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A CORRELATION FOR TOTAL BAND ABSORPTANCE OF RADIATING GASES*

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NOMENCLATURE

a,	dimensionless constant defined in equation (10);
A,	total band absorptance defined in equation (1) [cm^{-1}];
A_0,	spectroscopic constant [cm^{-1}];
$\bar{A}$,	dimensionless total band absorptance defined in equation (3);
B^2,	spectroscopic constant [$(\text{atm})^{-n}$];
C_0^2,	spectroscopic constant [cm^2/g];
f_1, f_2,	dimensionless functions defined in equation (2);
k,	spectral absorption coefficient [cm^2/g];
L,	mean beam length [cm];
P^n,	equivalent broadening pressure [$(\text{atm})^n$];
r,	geometric beam length [cm];
t,	dimensionless equivalent broadening pressure defined in equation (3);
u,	dimensionless mass path length defined in equation (3);
w,	mass path length [g/cm^2].

Greek symbols

ν,	wave number [cm^{-1}];
ϕ,	angle [deg];
Ω,	solid angle [steradian].

INTRODUCTION

The calculation of radiant energy transfer in bounded and unbounded systems requires a description of gaseous band absorption and emission phenomena. For practical purposes, it is often convenient to use the total or integrated band absorptance, A, defined as

$$A = \int_{\Delta\nu} A(\nu)\,\mathrm{d}\nu = \int_{\Delta\nu} (1 - e^{-k(\nu)w})\,\mathrm{d}\nu \qquad (1)$$

where $A(\nu)$ is the spectral absorptance, $\Delta\nu$ the effective band width, $k(\nu)$ the spectral absorption coefficient and w the mass path length. The spectral absorption coefficient $k(\nu)$ is, in general, a very rapidly varying function of frequency, and hence exact integration of equation (1) over a complex vibration–rotation band is formidable.

In one of the first attempts to describe total band absorptance analytically, Schack [1] approximated the spectral absorption coefficient as a function of reordered wavelength by a straight line and by a series of straight lines. The result for the straight-line approximation is a simple expression involving two correlation constants (two-parameter model), and is not a function of pressure. Penner [2] has pointed out that this approximation is of little value since it is valid only at high pressures where substantial overlapping of rotational lines occurs. Edwards and Menard [3] have also mentioned that as the mass path length becomes large, Schack's expression approaches a constant, instead of the logarithmic asymptote indicated by experimental results [4–7].

Much of the experimental band absorptance data available to date has been correlated by using linear, square root,

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and logarithmic functions of mass path length at small, intermediate and large values of mass path length respectively [4–7]. The range of applicability for each function, however, is uncertain and these correlations are not smoothly joined in the transition regions. To eliminate these difficulties, Edwards and Menard [3] have proposed a correlation procedure based on the statistical model of spectral lines and an assumed model of reordered frequency dependence of spectroscopic parameter (the exponential-wide-band model). Their correlation represents a continuous smooth function extending from a linear to a square root and to a logarithmic path length dependency. In later papers [8–10] they have employed this procedure to correlate successfully the total band absorptance for various bands of carbon monoxide, carbon dioxide, methane and water vapor. There exists, however, one drawback in this correlation, that it does not have a single explicit expression for the whole path-length range.

The present paper is to establish a single continuous explicit expression for the total band absorptance. This would provide not only a more convenient and direct way for correlation of experimental data [11], but also a formula highly desirable in many radiation calculations, such as the calculation of mean beam length by use of the total band absorption laws [12, 13].

A SINGLE CONTINUOUS CORRELATION

The exponential-wide-band model of Edwards yields the linear, square-root and logarithmic relations for small, intermediate and large path lengths, respectively. In dimensionless forms, they are

$$\bar{A} = u, \qquad \bar{A} = (ut)^{\frac{1}{2}} + f_1(t), \qquad \bar{A} = \ln\left[u f_2(t)\right] \quad (2)$$

where

$$\bar{A} \equiv A(A_0), \qquad u \equiv C_0^2 w, \qquad t \equiv B^2 P^n. \quad (3)$$

A_0, C_0^2, B^2 are correlation constants related to the basic spectroscopic parameters and are functions of temperature, P^n is the equivalent broadening pressure and $f_1(t)$ and $f_2(t)$ are certain functions of t [3]. The above relations agree well with those established from experimental results and analytical models except that a slightly different relation (square-root logarithmic relation) has been observed in the large path length region of the rigid-rotator model [3].

To construct a single continuous function $\bar{A}(u, t)$ for all values of u and t, the mathematical properties of such a function must be first specified. The function must possess the two asymptotes,

$$\lim_{u \to 0} \bar{A}(u, t) = u \quad (5)$$

$$\lim_{u \to \infty} \bar{A}(u, t) = \ln\left[u f_2(t)\right] \quad (6)$$

In addition, it has been shown recently that all band absorptance laws have certain fundamental properties in common [14]. Total band absorptance must be a positive, monotonically increasing function of mass path length

$$\bar{A}(u, t) \geqslant 0 \qquad \bar{A}_u(u, t) > 0 \quad (7)$$

and it must have its maximum slope at the optically thin limit,

$$\bar{A}_u(0, t) \geqslant \bar{A}_u(u, t) \quad (8)$$

where subscript u refers to the derivative with respect to u. One further restriction which must be placed on any proposed correlation based on the statistical or Elsasser models is that it must not possess inflection points,

$$\bar{A}_{uu}(u, t) \leqslant 0. \quad (9)$$

This criterion can be easily shown by differentiating the mean spectral absorptance given by these models twice.

A class of functions which satisfy all the requirements given in equations (5) to (9) are

$$\bar{A}(u, t) = \ln\left\{u f_2(t)\left[\frac{u + a}{u + a f_2(t)}\right] + 1\right\}, \qquad (a \geqslant 2) \quad (10)$$

No proof has been made on the uniqueness of this class of functions, and it is possible that there exist other functions which also satisfy the given requirements. It is felt, however, that the possible lack of uniqueness of equation (10) is of little practical importance if successful correlations with experimental data can be established from it. The value of a and the functional form of $f_2(t)$ will be chosen by comparing with the established correlation of experimental data. Since Edwards' correlation has been highly successful in correlating existing data of various vibration–rotation bands [8–10], attempt is made to determine a and $f_2(t)$ from the comparison with it.

COMPARISON WITH EDWARDS' CORRELATION

In Edwards' correlation, the function $f_2(t)$ in the logarithmic relation is given as

$$f_2(t) = t\,e^{2-t} \qquad (t \leqslant 1) \quad (11)$$

and

$$f_2(t) = e \qquad (t \geqslant 1) \quad (12)$$

where e is the base of the natural logarithm. The limiting relation at $t = 1$ implies that the spectral lines are overlapped to such an extent that the total band absorptance is no longer a function of pressure. It is interesting to note that Edward's correlation is a three-parameter characterization (in contrast to Schack's two-parameter characterization) of total band absorptance and reduces to a two-parameter one as the equivalent pressure becomes sufficiently large ($t \geqslant 1$).

If the function $f_2(t)$ as given in equation (11) and (12) is used in equation (10), it is found that among all values of a, $a = 2$ would give the best fit to Edwards' correlation. The comparison is shown in Fig. 1. The linear and logarithmic asymptotes are identical in the two correlations but the values given by equation (10) in the square-root region are sometimes 20 to 30 per cent higher. In general, the agreement is quite satisfactory.

Better agreement with Edwards' correlation may be achieved by employing various forms of $f_2(t)$. This is permissible here because no mathematical restrictions have been imposed on the pressure-dependent function $f_2(t)$ in the present development. Indeed, a single continuous function

$$f_2(t) = 2{\cdot}94\,[1 - \exp(-2{\cdot}60\,t)] \qquad (13)$$

has been found to give an excellent numerical agreement with the function given in equations (11) and (12), for the whole range of t, $0 < t < \infty$. Physically, equation (13) offers a more reasonable pressure-dependent function in that

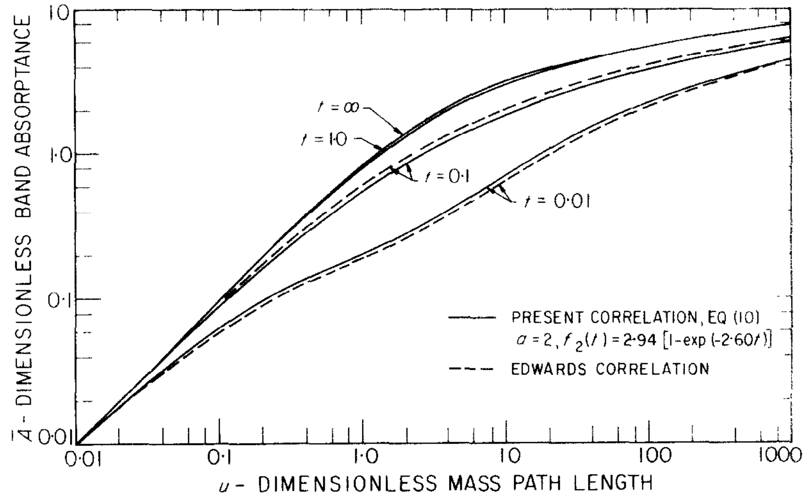

FIG. 1. Comparison between the present correlation [with $f_2(t)$ given by Edwards] and Edwards' correlation

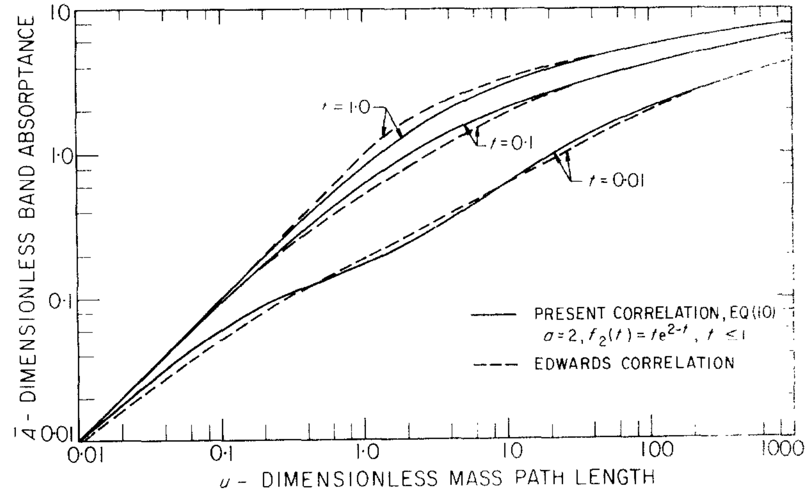

FIG. 2. Comparison between the present correlation [with $f_2(t)$ given by equation (13)] and Edwards' correlation.

it accounts for an increase, although small, in absorptance due to a pressure increase in the region $t > 1$. In the limit as $t \to \infty$, equation (10) reduces to a two-parameter characterization of total band absorptance. The correlation for various values of t is shown in Fig. 2, along with Edwards' correlation. The agreement is very good.

It should be pointed out that other values of $a(a > 2)$ may yield results in good agreement with correlations established from other band models of spectral lines. But these correlations must be confirmed by experimental data as in the case of Edwards' correlation.

CONCLUDING REMARKS

Certain interesting observations on the total band absorptance can be made as a result of the remarkable, if not too surprising, agreement between the present correlation and Edwards' correlation. It should be emphasized that Edwards' correlation is based on the statistical model of spectral lines and an exponential-wide-band model of reordered frequency dependence of basic spectroscopic constants. The present correlation, on the other hand, has little dependence on the detailed structure of a band, and is derived from a set of common mathematical properties of total band absorption. But the success of the present correlation still relies on the comparison with the result from the consideration of detailed structures, such as the determination of a from Edwards' correlation. The difference in these two approaches is quite analogous to that in the microscopic and macroscopic derivations of a thermodynamic formula.

Use of the proposed correlation has been recently made to correlate the infrared absorption data of carbon monoxide for a wide optical range [11]. In addition to its success of correlating data, it should be emphasized that this correlation, in contrast to any previous attempt, requires no arbitrary division of experimental data into various optical regions.

Application of the proposed correlation to the calculation of mean beam length may appear unnecessarily difficult because of the integration involved in such a calculation [13, 14]:

$$A(L) = \frac{1}{\pi}\int_{\Omega} A(r)\cos\phi \,\mathrm{d}\,\Omega \tag{14}$$

where r is the distance from the differential element to the confining surface, ϕ the angle between r and the normal to the differential element, and Ω the solid angle subtended by the gas volume. Indeed, simple functions (also simple to integrate) such as power-law and logarithmic correlations are available for respective optical regions. It should be realized, however, that for gas bodies with optical beam lengths (the product of geometric beam length r and partial pressure of the radiating gas) falling into a wide optical range, a single explicit function is needed for the expression of $A(L)$ over the whole ranges of Ω, even though it is always possible to split the integral on the right-hand side of equation (14) over different ranges of Ω in order to use simple absorption correlations. For the particular case that all optical beam lengths fall into one optical region (weak or strong), the linear and logarithmic asymptotic expressions of the proposed correlation can be used directly for calculation, just as they have been suggested in previous works [12, 13].

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