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MODELS FOR THE ANALYSIS OF CN VIOLET RADIATION BEHIND SHOCK WAVES IN AIR CONTAMINATED WITH CARBON-BEARING COMPOUNDS

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SUMMARY

Techniques for analyzing the effects of contamination radiation from the CN violet band system behind normal shock waves are presented. The problem is approached in two ways: (1) a simple chemical system is set up for a known contaminant, and (2) a computer program which utilizes a free-energy minimization technique is used to evaluate the CN particle concentration for given atom percentages of carbon. Equilibrium radiation from CN and air with various contamination levels is determined for shock velocities ranging from 20 000 to 44 000 ft/sec (6.10 to 13.41 mm/μsec) at free-stream pressures of 0.1 and 1 mm Hg. Radiation from CN in the chemical relaxation zone behind a normal shock with C_2N_2 as the free-stream contaminant is also considered. Results of the calculations are presented graphically and discussed.

The results indicate that radiation from contaminated air in the velocity range between 20 000 and 30 000 ft/sec (6.10 and 9.14 mm/μsec) may be dominated by small contamination levels. However, at speeds greater than about 33 000 ft/sec (10.06 mm/μsec), relatively high contamination levels can be tolerated for equilibrium lows because the contamination radiation is minor in comparison with air radiation. The alculations for the nonequilibrium condition further indicate that the ratio of CN radiaion to radiation from air in the chemical relaxation zone is comparable to the ratio for he equilibrium condition, and that contamination radiation probably presents no greater problem in the nonequilibrium region than in the corresponding equilibrium region.

INTRODUCTION

Carbon-bearing compounds in the presence of nitrogen at elevated temperatures are generally involved in a rather complex chemistry. For temperatures greater than about 3000° K, one of the byproducts is the cyano radical CN, which gives rise to a relatively intense radiation in the near-ultraviolet and visible-violet region of the spectrum. Very often, conditions are such that the carbon-bearing contaminants are difficult to remove from the system, and the CN violet radiation becomes a severe hindrance to spectrographic studies (refs. 1, 2, and 3).

The specific purposes of this report are to investigate the behavior of the CN contamination radiation for a fairly wide range of working conditions, and to provide an aid in defining tolerable free-stream contamination levels for radiation heat-transfer studies in shock tubes and expansion tubes. The problem of fulfilling these purposes has been approached in two ways. In the first method, the production of CN behind a normal shock is assumed to arise from small amounts of cyanogen C_2N_2 in the free stream. Calculations for both equilibrium and nonequilibrium conditions have been made for this assumption. A second method allows the shocked gas to contain a specified ratio of carbon atoms to air atoms and uses the minimum of free energy to define the resulting chemical equilibrium. The results of these two types of calculations are presented for various amounts of contaminant and over a range of shock speeds from 20 000 to 44 000 ft/sec (6.10 to 13.41 mm/μsec). These results should be an aid in analyzing observed contamination radiation from CN and will be indicative of the effects to be expected from a given contamination level.

SYMBOLS

F	radiative flux, W/cm^2
h	Planck constant, 6.63×10^{-34} J-sec
j	emission coefficient, W/cm^3-4π sr
k_f, k_b	forward and reverse chemical rate constants, cm^3/mole-sec
K_1	chemical equilibrium constant, $\frac{n_{CN}^2}{n_{C_2N_2}}$, moles/cm^3
K_2	chemical equilibrium constant, $\frac{n_C n_N}{n_{CN}}$, moles/cm^3
L	shock layer thickness, cm
M	constant, defined by equation (1), moles/g
n	molar concentration, moles/cm^3
p	pressure, mm Hg

r	nose radius, cm
t	time, sec
T	temperature, ^{o}K
V_s	shock speed, ft/sec (mm/μsec)
x	mole fraction
δ	shock detachment distance, cm
μ_C	number of carbon atoms per molecule
μ_u	number of unspecified atoms per molecule
ν	frequency of photon, Hz
ξ,y	distance behind shock front, cm
ρ	density, g/cm^3
ϕ	atom fraction of carbon

Subscripts:

1	free-stream conditions
2	conditions behind shock wave
o	standard atmospheric conditions
eq	equilibrium conditions

An asterisk (*) indicates an electronically excited state.

CALCULATIONS AND RESULTS

Determination of Radiance of CN Violet Band System

For a given number of CN particles whose excited states are populated according to a known gas temperature, the mean radiation per molecule for a particular transition may be determined from the oscillator strength, or f-number, of the transition along with the spectroscopic constants pertaining to the levels involved. For temperatures above approximately 4500° K, the CN violet band system resulting from the transition $B\,^2\Sigma \rightarrow X\,^2\Sigma$ is the predominant radiation from this molecule, with major band progressions located at 4216 Å, 3883 Å, and 3590 Å. The calculation of the radiance per molecule as a function of temperature for this band system was performed by Henry T. Woodward at Ames Research Center, using a computer program similar to that described in reference 4. This type of calculation is commonly referred to as the "smeared rotational line" model, and has been shown to be a very good approximation to the more refined single-line calculation when the band system is reasonably compact (ref. 5). The radiance of CN violet used in this paper is graphically presented in figure 1. It corresponds to an f-number of 0.020 and a heat of formation of CN of 8.2 eV.

Chemistry of CN Behind Normal Shock Waves

In certain radiation experiments performed in the Ames combustion-driven expansion tube, high levels of CN violet radiation were observed (ref. 1). This radiation occurred when mylar and teflon diaphragms were employed and indicated that foreign substances from the diaphragms containing carbon appeared in the test gas. In one particular case when air was used as the test gas, the total gas-cap radiation was measured as 23 watts for a model in a flow of speed 7.33 mm/μsec at $\rho_1/\rho_0 = 4.1 \times 10^{-3}$. For a ballistic range test under similar conditions ($V_s = 7.52$ mm/μsec at $\rho_1/\rho_0 = 4.1 \times 10^{-3}$), gas-cap radiation was observed to be 2.4 watts. This value is essentially a decade below that obtained in the expansion tube. From spectral analysis of the expansion-tube radiation, one-third of the integrated spectrum was estimated to be due to the CN violet band system, while most of the remainder consisted of atomic line radiation from metallic elements. Under these test conditions, the mole fraction of CN required to produce the radiation attributed to the violet band system is 1.8×10^{-3}.

Since a certain amount of contamination in the free stream may be unavoidable, it is of interest to learn the effects of CN for a range of operating conditions. One approach to the analysis is to allow the formation of CN behind a normal shock to result from a known amount of C_2N_2 contamination in the free stream. It is further assumed that the state variables of the shocked gas attain the same equilibrium values as for pure air. Normal-shock tables were used to determine these values (ref. 6). For a first approximation to the chemistry of the contaminant, the reactions considered were merely the formation and dissociation of CN:

$$C_2N_2 \overset{K_1}{\rightleftarrows} 2CN$$

$$CN \overset{K_2}{\rightleftarrows} C + N$$

The equilibrium constants, defined as

$$K_1 = \frac{n_{CN}^2}{n_{C_2N_2}}$$

and

$$K_2 = \frac{n_C n_N}{n_{CN}}$$

are functions of temperature only and may be computed from partition functions or tabulations of the free energy. The equilibrium constants, along with the conservation relation for carbon atoms

$$\frac{1}{\rho}\left(2n_{C_2N_2} + n_{CN} + n_C\right) = \text{Constant} \equiv M \tag{1}$$

may be used to evaluate the CN concentration at a given temperature and density as follows:

$$n_{CN} = \frac{-\left(K_2 + n_N\right) + \sqrt{\left(K_2 + n_N\right)^2 + \frac{8\rho M n_N^2}{K_1}}}{\frac{4n_N}{K_1}}$$

$$\approx \frac{\rho M n_N}{K_2 + n_N} \qquad \left(\frac{8\rho M}{K_1} << 1\right)$$

The results of this approach are illustrated in figures 2 and 3 for free-stream mole fractions of C_2N_2 of 0.0001, 0.001, and 0.005 for values of p_1 of 0.1 and 1 mm Hg with a free-stream temperature of 300° K. The concentration of nitrogen atoms behind the shock was assumed to have its pure-air value and was determined from equilibrium chemical properties of high-temperature air (ref. 7). Values of air radiation used in the figures were taken from Allen (ref. 8). Allen does not give the emission coefficient directly but plots the total radiative flux from one side of an infinite air slab of 1-cm thickness, taking absorption effects into account. For an optically thin gas, the flux F from an infinite slab is given by

$$F = \frac{1}{2} Lj$$

where L is the slab thickness. Since this relationship was used in obtaining the emission coefficient for air in figures 2 and 3, some inaccuracy exists in the air curves at conditions for which absorption effects are significant. These assumptions and approximations overestimate the CN concentration because the reactions involving CO, O, and the ionization of C have been neglected. Such an approach is admittedly only approximate, but this method of calculation is extremely simple and does show the trend to be expected at higher velocities, namely that the CN radiation becomes negligible as V_s increases.

The problem was approached in a different way by utilizing a computer program which determines the chemistry behind the shock by minimizing the free energy of the system. It was assumed that a small percentage of an unknown carbon-bearing compound was present in the free stream but had no effect on the cold-gas properties. A value was given to the ratio of carbon atoms to the total number of atoms in the gas, and the reaction chemistry of the carbon behind the shock was determined according to the free-energy minimization process. This calculation procedure is physically equivalent to a model in which the carbon-bearing contaminant is dissociated into its atoms with no energy drain from the shock wave and in which only the carbon atoms are chemically active with the ordinary air constituents. Again, this model would tend to provide an upper limit to the CN concentration, since intermediate species involved in breaking up the contaminant have not been considered. An advantage to this approach is that some idea of an acceptable contamination level for a carbon-bearing compound may be obtained from the results of the calculations. For example, if a contaminant A contains μ_C carbon atoms per molecule and μ_u atoms of additional unspecified elements, the atom fraction of carbon is

$$\phi = \frac{\mu_C x_A}{\left(\mu_u + \mu_C\right) x_A + 2\left(x_{N_2} + x_{O_2}\right)}$$

with

$$x_A + x_{N_2} + x_{O_2} = 1$$

If

$$x_A << x_{N_2} + x_{O_2}$$

the mole fraction of the compound is approximately

$$x_A \approx \frac{2\phi}{\mu_C} \tag{2}$$

Calculations have been made for atom fractions of carbon of 0.001, 0.005, 0.01, and 0.05 for p_1 values of 0.1 and 1 mm Hg. The results are shown graphically in figures 4 and 5. In these figures, the CN radiation reaches a maximum for $V_s \approx 26\ 000$ ft/sec (7.92 mm/μsec) and decreases rapidly thereafter. This trend, along with the rapid monotonic increase of the air radiation, illustrates the fact that the contamination radiation becomes negligible at the higher speeds.

For $V_s = 23\ 000$ ft/sec (7.01 mm/μsec), the CN radiation observed in the Ames expansion tube is approximately that calculated for $\phi = 0.05$, for equilibrium conditions behind the shock. (See fig. 5.) Equation (2) applied to a compound containing 10 carbon atoms per molecule gives a contaminant mole fraction of 0.01. Thus only 1-percent contamination of the test gas by such a compound could conceivably produce behind a shock CN violet radiation of an intensity several times that of air. However, radiation caused by this species at speeds greater than about 32 000 ft/sec (9.75 mm/μsec) could be completely ignored as far as radiation heat transfer is concerned. At the higher speeds, atomic and ion continuum radiation from the dissociated constituents of a contaminant is not expected to be significantly different from radiation from atoms and ions of pure air, since theoretical relationships for the various types of continua predict a weak dependence of the emission coefficient on the atomic weight of the species.

The CN violet radiation as calculated by the previously described methods is compared to a direct determination of j_{CN} in which the equilibrium mole fraction of CN was computed by using the normal-shock, free-energy minimization program of reference 9 (figs. 6 and 7). This program was applied to air with a free-stream mole fraction of C_2N_2 of 0.005. These results are compared with the first approximate solution for $\left(x_{C_2N_2}\right)_1 = 0.005$ and with the solution for a corresponding atom fraction of carbon of 0.005 at free-stream pressures of 0.1 and 1 mm Hg. Both methods predict accurately the velocity regime in which the CN radiation begins to decrease. Below 26 000 ft/sec

(7.92 mm/μsec), the first approximate solution leads to an overestimate of the number density of CN behind the shock, primarily because of neglect of the exchange reaction $CN + O \rightleftharpoons CO + N$. However, the results for $\phi = 0.005$ agree very well with the direct calculation over the entire velocity regime for which CN radiation is most significant.

Nonequilibrium Calculations

The problem of nonequilibrium radiation from CN was considered also. A computer program which calculates the finite-rate chemistry behind a normal shock was used to compute the concentration of CN as a function of distance. The pressure and temperature were assumed to increase discontinuously across the shock as determined from the conservation equations for mass, momentum, and energy. The reaction mechanism included the reactions considered most important for air, in addition to the following reactions involving the carbon compounds:

$$C_2N_2 + M \rightleftharpoons 2CN + M$$

$$CN + O \rightleftharpoons CO + N$$

$$CO + O + M \rightleftharpoons CO_2 + M$$

$$CO + M \rightleftharpoons C + O + M$$

$$CN + M \rightleftharpoons C + N + M$$

$$C + e^- \rightleftharpoons C^+ + 2e^-$$

Rates for reactions occurring in air were obtained from reference 10; the dissociation reaction used for C_2N_2 was that given in reference 11. The remaining reaction rates were taken from reference 12, with the exception of the ionization of carbon, for which a classical electron-impact formula was used to define the ionization rate.

For a first approximation to the radiation from the nonequilibrium zone, the internal energy modes were assumed to be in equilibrium with the local translational temperature. In order to obtain a more realistic model for the nonequilibrium radiation, it is necessary to account for the finite excitation time of the excited electronic levels of the radiating species. In the case of radiation from air, the experimental data of reference 13 were used to determine the distance behind the shock at which the radiation intensity was maximum. The intensity was then assumed to increase linearly with distance up to this point, and thereafter to be determined by the local translational temperature.

For the case of the CN violet, reaction rates involving the excitation of the $B\,^2\Sigma$ level of CN and the deexcitation by collisions and by spontaneous emission were employed according to the method of reference 14. The profiles for the CN violet were calculated by this method until the resulting intensity approached that obtained when the $B\,^2\Sigma$ level was populated according to the local translational temperature. The additional reactions and rates used for these calculations are

$$\mathrm{CN} + \mathrm{M} \rightleftarrows \mathrm{CN}^* + \mathrm{M} \quad \begin{cases} \left(k_f = \left(8.62 \times 10^{-13}\right) T^{5.50} e^{-37\,100/T} \dfrac{\mathrm{cm}^3}{\text{mole-sec}}\right) \\ \left(k_b = \left(8.62 \times 10^{-13}\right) T^{5.50} \dfrac{\mathrm{cm}^3}{\text{mole-sec}}\right) \end{cases}$$

$$\mathrm{CN}^* \rightarrow \mathrm{CN} + h\nu \qquad \left(k_f = 8.86 \times 10^{6}\ \mathrm{sec}^{-1}\right)$$

For each set of initial conditions, the emission coefficients of air and CN violet and the temperature and mole fraction of CN, along with the equilibrium asymptotes, are illustrated as functions of the distance behind the shock (figs. 8 and 9). The equilibrium values for T and x_{CN} were determined from the equilibrium normal-shock program (ref. 9), and the equilibrium air-radiation values are those obtained from Allen (ref. 8). The nonequilibrium air radiation was calculated from correlation formulas which give the emission coefficient per particle as a function of temperature for various transitions. These formulas for the air species are given in reference 15, and the CN violet radiation was calculated from a correlation formula determined from the data in figure 1. Needless to say, these formulas are extrapolated in certain instances to extremely high temperatures where they cannot be expected to remain very accurate. Furthermore, the oxygen and nitrogen line radiation and free-bound radiation, which are major contributors at high temperatures, have been omitted from the air-radiation calculations because acceptable correlation formulas were not immediately available. The omission of this radiation contributes to the variance between the value of the emission coefficient of air at 5 cm and the corresponding equilibrium value given by Allen for a velocity of 33 000 ft/sec (10.06 mm/μsec). (See fig. 8.)

One conclusion which may be drawn from these calculations is that the CN violet contamination radiation presents about the same problem in the chemical relaxation zone as in the equilibrium zone, at least for the lower shock speeds. However, the CN number density profile may differ considerably for various contaminants, and it is expected that the CN violet profile will not exhibit as rapid a rise in the nonequilibrium zone when a contaminant does not contain the CN radical in its molecular structure. To illustrate this, the nonequilibrium calculation has been performed for 1 percent CO_2 contamination. The resulting profile for the CN violet radiation is shown in figure 10 for $V_s = 23\,000$ ft/sec (7.01 mm/μsec) and $p_1 = 1$ mm Hg, and is compared with the calculation for 0.5 percent C_2N_2 contamination for the same speed and initial pressure. The equilibrium value shown is for $\phi = 0.005$.

The nonequilibrium normal-shock calculations may be used to estimate the effects to be expected for a blunt body in expansion-tube flow by assuming a linear velocity profile through a gas cap with shock detachment distance δ. The particle residence time in the shock layer may then be equated to the particle time after passage through a one-dimensional normal-shock wave. Use of the empirical expression for shock detachment distance for a spherical-nose radius r

$$\delta \approx 0.8 \frac{\rho_1}{\rho_2} r$$

together with the assumption of a linear velocity profile between the shock and the stagnation point, leads to the following approximate expression for particle residence time in the shock layer at a distance ξ behind the shock front:

$$t \approx -\frac{0.8r}{V_s} \ln\left(1 - \frac{\xi}{\delta}\right)$$

For a typical case of $r = 3$ cm, $V_s = 23\ 000$ ft/sec (7.01 mm/μsec), and $\xi = 0.75\delta$, the calculated particle time is 3.8 μsec. The distance behind the shock in the standing normal-shock calculation corresponding to this particle time is about 0.35 cm. Figures 10, 11, and 12 show that for these conditions, nonequilibrium effects must be taken into consideration. In order to illustrate this further, the radiation profiles have been integrated, and the ratios of contaminant radiance to air radiance are compared in table I. The ratio for the corresponding equilibrium case is also given.

TABLE I.- RATIOS OF CONTAMINANT RADIANCE TO AIR RADIANCE

Contaminant	$\dfrac{\int_0^{0.35} j_{CN}\,dy}{\int_0^{0.35} j_{air}\,dy}$
0.005 C_2N_2	0.212
.01 CO_2	.00452
ϕ = .005 (equilibrium only)	.187

The C_2N_2 contamination level of 0.005 is seen to lead to a nonequilibrium flux ratio about the same as the corresponding equilibrium ratio, whereas the CO_2 contaminant leads to a much lower flux ratio.

CONCLUDING REMARKS

Techniques for analyzing the effects of contamination radiation from the CN violet band system behind normal shock waves are presented. Equilibrium radiation from CN and air with various contamination levels is determined for shock velocities ranging from 20 000 to 44 000 ft/sec (6.10 to 13.41 mm/μsec) at free-stream pressures of 0.1 and 1 mm Hg.

For all equilibrium cases, the CN radiation is seen to be negligible at the higher velocities, even for the highest contamination levels considered, because of the depletion of the CN particle concentration by dissociation. However, at the lower speeds, relatively small amounts of contaminant may cause the air radiation to be completely dominated by the CN violet band system.

The primary conclusion drawn from the nonequilibrium calculations is that the CN violet contamination radiation can be about as severe in the chemical relaxation zone as for the corresponding equilibrium case. Of course, C_2N_2 is an unrealistic shock-tube contaminant and was used in the calculations only as a convenient method of obtaining CN behind the shock. More realistic contaminants might include various forms of hydrocarbon oils and perhaps graphitic carbon. For these types of contaminants, the production of CN behind the shock would not take place nearly as rapidly as for the calculated cases and, in many instances, the CN number density would be monotonically increasing to its equilibrium value.

Langley Research Center,
National Aeronautics and Space Administration,
Langley Station, Hampton, Va., November 17, 1966,
129-01-08-35-23.

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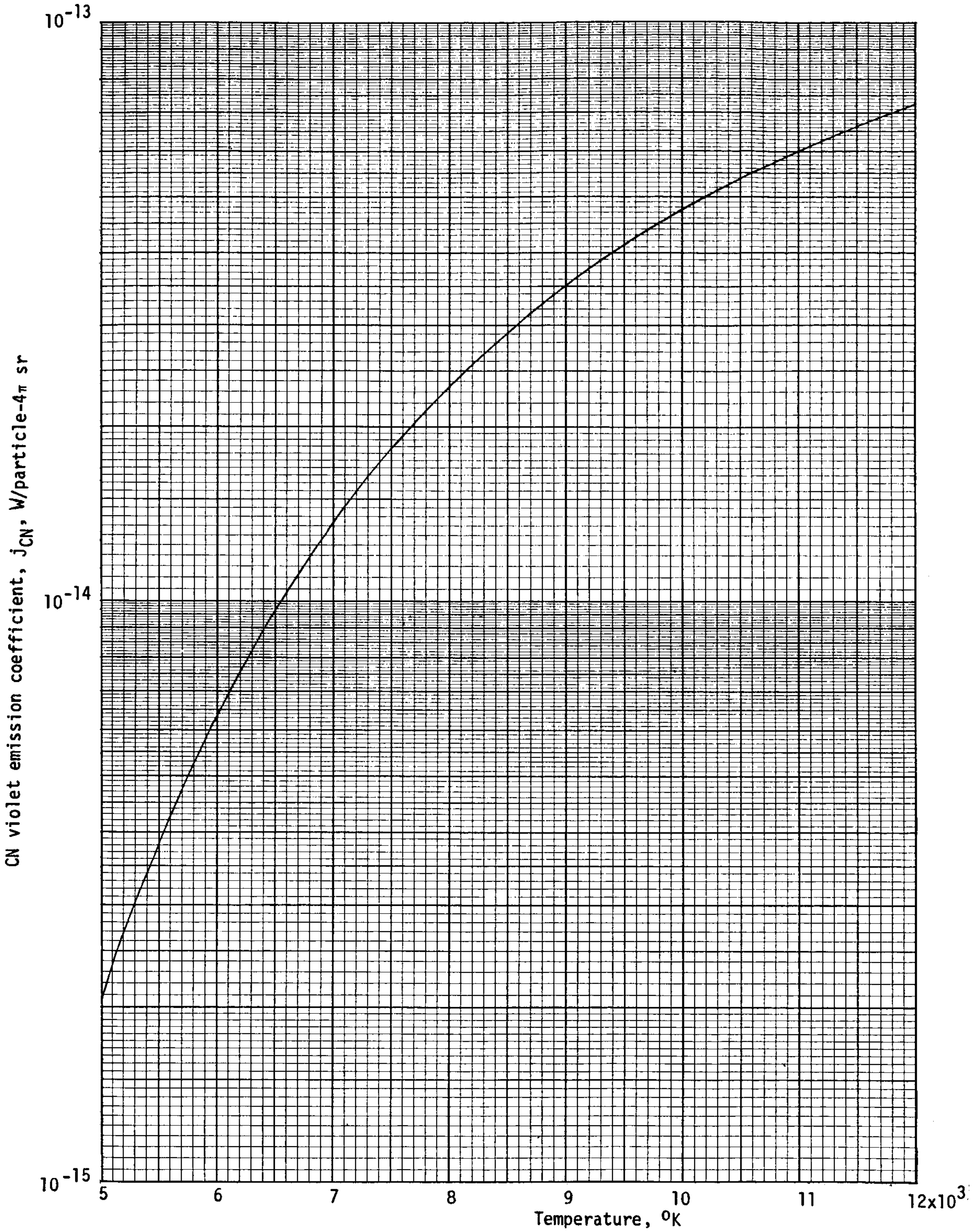

Figure 1.- Average CN violet emission coefficient per particle for an f-number of 0.020.

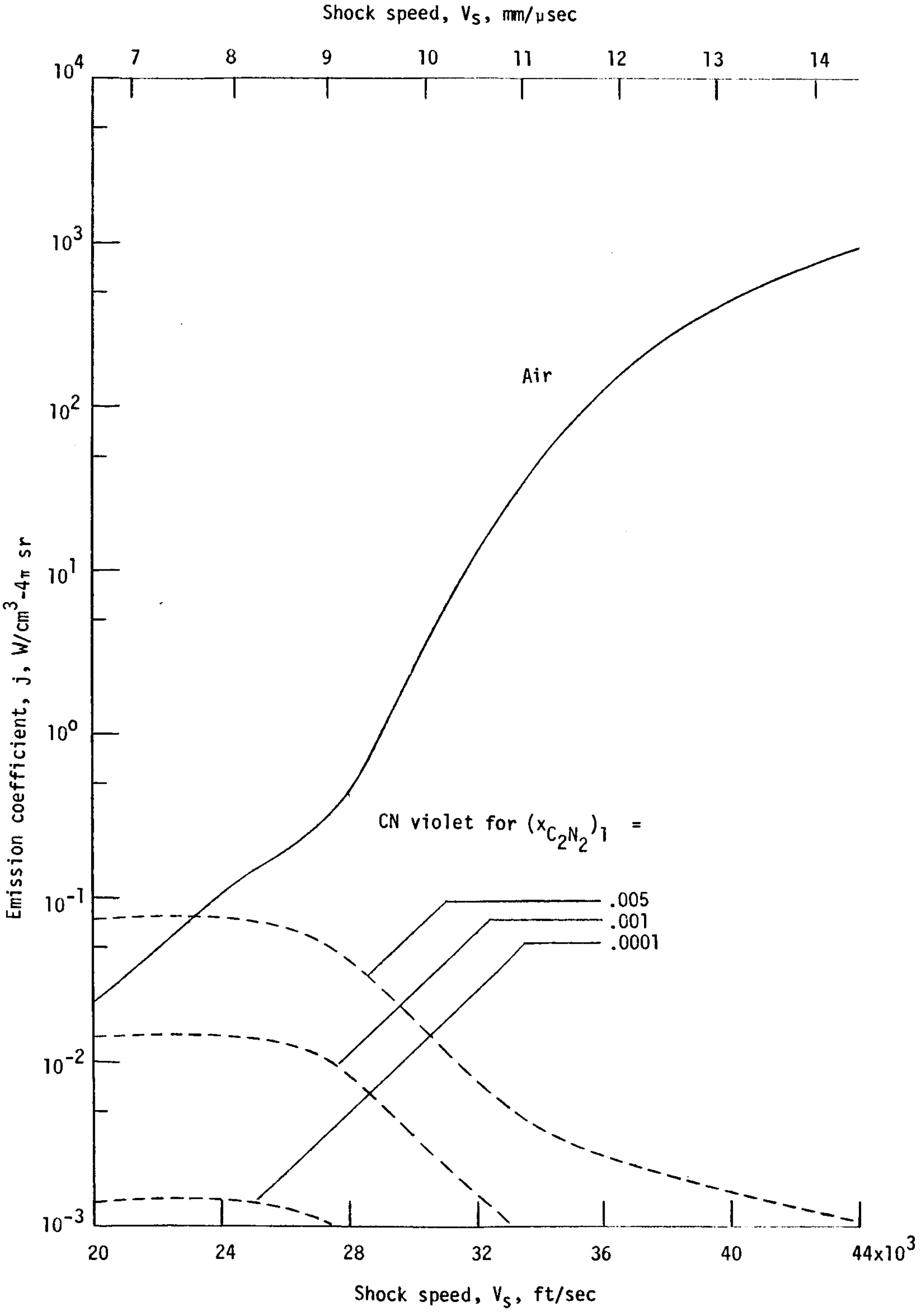

Figure 2.- Equilibrium emission coefficients for air and CN violet for p_1 = 0.1 mm Hg with C_2N_2 contaminant – approximate solution.

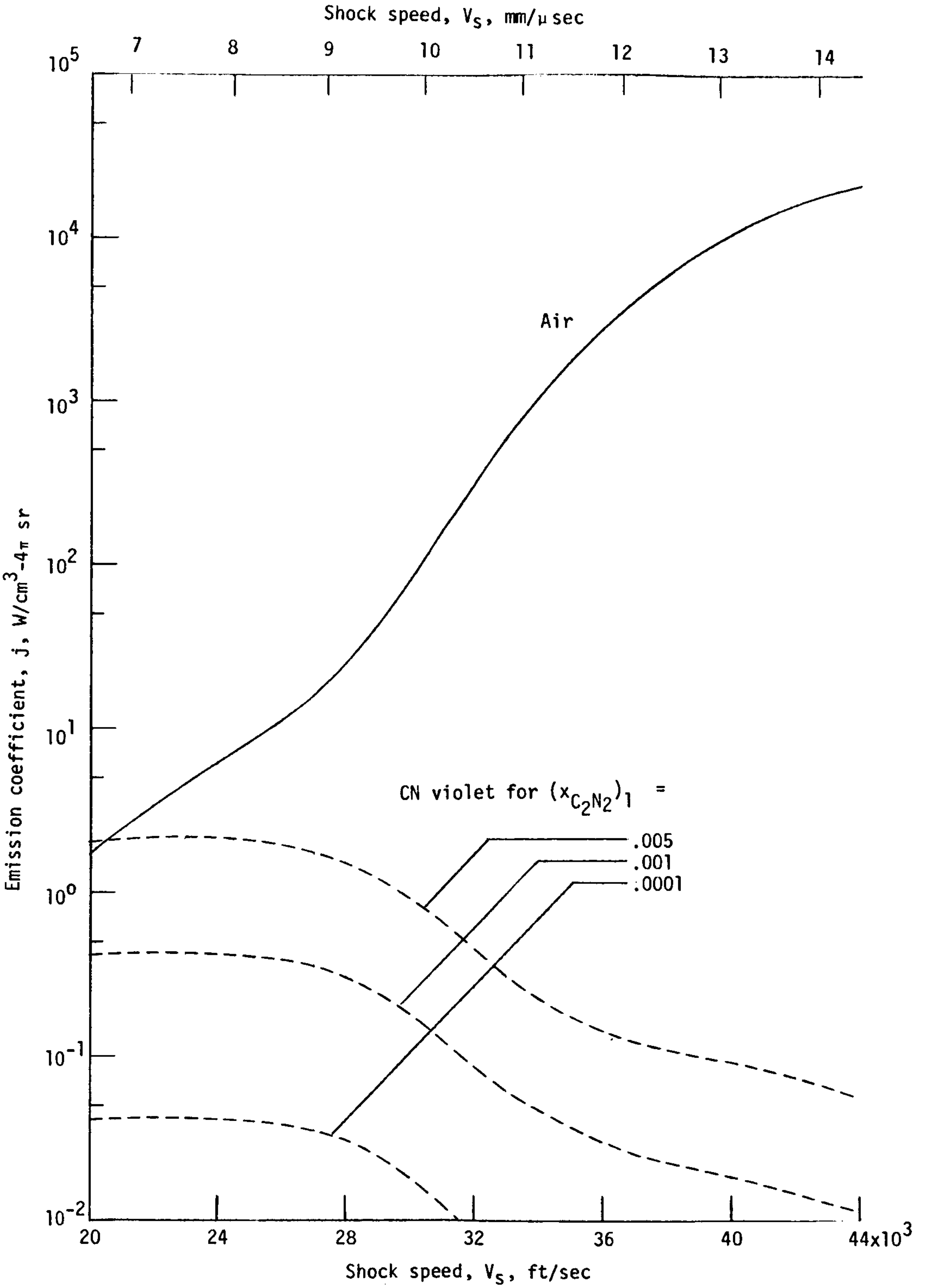

Figure 3.- Equilibrium emission coefficients for air and CN violet for $p_1 = 1$ mm Hg with C_2N_2 contaminant - approximate solution.

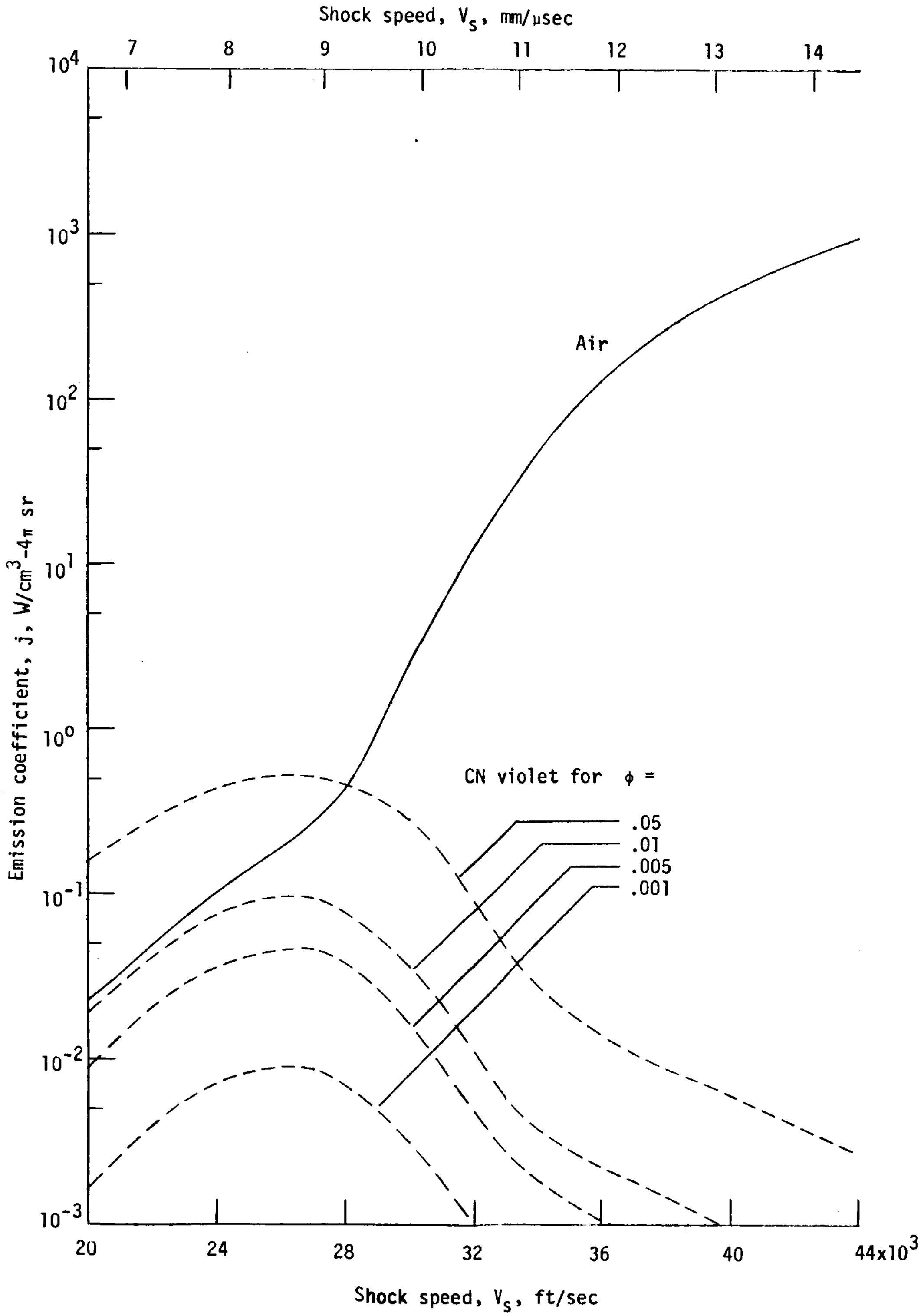

Figure 4.- Equilibrium emission coefficients for air and CN violet for $p_1 = 0.1$ mm Hg with specified carbon atom fraction.

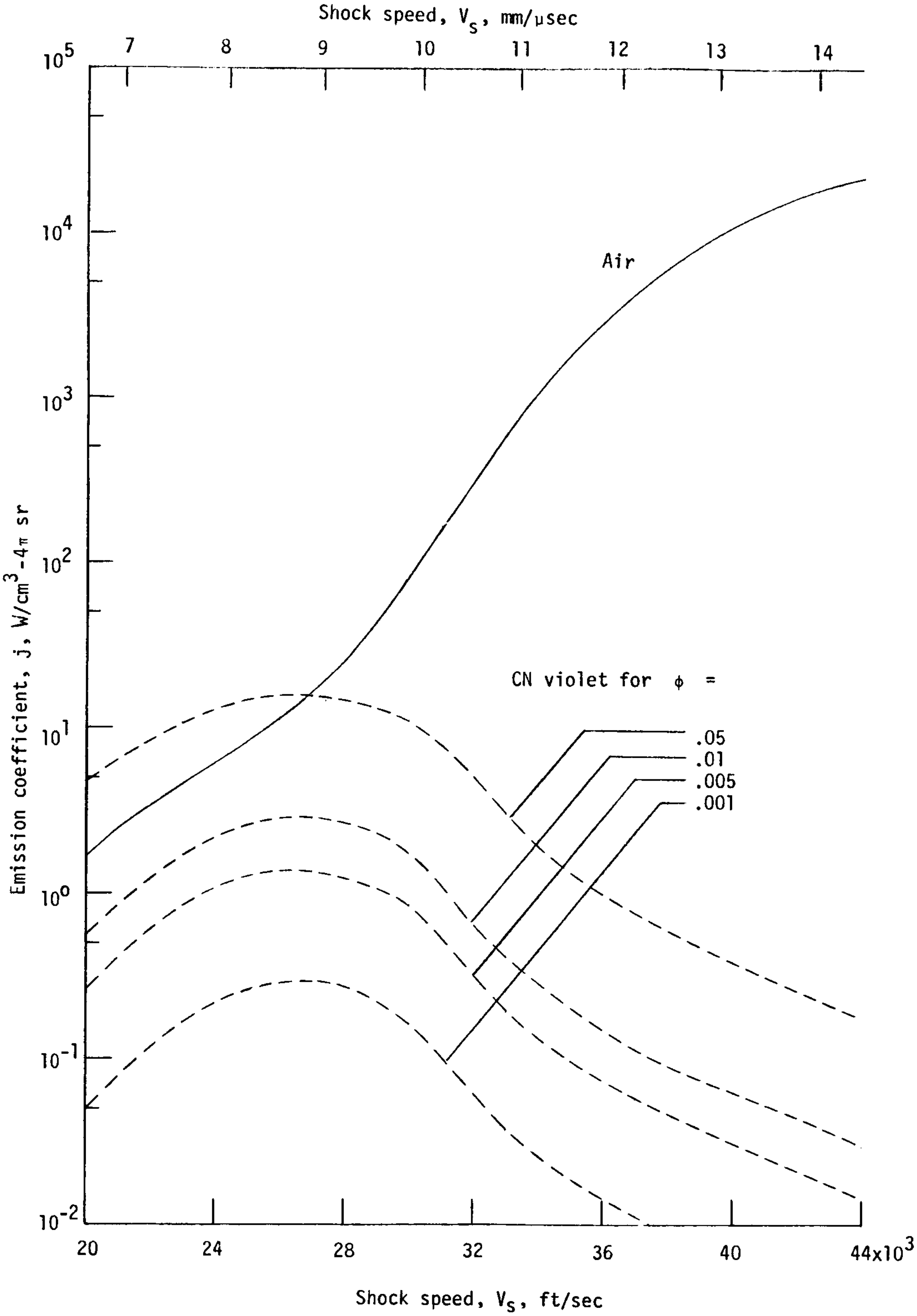

Figure 5.- Equilibrium emission coefficients for air and CN violet for $p_1 = 1$ mm Hg with specified carbon atom fraction.

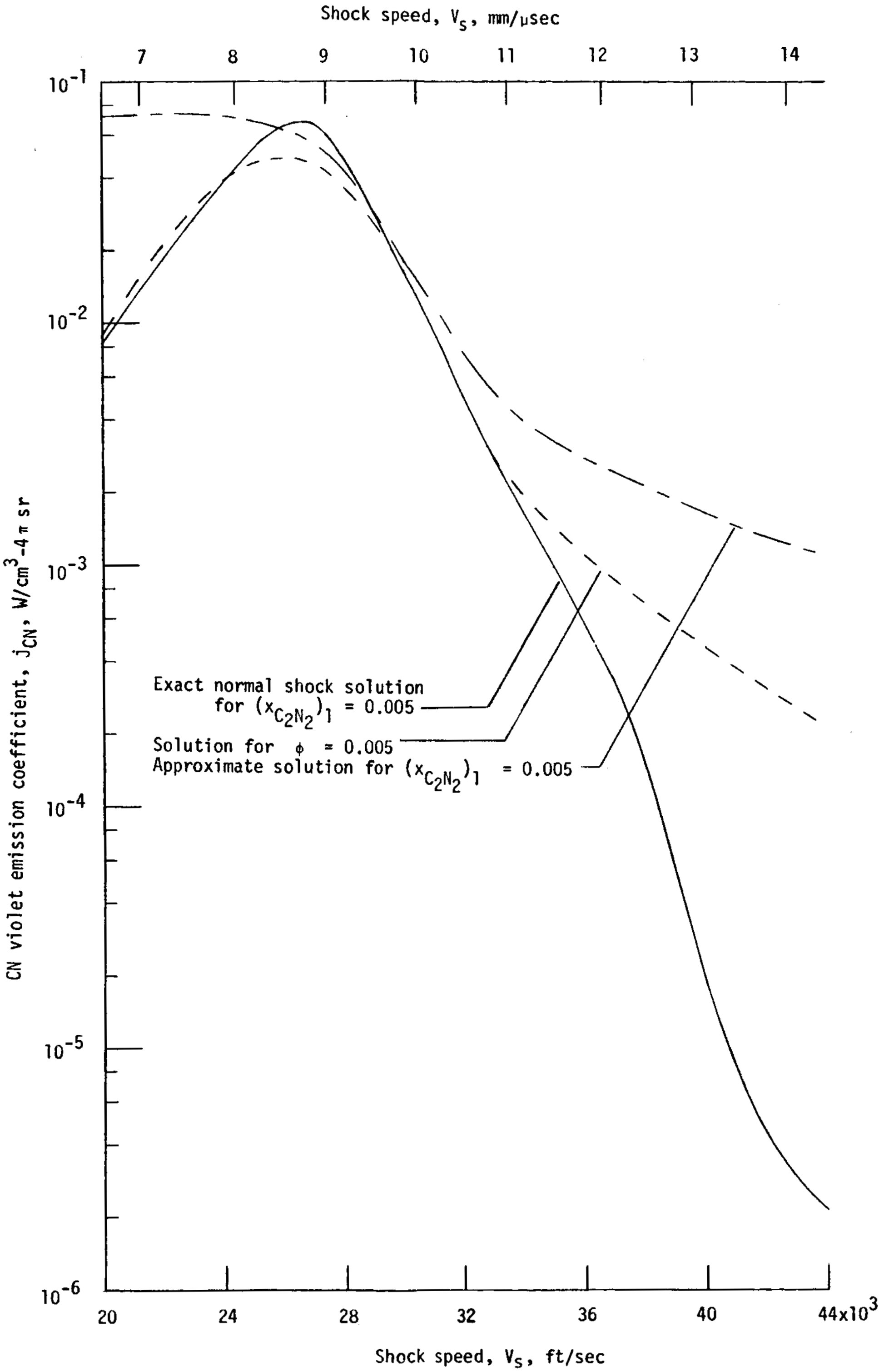

Figure 6.- Comparison of solutions for equilibrium CN violet emission coefficient for $p_1 = 0.1$ mm Hg.

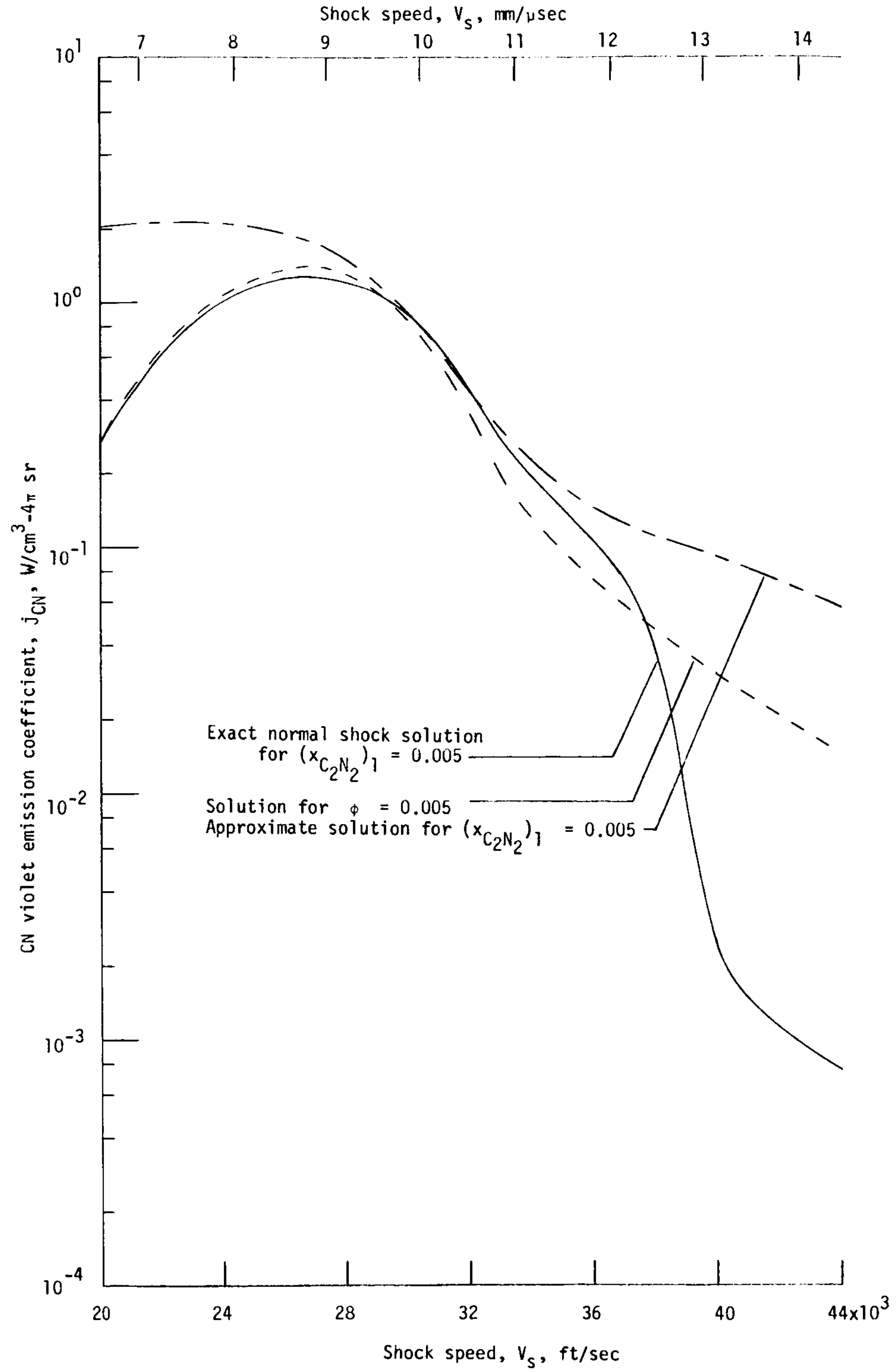

Figure 7.- Comparison of solutions for equilibrium CN violet emission coefficient for $p_1 = 1$ mm Hg.

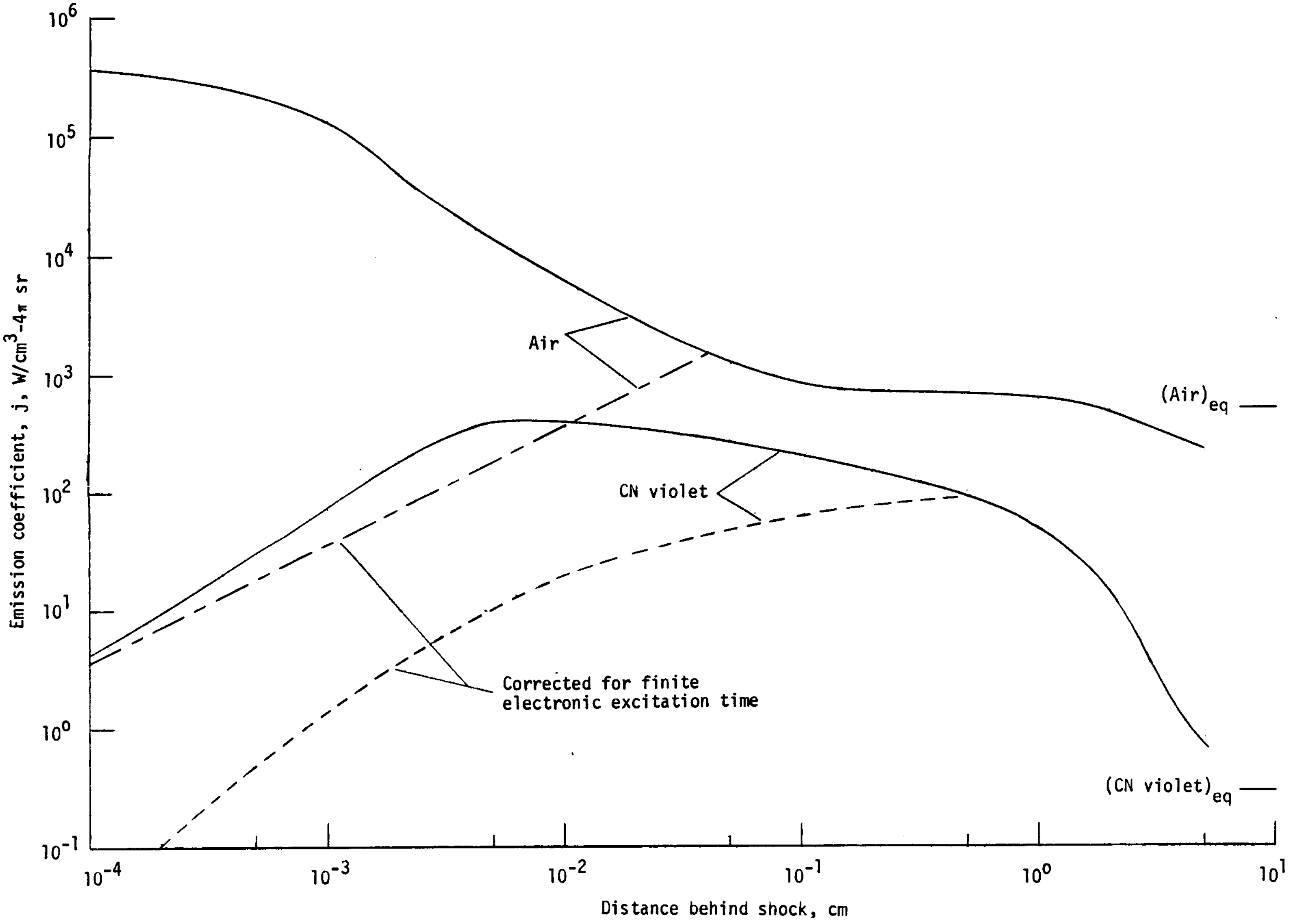

Figure 8.- Nonequilibrium profiles of emission coefficients for air and CN violet for initial contamination level of 0.5 percent C_2N_2. V_s = 33 000 ft/sec (10.06 mm/μsec); p_1 = 1 mm Hg.

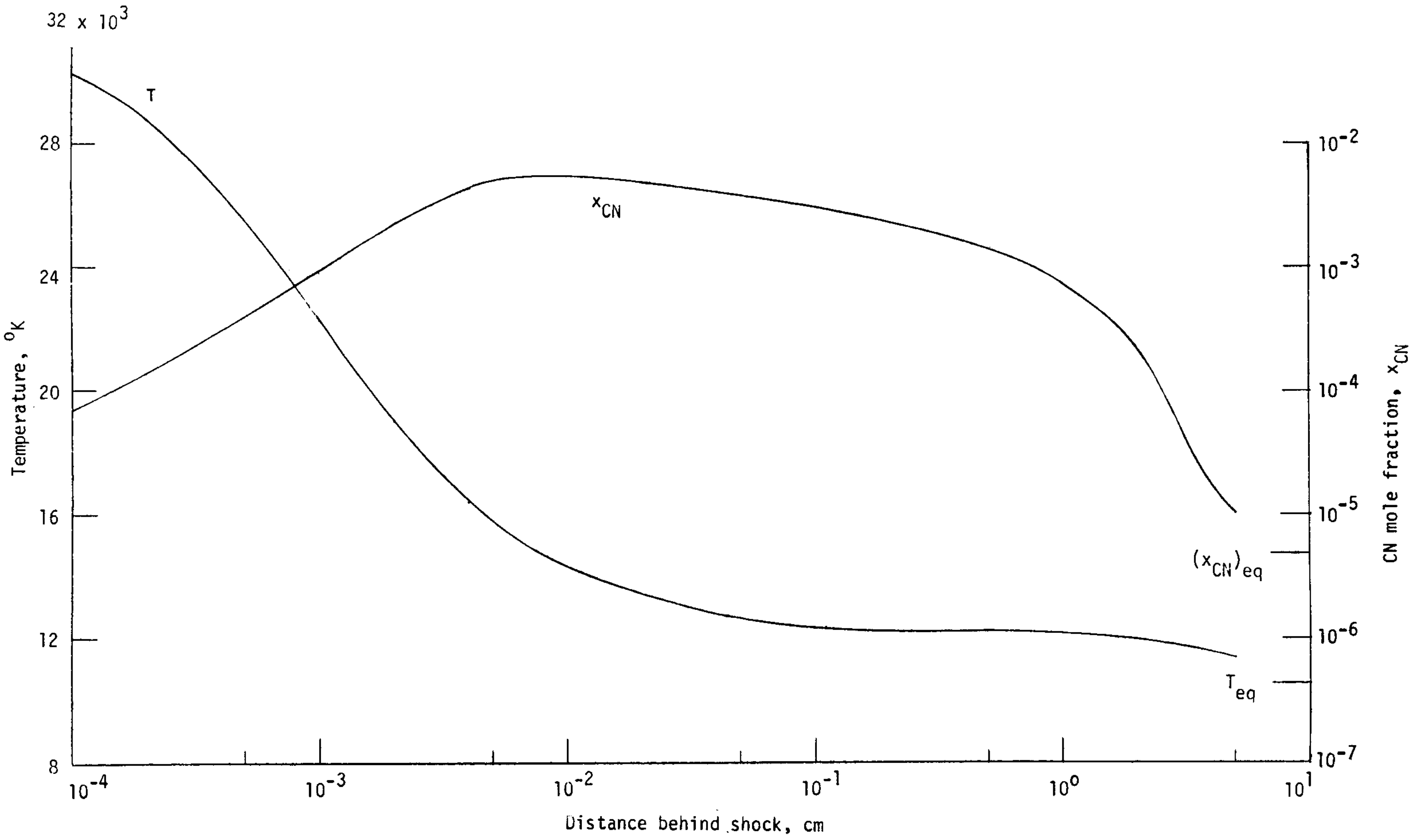

Figure 9.- Nonequilibrium profiles of temperature and mole fraction of CN for initial contamination level of 0.5 percent C_2N_2. V_s = 33 000 ft/sec (10.06 mm/μsec); p_1 = 1 mm Hg.

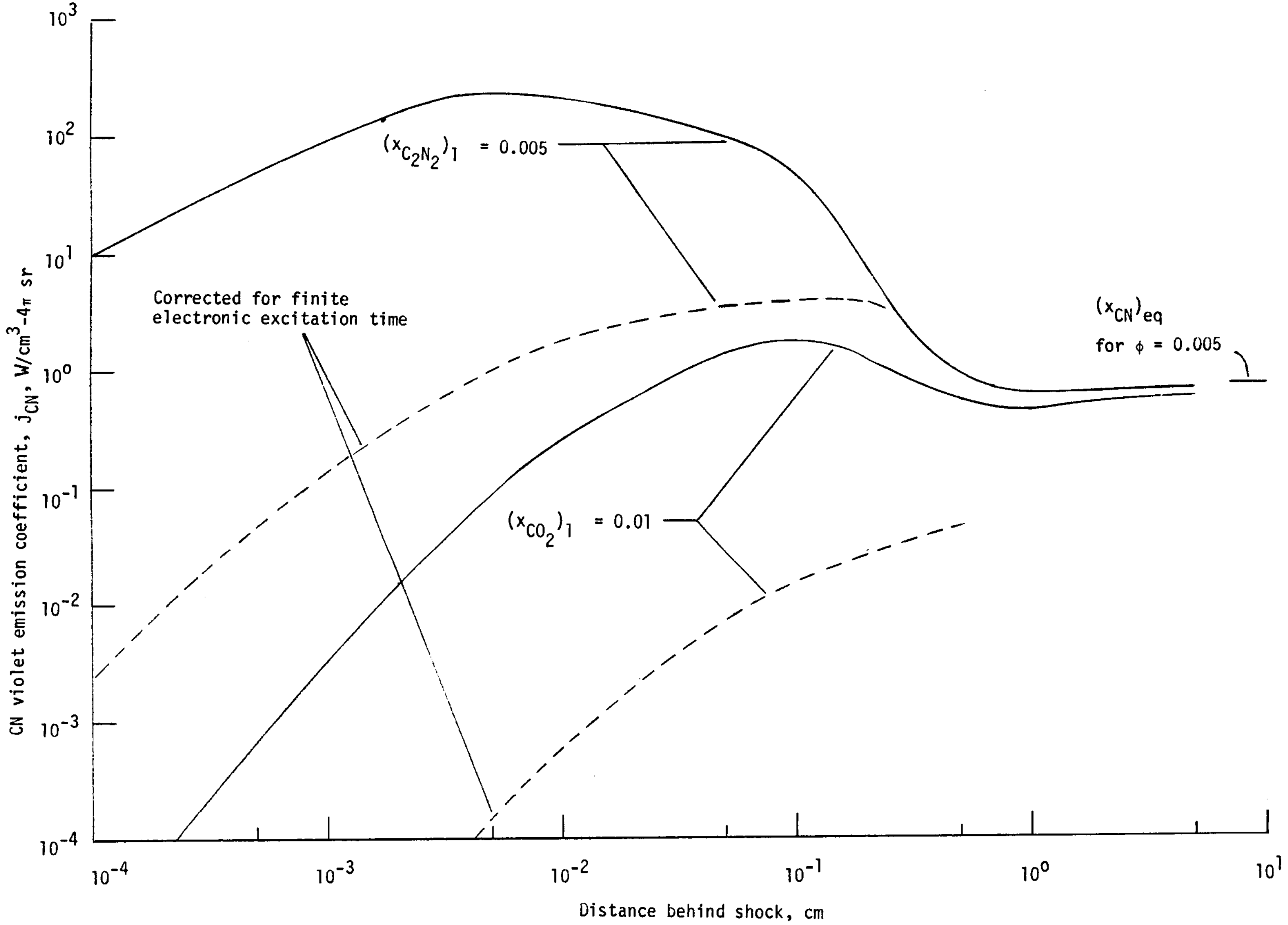

Figure 10.- Comparison of nonequilibrium emission coefficients of CN violet for initial contamination levels of 0.5 percent C_2N_2 and 1.0 percent CO_2. V_s = 23 000 ft/sec (7.01 mm/μsec); p_1 = 1 mm Hg.

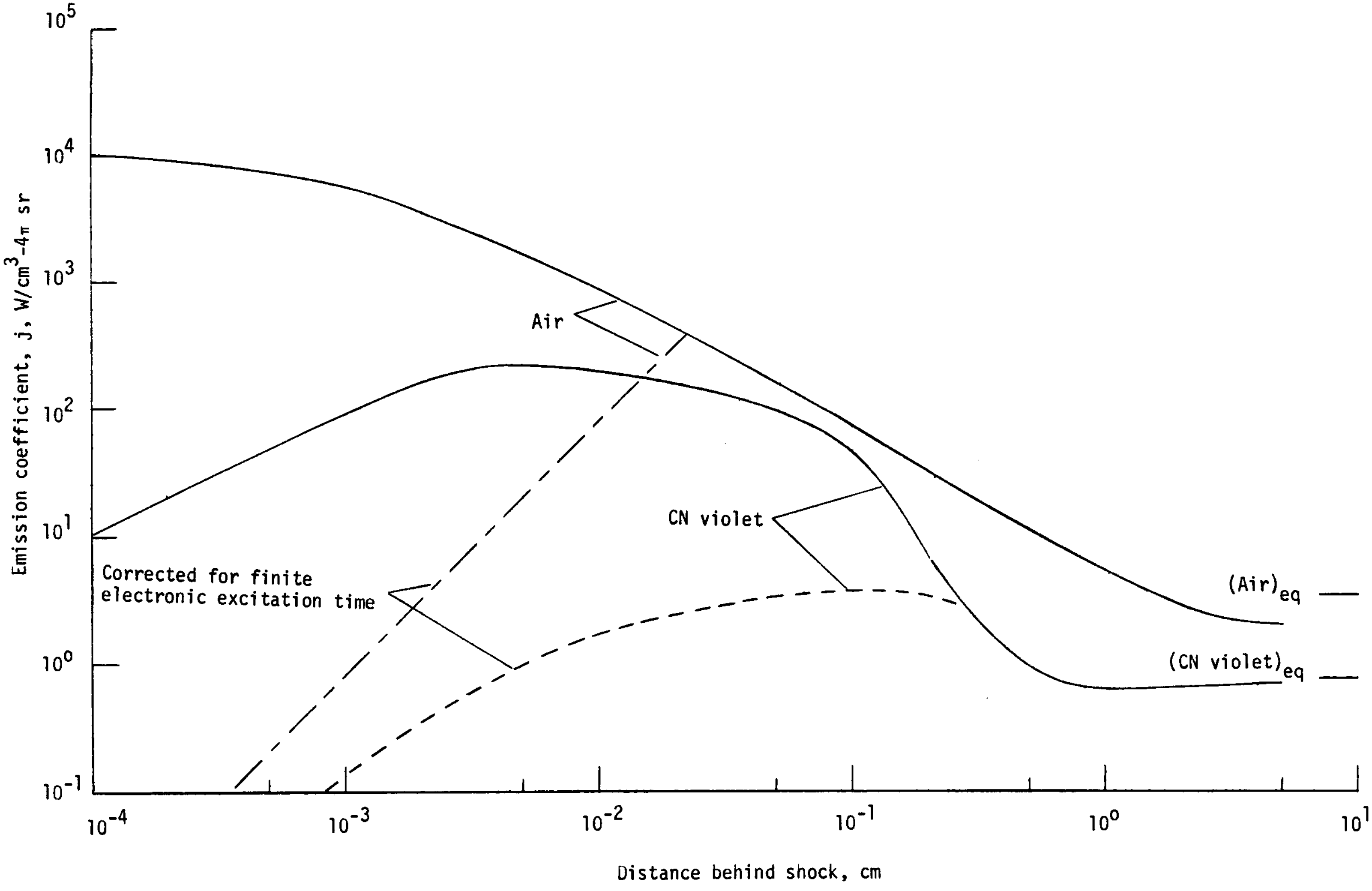

Figure 11.- Nonequilibrium profiles of emission coefficients for air and CN violet for initial contamination level of 0.5 percent C_2N_2. V_s = 23 000 ft/sec (7.01 mm/μsec); p_1 = 1 mm Hg.

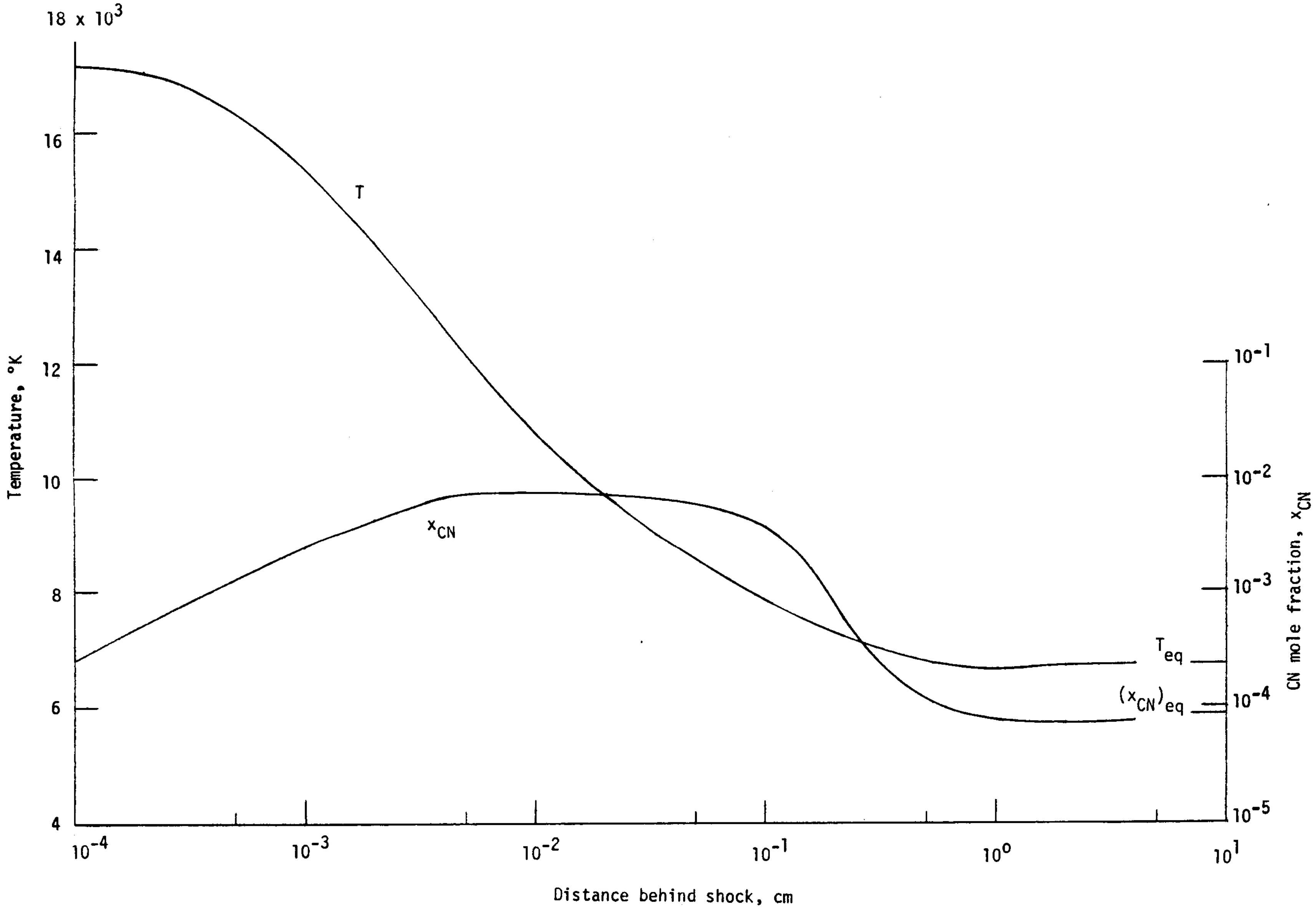

Figure 12.- Nonequilibrium profiles of temperature and mole fraction of CN for initial contamination level of 0.5 percent C_2N_2. V_s = 23 000 ft/sec (7.01 mm/μsec); p_1 = 1 mm Hg.

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