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Abstract

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ON REGULARITIES OF THE TEMPERATURE RADIATION OF A FLAME

(Presented by Academician N. N. Semenov, 30 X 1959)

According to experimental data, the laws of Wien, Stefan–Boltzmann, and others, often used in temperature measurements, are far from always obeyed under flame conditions ([1–5]). Theoretical treatments of the question are mainly limited to general propositions concerning the blackness of flames ([6–11]).

Let us carry out a calculation for a specific, narrowly defined model:

1) To the flame we apply the principle of "point-instantaneous" equilibrium, i.e., at each "point" of the flame, for its "instantaneous" temperature $T(x, y, z, t)$, the Gibbs distribution of molecules, atoms, and electrons in the gas is obeyed (and of electrons inside the solid inorganic particles present in the flame), and, consequently, also the Kirchhoff–Planck law

$$B_\nu(x, y, z, t) \equiv \frac{\eta_\nu(x, y, z, t)}{\alpha_\nu(x, y, z, t)} = \frac{2h\nu^3}{c^2}\left[\exp\left(\frac{h\nu}{kT(x, y, z, t)}\right) - 1\right]^{-1}; \quad (1)$$

where η_ν and α_ν are the mass coefficients of emissive and absorptive capacity of the medium; h is Planck' s constant; ν is the frequency; c is the speed of light; k is Boltzmann' s constant.

2) The flame is regarded as a certain continuum with density $\rho(x, y, z, t)$, and the equation of radiative transfer without allowance for scattering of light is used

$$dI_\nu/d\tau_\nu = B_\nu - I_\nu, \quad (2)$$

where I_ν is the intensity of monochromatic radiation; $\tau_\nu = \int \alpha_\nu \rho\, ds$ is the optical path of the ray; s is the geometrical path of the ray.

3) The concentration of solid inorganic particles in the flame is small, and the density of the continuum obeys the Clapeyron equation

$$P(t)M(x, y, z, t) = \rho(x, y, z, t)RT(x, y, z, t), \quad (3)$$

where P is the pressure; M is the molecular weight of the medium; R is the gas constant.

If each component of the gas mixture emits its own radiation (for example, CO_2 and H_2O), then, obviously, equations (1), (2), and (3) are written separately for each component, and in the subsequent formulas the pressure P denotes the partial pressure of the given component, and M its molecular weight.

4) For a flame, averaging over frequency of the quantities α_ν and τ_ν is physically justified (for example, $\lg \frac{\alpha_{\nu\max}}{\alpha_{\nu\min}} \ll 1$, according to the data of V. M. Mal' tsev and P. F. Pokhil).

5) The quantities η_ν, α_ν, T, ρ, and M vary within the investigated region of the flame not very strongly, and, for the characterization of the latter as a whole, their averaging over coordinates is physically justified.

The first limitation, being the most fundamental and stringent, is not fulfilled for all flames. It means that the inequalities $\tau_p \sim \tau_{\text{phys}} \ll \delta t \ll \tau_{\text{voz}}$, $l_p \ll \delta x,\ \delta y,\ \delta z \ll l_{\text{izm}}$ are satisfied, where τ_p is the time for the establishment of the equilibrium distribution of particles (molecules, atoms, and electro-

ones); τ_{rad} is the time for establishing equilibrium radiation; τ_{pert} is the characteristic time of the perturbing processes (chemical reactions, emission, etc.); l_p is the particle mean free path; l_{meas} is the characteristic length over which the temperature changes appreciably. In other words, in each volume $\delta V = \delta x\,\delta y\,\delta z$, during each time interval δt, a **local-time equilibrium** is fully established, since in the phase subsystem $(\delta V,\ \delta t)$ statistical laws are already applicable and the Gibbs distribution of particles over energies is obeyed, but the influence of perturbing factors and of the temperature gradient has not yet made itself felt. The indicated inequalities mean quasi-closure of the phase subsystem $(\delta V,\ \delta t)$, when the chemical and radiative nonequilibrium of the whole system only weakly affects the behavior of the subsystem, i.e., the number of collision events between particles without chemical transformations and without emission or absorption of light is much greater than the number of chemical-transformation events and the number of events of emission and absorption of light. However, for mathematical treatment it is more convenient to pass to the limit $\delta V,\ \delta t \to 0$, as if **point-instantaneous** equilibrium were realized.

In the absence of external light sources, the solution of equation (2) has the form $I_\nu = \varepsilon_\nu \bar{B}_\nu$, $\varepsilon_\nu = 1 - \exp(-\tau_\nu)$, where ε_ν is the selective degree of blackness of the flame.

The optical path τ_ν, according to (3), is equal to

$$\tau_\nu = \int \alpha_\nu \rho\, ds = \int \alpha_\nu \left(\frac{PM}{RT}\right) ds = \frac{\bar{\alpha}_\nu P \bar{M} s}{R\bar{T}},$$

i.e.,

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$$\varepsilon_\nu = 1 - \exp\left(-\frac{\bar{\alpha}_\nu P\bar{M}s}{R\bar{T}}\right),$$

$$I_\nu = \left[1 - \exp\left(-\frac{\bar{\alpha}_\nu P\bar{M}s}{R\bar{T}}\right)\right]\frac{2h\nu^3}{c^2}\left[\exp\left(\frac{h\nu}{kT}\right) - 1\right]^{-1}, \tag{4}$$

(here and below the overbar denotes averaging of the quantity over the coordinates).

The wavelength λ_m of maximum intensity in the spectral distribution of the flame light is determined from the condition

$$\frac{dI_\lambda}{d\lambda} = \left[\frac{d}{d\lambda}\left(\frac{I_\nu d\nu}{d\lambda}\right)\right] = 0. \tag{5}$$

Using (4) and (5), we obtain $\left(\theta = k\bar{T}/h\nu = k\lambda\bar{T}/hc\right)$

$$\frac{\exp\tau_{\lambda_m} - 1}{\tau_{\lambda_m}} = \frac{\theta_m^2[1 - \exp(-1/\theta_m)]}{5\theta_m[1 - \exp(-1/\theta_m)] - 1}\frac{1}{\tau_{\lambda_m}}\left(\frac{\partial\tau_{\lambda_m}}{\partial\theta}\right)_{\lambda_m} \tag{6}$$

or

$$\lambda_m\bar{T} = \frac{hc}{k}\theta_m(\tau_{\lambda_m}) = f\left(\frac{\bar{\alpha}_{\lambda_m}P\bar{M}s}{R\bar{T}}\right),$$

i.e., for a flame the product $\lambda_m\bar{T}$ is not a constant quantity, but a function of the experimental conditions. To determine it analytically, it is necessary to know the dependence $\tau_\lambda(\theta)$, which differs for different gases. Thus, for gases with hydrogen-like atoms, according to the quantum-mechanical model ([12]), $\tau_\lambda \sim \theta^3[1 - \exp(-1/\theta)]$.

Hence we obtain the function $\theta_m(\tau_{\lambda_m})$, determined by the transcendental equation

$$\frac{\exp\tau_{\lambda_m} - 1}{\tau_{\lambda_m}} = \frac{(3\theta_m + 1)[1 - \exp(-1/\theta_m)] - 1}{5\theta_m[1 - \exp(-1/\theta_m)] - 1}. \tag{7}$$

The solution of equation (7) can be tabulated:

τ_{λ_m}	0.000	0.500	1.000	1.500	2.000	2.500	3.000	3.500	4.000	4.500	5.00	∞
$\theta(\tau_{\lambda_m})$	0.500	0.378	0.313	0.247	0.250	0.234	0.223	0.216	0.211	0.208	0.206	0.201
$\lambda_m T$	0.715	0.541	0.448	0.392	0.358	0.335	0.319	0.309	0.302	0.298	0.295	0.288

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Let us express analytically two limiting cases:

1) Optically thin flame ($\tau_{\lambda_m} \ll 1$):

$$\lambda_m \overline{T} = \left(0.715 - 0.961 \frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}}\right) \text{ cm} \cdot \text{deg}. \tag{8}$$

Use of Wien' s law would lead to an underestimate of the measured temperature by a factor of ~ 2.5. However, formula (8) is valid if the spectrum remains continuous. In fact, the spectra for optically thin flames in most cases have a line-band structure, i.e., one observes a series of characteristic wavelengths $\lambda_{\max}$ not obeying Wien's law, where the light-intensity distribution has corresponding maxima.

2) Optically thick flame ($\tau_{\lambda_m} \gg 1$):

$$\lambda_m \overline{T} = \left[0.288 + 0.171 \frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}} \exp\left(-\frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}}\right)\right] \text{ cm} \cdot \text{deg}. \tag{9}$$

Wien' s law is obeyed only in the limiting case of an optically thick flame, $\lambda_m \overline{T} \to 0.288$ cm $\cdot$ deg as $\tau_{\lambda_m} \to \infty$.

For gases with non-hydrogen-like atoms, formulas (7), (8), and (9) are somewhat more complicated, but their fundamental character is preserved:

$$\lambda_m \overline{T} = f_1(\theta_m) + f_2(\theta_m) \frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}} \left[\exp\left(\frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}}\right) - 1\right]^{-1}.$$

Thus, for a more general dependence $\tau_\lambda \sim \theta^\beta [1 - \exp(-1/\theta)]$, where $\beta = \text{const} \leq 0$, and $[1 - \exp(-1/\theta)]$ is the Rosseland factor reflecting Einstein' s negative absorption of light, we obtain the tabulated function $\theta_m(\tau_{\lambda_m}, \beta)$ satisfying the equation

$$\frac{\exp \tau_{\lambda_m} - 1}{\tau_{\lambda_m}} = \frac{(\beta \theta_m + 1)[1 - \exp(-1/\theta_m)] - 1}{5\theta_m [1 - \exp(-1/\theta_m)] - 1}. \tag{10}$$

From this follow two limiting cases:

1) For $\tau_{\lambda_m} \ll 1$

$$\lambda_m \overline{T} = \frac{hc}{k} \theta_m = \frac{1.43}{5 - \beta} \left[1 - \left(\frac{5}{5 - \beta} - \frac{1}{1 - \exp(\beta - 5)}\right) \frac{\overline{a}_{\lambda_m} P \overline{M} s}{R \overline{T}}\right] \text{ cm} \cdot \text{deg}.$$

2) For $\tau_{\lambda_m} \gg 1$

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$$\lambda_m \overline{T} = \frac{hc}{k}\theta_m =$$

$$= \left[0.288 + (\beta \cdot 0.0576 - 0.0020)\frac{\overline{a}_{\lambda_m} P\overline{M}s}{R\overline{T}} \exp\left(-\frac{\overline{a}_{\lambda_m} P\overline{M}s}{R\overline{T}}\right)\right] \text{ cm} \cdot \text{deg}.$$

Next, the flux of monochromatic radiation of the flame is $H_\nu = \int_\Omega I_\nu d\Omega$, where Ω is the solid angle. In measurements of flame temperature, narrow-aperture beams of light are usually used, i.e.,

$$H_\nu \simeq I_\nu \Delta\Omega = \left[1 - \exp\left(-\frac{\bar{a}_\nu P\bar{M}s}{R\bar{T}}\right)\right] \frac{2h\nu^3}{c^2} \left[\exp\left(\frac{h\nu}{kT}\right) - 1\right]^{-1} \Delta\Omega. \quad (11)$$

Averaging the selective degree of blackness of the flame over frequency, for the total radiation flux we obtain

$$H = \int_0^\infty H_\nu d\nu = \left[1 - \exp\left(-\frac{\bar{a}P\bar{M}s}{R\bar{T}}\right)\right] \frac{\Delta\Omega}{\pi}\sigma T^4, \quad (12)$$

where σ is the Stefan–Boltzmann constant.

The logarithmic temperature coefficient is equal to

$$a_T = \frac{\partial \ln H}{\partial \ln T} = 4 - \frac{\bar{a}P\bar{M}s}{R\bar{T}} \left[\exp\left(\frac{\bar{a}P\bar{M}s}{R\bar{T}}\right) - 1\right]^{-1} = 4 - a.$$

Similarly, the remaining logarithmic coefficients are equal to

$$a = \frac{\partial \ln H}{\partial \ln \bar{a}} = \frac{\partial \ln H}{\partial \ln P} = \frac{\partial \ln H}{\partial \ln M} = \frac{\partial \ln H}{\partial \ln s} = \frac{\bar{a}P\bar{M}s}{R\bar{T}} \left[\exp\left(\frac{\bar{a}P\bar{M}s}{R\bar{T}}\right) - 1\right]^{-1}.$$

Thus, in the limiting case of an optically dense flame, the influence of pressure, structure, and geometry of the flame is ultimately eliminated, and the Stefan–Boltzmann law is obeyed.

In the literature, Schack' s empirical formulas ([1,2]) are usually cited, with power-law dependences $H = \text{const} \cdot P^{a_P} s^{a_S} T^{a_T}$, where a_P, a_S, and a_T are empirically selected quantities, constant within a narrow range of conditions $(P, s, \bar{T})$. Thus, for the radiation flux of CO_2 and H_2O vapor at $\bar{T} \simeq 700$—800°K, Schack, who

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unfortunately did not investigate the influence of all parameters, selected the dependences: 1) for CO_2

$$H \simeq 3.5(Ps)^{0.33}\left(\frac{T}{100}\right)^{3.5}\frac{\text{kcal}}{\text{m}^2\text{h}};$$

2) for H_2O

$$H \simeq 3.5\,P^{0.80}s^{0.60}\left(\frac{T}{100}\right)^{3}\frac{\text{kcal}}{\text{m}^2\text{h}}.$$

As shown above, the dependences agree well with formula (12).

Deviations from the Stefan–Boltzmann law under conditions of an optically non-dense flame introduce significant corrections into the formulas used in determining the radiation, brightness, and color temperatures of a flame.

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Note: Figure translations are in progress. See original paper for figures.

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