (MOL) для решения возникающей жёсткой системы дифференциальных уравнений в частных производных. Количественные результаты показывают, что вытеснение гелиевого трассера мономером метилметакрилата (MMA) увеличивает эффективное сечение рэлеевского рассеяния в 3,1 раза. Показано, что игнорирование этого изменения состава приводит к занижению температуры примерно на 650 К в квазистационарной фазе газификации ( $T_s \approx 593,4$  К). Кроме того, моделирование подтверждает, что характерное «проваливание температуры», наблюдаемое в исходных экспериментальных данных LRS в момент начала воспламенения, является следствием изменения состава, а не реальным тепловым эффектом. Данная работа формирует физически обоснованный подход к устранению смещения в оптических измерениях с

использованием динамических поправочных коэффициентов, обеспечивая масштабируемую методологию для высокоточной термометрии в пиролизирующихся системах с переменным составом.

*Ключевые слова:* пиролиз ПММА, рэлеевское рассеяние, композиционная погрешность, эффект продувки, метод линий, тепловой пограничный слой, численное моделирование, перенос компонентов, метилметакрилат, оптическая диагностика, теплопроводность, термометрия

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