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The unsteady-state energy conservation equation for a small spherical particle in LII modeling

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ABSTRACT This paper discusses the formalization of the energy variation rate of a small particle encountered in the modeling of an unsteady-state soot particle temperature and diameter during laser-induced incandescence. A derivation of the particle energy equation is presented based on the first law of thermodynamics applied to an open system. Problems associated with an incorrect particle internal energy variation rate used in the literature are discussed. Numerical calculations are presented to demonstrate the effects of several incorrect particle internal energy variation rates on the calculated particle temperature and diameter.

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1 Introduction

Laser-induced incandescence (LII) has become a popular non-intrusive technique for measuring the volume fraction and particle size (mean or distribution) of combustion-generated nano-sized soot particles in many different applications [1–13]. It has also been demonstrated that LII can also be used to infer the size of non-carbon nano-particles [14, 15]. In LII, nano-particles, such as soot, suspended in a carrier gas are first thermally excited using a high power laser pulse with a duration of about 20 ns. The particles inside the laser beam are rapidly heated to a temperature significantly higher than that of the surrounding gas by the end of the laser pulse. After the laser pulse, the particles start to cool primarily through sublimation (if the particle temperature is higher than about 3400 K [13]) and by conduction heat loss. As the particle temperature decreases, conduction becomes increasingly important and eventually is the dominant heat loss mechanism. Although thermal radiation is the basis of LII, it in general remains negligible, compared to heat conduction, as a particle heat loss mechanism at atmospheric or higher pressures. Due to their larger surface area-to-volume ratios, small particles cool faster than larger ones. This forms the principle of LII based particle sizing techniques.

To determine the particle size distribution, it is essential to have an accurate LII model to describe the variation of particle temperature with time, which includes both the physical model and the optical and thermal properties of the particles. In addition, the initial particle diameter distribution and the surrounding gas properties, such as pressure, temperature, and compositions, are also required. As shown in a recent comprehensive comparison of different LII models [16], the accurate calculation of the particle laser energy absorption rate is greatly limited by our knowledge of the value of soot absorption function $E(m)$. Accurate calculation of the particle conduction heat loss rate is possible, provided a reliable thermal accommodation coefficient is available. Modeling of the sublimation heat loss continues to exhibit large differences among LII models used by different researchers [16].

Besides the different treatments of the particle laser energy absorption rate and the various particle heat loss mechanisms among LII models documented in [16], there exists two different expressions for the particle internal energy variation rate: one is used by Kock et al. [14, 16] and Hofmann et al. [16, 18] (hereinafter referred to as K&H), and the other is used by all other researchers. According to Hofmann et al. [18], these two different expressions for the particle internal energy variation rate lead to a rather significant difference in the calculated particle temperature decay rate, even at low laser fluences. Such an effect implies that a rather large error is incurred in the thermal accommodation coefficient determined by the low-fluence LII technique described in [19] if an incorrect particle internal energy variation rate expression were used.

An attempt to ascertain how to evaluate the particle internal energy variation rate is the motivation of the present study. More specifically the objectives of this paper are:

1. To discuss the consequences of using the two different particle internal energy variation rate expressions.
2. To present a derivation of the energy conservation equation for a small particle from the first law of thermodynamics and establish how the particle internal energy variation rate should be evaluated.
3. To compare the numerical results of soot particle temperature and diameter obtained using the different expressions for the particle internal energy variation rate at two laser fluences.

Throughout this study we consider particles that are sufficiently small such that their temperature is uniform, i.e., there

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is no internal temperature gradient. Such an assumption is commonly made in LII studies.

2 Theory

2.1 *Expressions for the particle internal energy variation rate in LII model*

The numerical model describing the energy and mass conservation of a single isolated nano-sized spherical particle in LII can be written as [16]

$$Q_{\mathrm{int}} = Q_{\mathrm{abs}} + Q_{\mathrm{sub}} + Q_{\mathrm{cond}} + Q_{\mathrm{rad}} \,, \tag{1}$$

$$\frac{\mathrm{d}M_{\mathrm{p}}}{\mathrm{d}t} = J_{\mathrm{sub}} \,, \tag{2}$$

where Q_{int} on the left hand side of (1) represents the rate of the particle internal energy change, the terms on the right hand side of (1) stand for the particle laser energy absorption rate, heat loss rates due to sublimation, heat conduction, and radiation, respectively. Other particle heat transfer mechanisms, such as oxidation, described by Michelsen [16, 17] are not taken into account in this study. In (2), M_{p} is the particle mass, t is time, and J_{sub} is the total mass sublimation rate from the particle surface. Although the expressions for the terms on the right hand side of (1) are somewhat different in different LII models [16], they are more or less similar with the largest difference being in the expression for the sublimation terms Q_{sub} and J_{sub}. However, there also exists two substantially different expressions for the particle energy variation rate, which is the primary concern of the present study.

In most LII models used by different researchers, this term is written as

$$Q_{\mathrm{int}} = V_{\mathrm{p}} \varrho_{\mathrm{p}} c_{\mathrm{p}} \frac{\mathrm{d}T}{\mathrm{d}t} \,, \tag{3}$$

where $V_{\mathrm{p}} = \pi d_{\mathrm{p}}^3/6$ is the particle volume, d_{p} is the particle diameter, ϱ_{p} is the particle density (assuming temperature independent), c_{p} is the particle specific heat (temperature dependent), and T is the particle temperature. Substitution of (3) into (1) leads to the derivative of particle temperature with respect to time as

$$\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{Q_{\mathrm{abs}} + Q_{\mathrm{sub}} + Q_{\mathrm{cond}} + Q_{\mathrm{rad}}}{M_{\mathrm{p}} c_{\mathrm{p}}} \,, \tag{4}$$

where $M_{\mathrm{p}} = V_{\mathrm{p}} \varrho_{\mathrm{p}}$ is the particle mass. It is noted that in the LII studies conducted by K&H, the following expression was used for the particle internal energy variation rate

$$Q_{\mathrm{int}} = \frac{\mathrm{d}}{\mathrm{d}t} \left(V_{\mathrm{p}} \varrho_{\mathrm{p}} c_{\mathrm{p}} T \right) = \frac{\mathrm{d}}{\mathrm{d}t} \left(M_{\mathrm{p}} c_{\mathrm{p}} T \right) \,. \tag{5}$$

By using the following expansion

$$\begin{aligned} Q_{\mathrm{int}} &= \frac{\mathrm{d}}{\mathrm{d}t} (M_{\mathrm{p}} c_{\mathrm{p}} T) = M_{\mathrm{p}} c_{\mathrm{p}} \frac{\mathrm{d}T}{\mathrm{d}t} + M_{\mathrm{p}} T \frac{\mathrm{d}c_{\mathrm{p}}}{\mathrm{d}t} + c_{\mathrm{p}} T \frac{\mathrm{d}M_{\mathrm{p}}}{\mathrm{d}t} \\ &= \left(M_{\mathrm{p}} c_{\mathrm{p}} + M_{\mathrm{p}} T \frac{\mathrm{d}c_{\mathrm{p}}}{\mathrm{d}T} \right) \frac{\mathrm{d}T}{\mathrm{d}t} + c_{\mathrm{p}} T \frac{\mathrm{d}M_{\mathrm{p}}}{\mathrm{d}t} \,. \end{aligned} \tag{6}$$

K&H obtained the derivative of particle temperature with respect to time as

$$\frac{\mathrm{d}T}{\mathrm{d}t} = \frac{Q_{\mathrm{abs}} + Q_{\mathrm{sub}} + Q_{\mathrm{cond}} + Q_{\mathrm{rad}} - c_{\mathrm{p}} T J_{\mathrm{sub}}}{M_{\mathrm{p}} (c_{\mathrm{p}} + T \,\mathrm{d}c_{\mathrm{p}}/\mathrm{d}T)} \,. \tag{7}$$

It is therefore evident that there are two extra terms in the particle energy equation in the LII models of K&H, (7), compared to models used by others, (4). One of these terms is in the numerator ($-c_{\mathrm{p}} T J_{\mathrm{sub}}$), the other ($M_{\mathrm{p}} T \,\mathrm{d}c_{\mathrm{p}}/\mathrm{d}t$) is in the denominator. There are in effect two 'sublimation' terms in (7): one is the 'normal' sublimation term Q_{sub}, which is in most LII models written as

$$Q_{\mathrm{sub}} = \frac{\Delta H_{\mathrm{v}}}{M_{\mathrm{v}}} \frac{\mathrm{d}M_{\mathrm{p}}}{\mathrm{d}t} \,, \tag{8}$$

the other, $-c_{\mathrm{p}} T J_{\mathrm{sub}}$, originates from the variation rate of the particle internal energy due to mass loss.

The qualitative effects of these two extra terms can be easily understood within the context of LII of soot where the specific heat increases with temperature, i.e., the term $T \,\mathrm{d}c_{\mathrm{p}}/\mathrm{d}T > 0$, and the particle loses mass, i.e., $J_{\mathrm{sub}} < 0$ and $-c_{\mathrm{p}} T J_{\mathrm{sub}} > 0$. Therefore, at low-fluence LII where the mass loss rate is negligible, the particle temperature variation rate is reduced by the extra term, which means that the peak particle temperature is lowered and the particle temperature decays more slowly after the laser pulse. By contrast, at high fluences, the term $-c_{\mathrm{p}} T J_{\mathrm{sub}}$ in the numerator of (7) behaves like a strong source term that will increase the particle temperature.

In the derivation of particle internal energy variation rate of K&H, it is implicitly assumed that the particle internal energy U is defined as

$$U = M_{\mathrm{p}} c_{\mathrm{p}} T \tag{9}$$

and the particle internal energy variation rate is evaluated as $Q_{\mathrm{int}} = \mathrm{d}U/\mathrm{d}t$. The specific heat c_{p} is in general a function of T and is linked to the particle internal energy U, for example in a study of nano-sized soot particle combustion, Hiers [20, 21], as

$$U = M_{\mathrm{p}} u = M_{\mathrm{p}} \left(\int_{T_{\mathrm{ref}}}^{T} c_{\mathrm{p}} \,\mathrm{d}T + u_{\mathrm{ref}} \right) \,, \tag{10}$$

where u is the specific (per unit mass) particle internal energy, T_{ref} and u_{ref} are respectively the reference temperature and the specific particle internal energy at the reference temperature, which were set to 0 J and 0 K respectively in [20, 21]. Equation (10) reduces to the expression used by K&H, (9), only under the assumption that the particle specific heat c_{p} is temperature independent. Consequently, the second term on the right hand side of (6) should vanish.

Another way to illustrate this point is that the particle internal energy expression used by K&H is in generally inconsistent with the definition of specific heat. For a solid material like soot (assumed to be graphite here), its specific heat at constant pressure is almost the same as that at constant volume and there is in general no need to differentiate the two. The specific heat of a solid material is defined as

$$c_{\mathrm{p}} \equiv \frac{\partial u}{\partial T} \,. \tag{11}$$

The particle internal energy expression used by Hiers [20, 21], (10), is consistent with this definition of specific heat, whether the specific heat c_p is temperature dependent or not. However, the definition of specific heat, (11), is not recovered in the expression of K&H ($u = c_\mathrm{p} T$) unless a temperature independent c_p is assumed.

By allowing both the particle temperature and its mass (or diameter) to vary with time, Hiers [20, 21] derives the particle energy conservation equation using the following expansion

$$\frac{\mathrm{d}U}{\mathrm{d}t} = \frac{\partial U}{\partial T}\frac{\mathrm{d}T}{\mathrm{d}t} + \frac{\partial U}{\partial M_\mathrm{p}}\frac{\mathrm{d}M_\mathrm{p}}{\mathrm{d}t} = M_\mathrm{p} c_\mathrm{p}\frac{\mathrm{d}T}{\mathrm{d}t} + u\frac{\mathrm{d}M_\mathrm{p}}{\mathrm{d}t}\,. \tag{12}$$

The first term on the right hand side of (12) is identical to the particle internal energy variation rate commonly used in LII studies, (3), and the first term on the right hand side of (6). The second term on the right hand side of (12) is equivalent to the third term on the right hand side of (6). The appearance of the second term on the right hand side of (6) is physically erroneous as mentioned above. Because of such inconsistent expression, the third term on the right hand side of (6) is also inadequate, i.e., $c_\mathrm{p} T$ should be replaced by $\int_{T_\mathrm{ref}}^{T} c_\mathrm{p}\,\mathrm{d}T$. It is evident that (12) reduces to (3) in the absence of particle mass loss. On the contrary, (6) does not reproduce (3) even when there is no particle mass loss. The effect of using these different expressions for the particle internal energy variation rate on the calculated particle temperature and diameter is shown later. It suffices to mention here that neither the expression of K&H, (6), nor that of Hiers [20, 21], (12), for the particle internal energy variation rate is correct.

2.2 *Derivation of the energy conservation equation for a small particle with mass loss*

Before a rigorous derivation for the unsteady-state energy conservation equation of a small particle subject to mass loss is presented, it is worth pointing out that the particle energy conservation equation used in LII studies, (1) with either (3) or (6) for the particle internal energy variation rate, is written somewhat heuristically rather than based on a sound theoretical derivation. Central to the problem is how the particle internal energy variation rate should be evaluated in situations where the particle undergoes heat and mass transfer simultaneously. Although K&H used an inappropriate expression for the particle internal energy, compare (9) to (10), it is quite clear that their motivation for using a different expression for the particle internal energy variation rate from the commonly used one written in (3) is to account for the additional particle internal energy variation caused by the particle mass loss through sublimation at high laser fluences, which is apparently not included in (3). This reasoning seems to provide some support to the particle internal energy variation rate expression used by K&H. Such seemingly correct expression for the particle internal energy variation rate has been used not only in LII studies but also in other fields, e.g., in combustion by Hiers [20, 21].

An obvious sign that (6) and (12) are problematic is that they require the absolute value of the particle specific internal energy u. It is well known that we have no means of measuring the absolute value of internal energy, which is dependent on a reference temperature. Only changes in the internal energy, which is independent of the reference temperature, can be measured. The fundamental flaw of the particle energy equation suggested by K&H and Hiers can also be illustrated as follows. Let us consider an idealized scenario where the particle undergoes only a ‘sublimation’ process that has no heat of sublimation, i.e, the particle experiences no heat loss but only mass transfer. Under these assumptions, it is evident that the particle temperature should physically remain constant while its diameter decreases with time. However, use of either (6) or (12) as the particle internal energy variation rate in (1) suggests that the particle temperature increases while its diameter decreases with time, which is clearly incorrect.

The starting point of the rigorous derivation of the unsteady-state energy conservation equation of a small particle is the first law of thermodynamics, i.e., the principle of energy conservation, applied to an open system. Consider a small particle shown in Fig. 1. The surface of the particle at time t is represented by the solid line. At a small time interval Δt later, i.e., at time $t + \Delta t$, the particle surface moves to a new position due to sublimation or any other mass loss mechanism, such as surface reaction, as indicated by the dashed line. According to the first law of thermodynamics applied to the sublimating particle considered here, which is an open system, the change in the total energy stored in the particle during this time interval can be written as

$$\Delta E = \delta Q + \Delta P - \delta W\,, \tag{13}$$

where ΔE is the variation in the total energy of the particle during time interval Δt. The total energy E includes the bulk kinetic energy, potential energy, and internal energy. For a motionless small particle considered in the present context, the change in the particle total energy is caused entirely by the change in the internal energy, i.e., $\Delta E = \Delta U$. Symbol δQ is the net heat transfer to the particle and δW is the work done by the particle, which is absent in the present study. It is noticed that there is another term, ΔP, on the right hand side of (13), which accounts for the particle energy change associated with the mass entering and with the mass leaving the system (here the particle). Dividing (13) by Δt leads to

$$\frac{\Delta U}{\Delta t} = \frac{\delta Q}{\Delta t} + \frac{\Delta P}{\Delta t}\,. \tag{14}$$

Assuming the particle density remains constant, i.e., temperature independent, which is often made in LII studies, the left

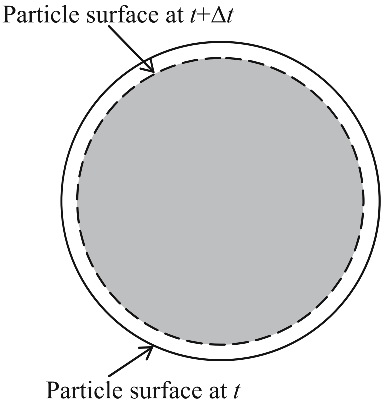

FIGURE 1 A schematic of the particle surface at time t and $t + \Delta t$

hand side of (14) can be written as

$$\frac{\Delta U}{\Delta t} = \frac{V_\mathrm{p}(t+\Delta t)\varrho_\mathrm{p} u(t+\Delta t) - V_\mathrm{p}(t)\varrho_\mathrm{p} u(t)}{\Delta t} = V_\mathrm{p}(t)\varrho_\mathrm{p}\frac{u(t+\Delta t)-u(t)}{\Delta t} + \frac{\Delta V}{\Delta t}\varrho_\mathrm{p} u(t+\Delta t)\,, \qquad (15)$$

where $\Delta V = V_\mathrm{p}(t+\Delta t) - V_\mathrm{p}(t)$ is the change in the particle volume, which is negative in the present context. As $\Delta t \to 0$, the first term on the right hand side of (15) becomes

$$\lim_{\Delta t\to 0} V_\mathrm{p}(t)\varrho_\mathrm{p}\frac{u(t+\Delta t)-u(t)}{\Delta t} = V_\mathrm{p}(t)\varrho_\mathrm{p}\frac{\partial u}{\partial t}\,. \qquad (16)$$

Assuming the particle surface recession radial velocity associated with sublimation is v_n at time t, one has

$$\Delta V = v_n \Delta t A_\mathrm{p}\,, \qquad (17)$$

where $A_\mathrm{p} = \pi d_\mathrm{p}^2$ is the particle surface area at time t. The second term on the right hand side of (15) is therefore equal to, as $\Delta t \to 0$,

$$\frac{\Delta V}{\Delta t}\varrho_\mathrm{p} u(t+\Delta t) = v_n A_\mathrm{p}\varrho_\mathrm{p} u\,. \qquad (18)$$

As $\Delta t \to 0$, the first term on the right hand side of (14) is the heat transfer rate to the particle, which can be evaluated as the net effect of all the heat transfer processes considered, i.e.,

$$\lim_{\Delta t\to 0}\frac{\delta Q}{\Delta t} = Q_\mathrm{abs} + Q_\mathrm{sub} + Q_\mathrm{cond} + Q_\mathrm{rad}\,. \qquad (19)$$

The rate of energy carried away by the mass lost from the particle in time interval Δt can be written as

$$\frac{\Delta P}{\Delta t} = \frac{\left(V_\mathrm{p}(t+\Delta t) - V_\mathrm{p}(t)\right)\varrho_\mathrm{p} u(t)}{\Delta t}\,, \qquad (20)$$

which is negative since the particle loses energy. This energy loss from the particle due to mass loss should not be confused with the sublimation heat loss term, though both are caused by removal of mass from the particle. The former accounts for the (internal) energy loss from the particle due to mass loss without phase change, while the latter accounts for additional energy loss due to phase change. Substitution of (17) into (20) leads to

$$\frac{\Delta P}{\Delta t} = v_n A_\mathrm{p}\varrho_\mathrm{p} u\,. \qquad (21)$$

Substitution of (15), (19), and (21) into (14), we obtain

$$V_\mathrm{p}\varrho_\mathrm{p}\frac{\partial u}{\partial t} + v_n A\varrho_\mathrm{p} u = Q_\mathrm{abs} + Q_\mathrm{sub} + Q_\mathrm{cond} + Q_\mathrm{rad} + v_n A\varrho_\mathrm{p} u\,. \qquad (22)$$

It is evident that the rate of particle energy loss due to removal of mass, which also has internal energy, from the particle (the last term on the right hand side), cancels out with the particle internal energy variation rate caused by mass loss, the second term on the left hand side. The physical significance of this cancellation is clear: removal of mass from the particle without phase change reduces the total particle internal energy, but should not change the internal energy per unit mass, i.e., the particle temperature. Upon using $\partial u/\partial t = \partial u/\partial T\,\mathrm{d}T/\mathrm{d}t = c_\mathrm{p}\,\mathrm{d}T/\mathrm{d}t$, (22) can now be written as

$$V_\mathrm{p}\varrho_\mathrm{p} c_\mathrm{p}\frac{\mathrm{d}T}{\mathrm{d}t} = Q_\mathrm{abs} + Q_\mathrm{sub} + Q_\mathrm{cond} + Q_\mathrm{rad}\,, \qquad (23)$$

which suggests that the particle internal energy variation rate should be calculated as that written in (3), but not those in (6) or (12). In other words, when the particle energy equation is written in the form of (1) the particle internal energy variation rate should be evaluated only by the temperature variation. The above derivation of the particle energy equation indicates that the particle energy equation suggested by K&H and that by Hiers [20] are incorrect, since the particle energy loss via mass loss, which also carries energy away from the particle, is neglected in their formulations. Under the assumptions of the idealized ‘sublimation’ scenario discussed earlier, (23) predicts that the particle temperature remains unchanged with time, though the particle diameter decreases, as expected. In addition, the absolute value of the particle internal energy is no longer required. This particle energy equation, (23), is also consistent with that used by most researchers in the combustion field for studying small carbon particle or droplet combustion, e.g. [22, 23].

3 Results and discussion

To demonstrate the effect of the particle internal energy variation rate expression on the calculated particle temperature and diameter in the entire process of LII, numerical calculations were conducted using the three different expressions, (3), (6), and (12). The surrounding gas is air at $p_\mathrm{g} = 1$ bar and $T_\mathrm{g} = 1800$ K. A monodisperse isolated spherical soot particle of $d_\mathrm{p} = 30$ nm was assumed and a laser wavelength of 532 nm. The spatial laser energy distribution was assumed to be top-hat and the temporal distribution is shown in Fig. 2. These conditions are identical to those used in the LII model comparisons documented in [16]. In our calculations, the following parameters were used: the soot

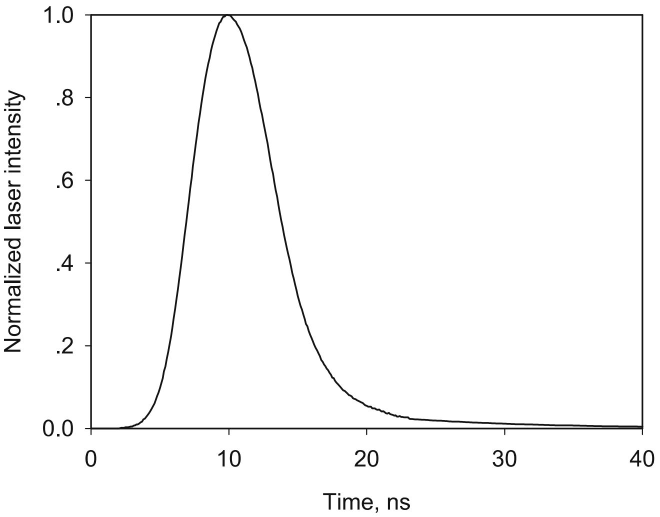

FIGURE 2 The normalized temporal laser intensity distribution

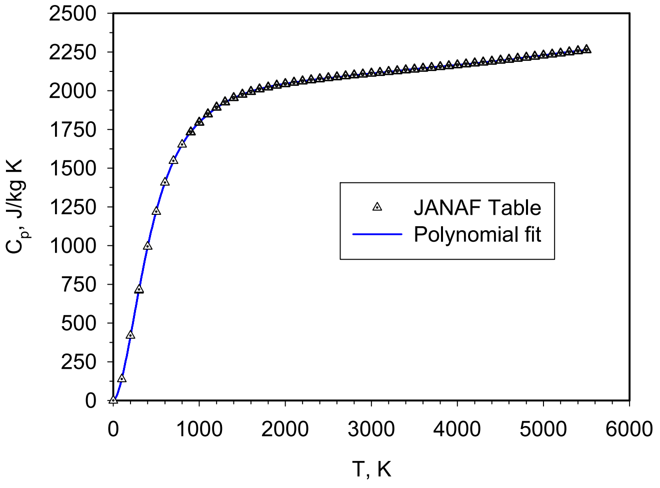

FIGURE 3 The specific heat of graphite from the JANAF tables

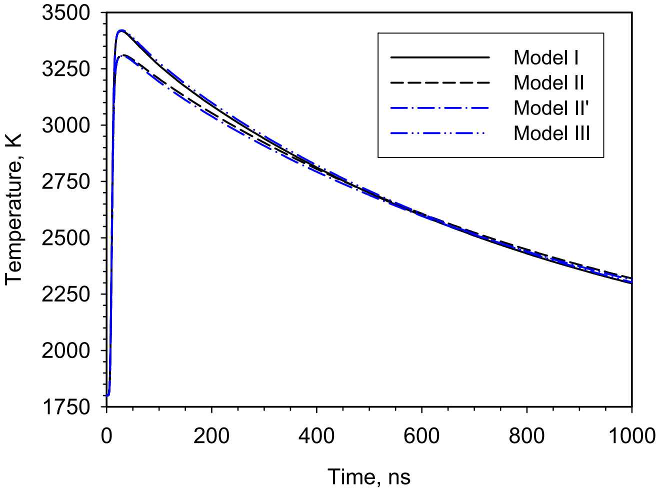

FIGURE 4 Effect of the particle internal energy variation rate expression on the calculated particle temperature at a low laser fluence of 0.5 mJ/mm^2

density is $\varrho_p = 1900\,\mathrm{kg/m^3}$, specific heat of soot is temperature dependent shown in Fig. 3 with its polynomial fit given in [16], the thermal accommodation coefficient of soot in air is assumed to be $\alpha = 0.37$, the soot absorption function $E(m)$ at 532 nm is 0.38, the effective sublimation coefficient is 0.77. Both sublimation and heat conduction occur in the free-molecular regime under the specified conditions. Further details of the LII model developed at NRC can be found elsewhere [16]. Calculations were carried out at two laser fluences of 0.5 mJ/mm^2 and 7 mJ/mm^2 to demonstrate the influence of these three particle internal energy variation rate expressions at low and high fluences, respectively. To help the presentation of the numerical results, the particle energy equation based on the commonly used particle internal energy variation rate expression, (3), is referred to as model I, while those based on (6) and (12) are referred to as model II and model III, respectively. It is noticed that in model III, the reference temperature for the particle internal energy was set to 0 K, as suggested by Hiers [20].

3.1 *Low fluence case*

Results of the low fluence case are shown in Fig. 4. At such a low laser fluence, there is essentially no soot sublimation and the particle diameter remains almost unchanged [16], i.e, the soot mass loss rate is negligibly small. It is noticed that there are two particle temperature curves in Fig. 4 calculated from model II: one is using the correct form of (7) (model II), the other is using a deliberately altered form of (7) with the term $-c_p T J_{\text{sub}}$ in the numerator changed to $c_p T J_{\text{sub}}$ (model II′), i.e., this term is switched from a source term to a sink term for the particle temperature. The reason for changing the sign of this term in model II′ in the simulation is given later in the discussion of high fluence results. It is evident that model II predicts a lower peak particle temperature than model I, by about 108 K, which is anticipated due to the appearance of a positive term $T\,\mathrm{d}c_p/\mathrm{d}T$ in the denominator of (7). For the very same reason, the decay rate of particle temperature predicted by model II is slower than that predicted by model I. The difference between the results of model I and model II are qualitatively similar to those reported by Hofmann et al. [16, 18]. Due to the very low sublimation rate in this low fluence case, results of model III is almost identical to those of model I. These numerical results are consistent with the discussions given earlier, i.e, model III reduces to model I while model II does not in the limit of very low laser fluences. A closer look at Fig. 4 indicates that the particle temperatures predicted by model III is slightly higher than those by model I, since the term $-u J_{\text{sub}}$ (playing the same role as $-c_p T J_{\text{sub}}$ in model II) acts as a source term in the particle energy equation. Results of model II′ in this case are also very close to, but slightly lower than, those from model II, again due to the very low sublimation rate.

3.2 *High fluence case*

Results of the high fluence case are compared in Figs. 5 and 6 for the particle temperature and diameter, respectively. In this high fluence case, the peak particle temperatures predicted by these models are respectively 4743 K (model I), 5304 K (model II), 4642.6 K (model II′), and 4990.7 K

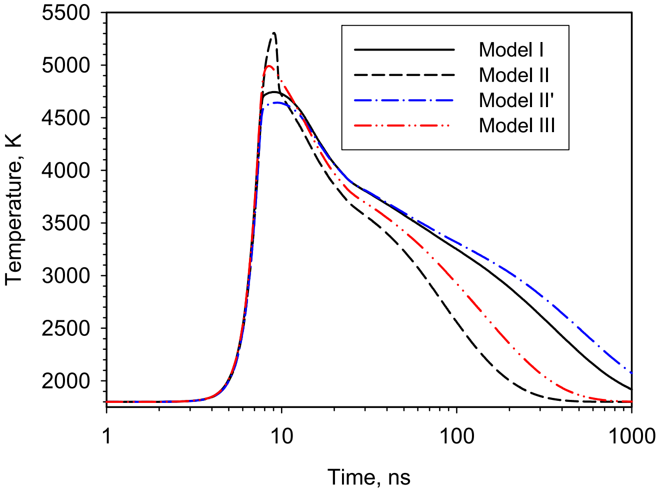

FIGURE 5 Effect of the particle internal energy variation rate expression on the calculated particle temperature at a high laser fluence of 7 mJ/mm^2

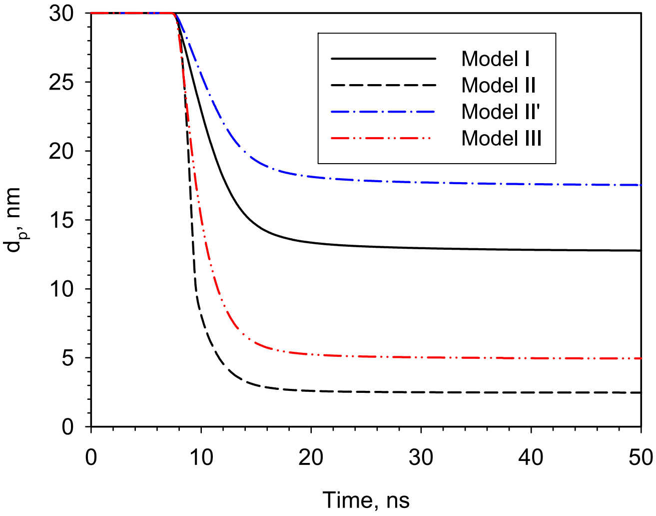

FIGURE 6 Effect of the particle internal energy variation rate expression on the calculated particle diameter at a high laser fluence of 7 mJ/mm^2

(model III). The peak temperature from model II is about 560 K higher than that from model I. The higher peak particle temperature from model II at high fluences is consistent with the analysis presented earlier. However, it is qualitatively different from those reported by Hofmann et al. [16, 18], who obtained lower peak particle temperatures using their ‘correct’ particle internal energy variation rate expression. As an attempt to understand the difference between the results of model II and those reported in [16, 18], the sign of the term $-c_p T J_{\mathrm{sub}}$ in the numerator of (7) is deliberately changed to $c_p T J_{\mathrm{sub}}$ as in model II′. With this change, the peak particle temperature predicted by model II′ is about 100 K lower than that by model I. Such a difference between the peak particle temperatures predicted by model II′ and model I is in qualitative and even reasonably quantitative agreement with that documented in [16]. This exercise suggests that Hofmann et al. [16, 18] implemented the term $-c_p T J_{\mathrm{sub}}$ in the numerator of their particle energy equation, (7), incorrectly. Otherwise, there seems no other ways to explain the results of Hofmann et al. presented in [16, 18].

The peak temperature from model III is about 248 K higher than that from model I. The higher peak particle temperature at high fluences predicted by models II and III are actually expected, since the term $-c_p T J_{\mathrm{sub}}$ in model II or the term $-u J_{\mathrm{sub}}$ in model III acts as a source term to enhance the particle temperature. As mentioned earlier, such a source term in the particle energy equation results in unrealistically higher peak particle temperature. The non-equilibrium soot particle temperature reported by Hiers [20] for soot undergoing oxidation should be treated with caution since the results were based on an incorrect particle energy equation.

As a result of much higher particle temperatures predicted by models II and III, the particle diameters predicted by these models decrease much faster than that predicted by model I, Fig. 6. The particle diameter history from model II′ shown in Fig. 6 is also very close to that obtained by the Hofmann model [16], further support the conjecture that Hofmann et al. [16, 18] implemented the term $-c_p T J_{\mathrm{sub}}$ incorrectly.

4 Conclusions

The different expressions for the internal energy variation rate of a small particle, subject to mass loss used in laser-induced incandescence modelling were evaluated. Through the application of the first law of thermodynamics to an open system, the derivation of the energy conservation equation for a small sublimating particle was presented. It was found that the commonly used expression for the particle internal energy variation rate in laser-induced incandescence studies is correct, while the more recent expressions suggested by K&H for laser-induced incandescence studies, and by Hiers for soot particle combustion are actually incorrect. These incorrect expressions for the particle internal energy variation rate lead to a physically erroneous high peak particle temperature at high laser fluences due to the introduction of a nonphysical source term in the energy equation. Accordingly, these incorrect expressions predict a much smaller particle diameter at the end of the laser pulse due to overestimation of the sublimation rate. As a result of using an inconsistent specific particle internal energy definition, the expression for the particle internal energy variation rate of K&H does not recover the correct expression in the regime of low laser fluences where the sublimation rate vanishes. Instead, their expression predicts a lower peak particle temperature and a slower particle temperature decay rate after the laser pulse. Based on the numerical evidence provided in this study, it is very likely that Hofmann et al. implemented the nonphysical energy source term, associated with the mass loss in the particle energy equation, incorrectly in their numerical calculations.

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