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13. ABSTRACT (Maximum 200 words) A phenomenon observed during plume diagnostic measurements acquired during simulated altitude chamber tests of aluminum-loaded solid-propellant rocket motors is discussed, and analytical results are presented. The results of a line-by-line calculation of the aluminum chloride (AlCl) feature are discussed and compared with high-resolution ultraviolet (UV) spectral data.					
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THE AICI ABSORPTION FEATURE IN SOLID ROCKET PLUME RADIATION*

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Abstract

An absorption feature in the mid-ultraviolet spectral region of the radiation from aluminized solid-propellant rocket plumes has been identified as the $X^1\Sigma \rightarrow A^1\Pi$ transition of aluminum chloride (AlCl). A proposed model of the radiative transfer has the continuum radiation from the aluminum oxide particulate core passing through the outer gas boundary. A line-by-line calculation is then performed in a temperature-density-path length parameter space to attempt a fit to the data. The final temperature, density, and path length agreed well with flow-field calculations. The spectral agreement between model and data was marginal, but sufficient spectral structure and band shape fits were found to merit further work on this mechanism as a diagnostic tool in solid-propellant rocket studies.

Introduction

An interesting feature in the mid-ultraviolet spectrum of the radiation from aluminized solid-propellant rocket exhausts has been noted several times in unpublished reports. This feature is a deep absorption near 262 nm of the otherwise continuum spectrum typical of hot aluminum oxide particles. It has been attributed to the aluminum chloride (AlCl) A-X transition, although the spectral resolution was not sufficient to resolve bandheads.¹ Recently, spectra were obtained having sufficient resolution to identify the vibrational bands and these will be reported here. Curiously, spectra obtained from an on-board spectrometer viewing a rocket plume having an aluminized propellant also contained this feature.²

One obvious question arising from such an inherent spectral feature is "Can it be used to diagnose any properties of the rocket exhaust flow?" To

that end, a modeling effort was undertaken and this paper reports on the success of that work.

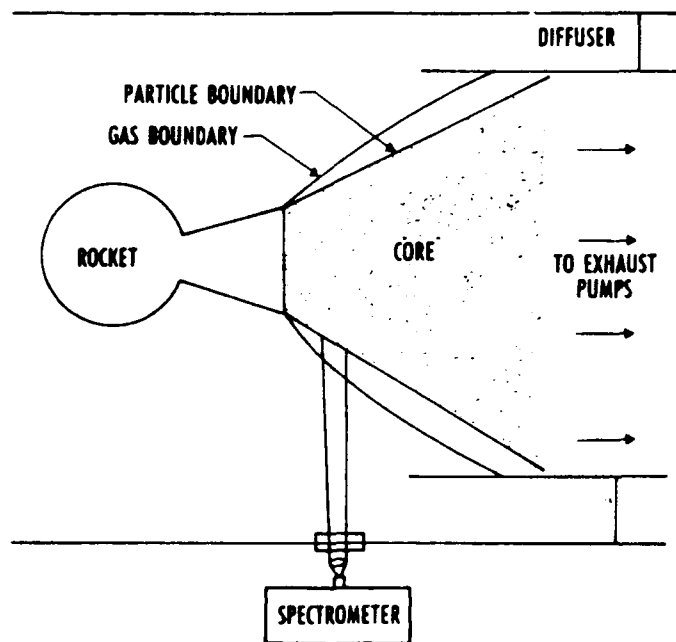


Fig. 1. Schematic of solid rocket motor test configuration used for UV spectral measurements.

Description of the Spectra

When a moderate resolution (~ 0.8 nm) ultraviolet scanning monochromator is pointed normal to the axis of an exhaust plume from an aluminized solid-propellant rocket motor being fired in an altitude test cell (Fig. 1), the spectra shown in Fig. 2 results. The continuum on either side of the feature at 262 nm and the thermal behavior of the radiation in the visible and infrared regions suggests that the feature is due to absorption of a gaseous specie. A quick examination of the predicted products of combustion of ammonium perchlorate with aluminum and hydrocarbon solids along with a trip through Pearce and Gaydon's³ atlas of spectra reveals that the most

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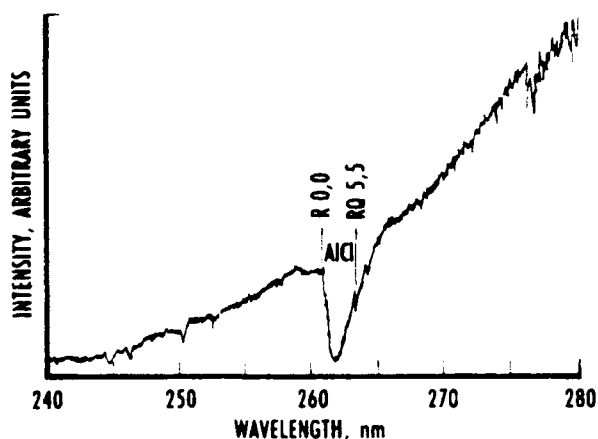


Fig. 2. Moderate resolution mid-UV spectra from solid-propellant rocket motor plume.

likely absorber is aluminum chloride (AlCl). To investigate this identification more thoroughly, higher-resolution spectra were obtained during a subsequent test. Because of the small vibrational ($\sim 450 \text{ cm}^{-1}$) and rotational ($\sim 0.25 \text{ cm}^{-1}$) constants of both AlCl states, a spectral resolution of about 0.1 nm was required to produce the band structure. The resulting spectrum (Fig. 3), indeed does show the expected structure, and many of the bands listed in Pearce and Gaydon, Table 1, are identified.

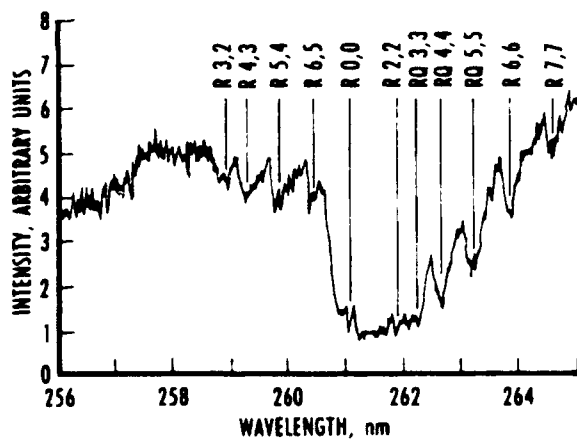


Fig. 3. High resolution UV spectra from solid-propellant rocket motor plume.

The AlCl Molecule

Aluminum chloride sublimates at temperatures above 460 K. A calculation of the plume properties using the JANNAF Standard Plume flow-field codes⁴ gives the solid and gaseous temperature profiles of Fig. 4 for this particular rocket plume. The minimum predicted temperature is about 600 K so that the AlCl is gaseous. Furthermore, the predicted mole fraction of AlCl is about 2.6 percent, and should follow the static pressure radial profile (Fig. 4).

Table 1. Bandheads of the AlCl $A^1\Pi - X^1\Sigma$ System

λ	I	V,V	λ	I	V,V
2708.9 R	2	R 7,9	2625.0 R	5	Q 3,3
2702.3 R	3	R 6,8	2622.4 R	4	R 3,3
2696.4 R	3	R 5,7	2620.0 R	4	Q 2,2
2692.8 R	5	Q 4,6	2618.2 R	3	R 2,2
2685.7 V	6	Q 2,4	2617.0 R	4	Q 1,1
2683.1 V	6	Q 1,3	2614.4 R	8	Q 0,0
2681.1 V	4	Q 0,2	2610.2 R	6	R 0,0
2649.7 V	4	Q 1,2	2606.7 R	2	R 6,5
2647.5 V	6	Q 0,1	2600.7 R	3	R 5,4
2644.9 R	2	R 7,7	2695.4 R	2	R 4,3
2638.1 R	3	R 6,6	2590.8 R	2	R 3,2
2632.8 R	3	Q 5,5	2586.7 R	2	R 2,1
2632.2 R	3	R 5,5	2564.3 R	1	R 4,2
2627.8 R	4	Q 4,4	2559.6 R	1	R 3,1
2627.0 R	3	R 4,4	2555.5 R	1	R 2,0

NOTE: THE LETTERS R AND V AFTER THE WAVELENGTH INDICATE THE DIRECTION OF DEGRADATION OF THE BAND, WHEREAS THE NATURE OF THE HEAD (R AND Q) IS INDICATED BEFORE THE VIBRATIONAL QUANTUM NUMBERS.

REF.: THE IDENTIFICATION OF MOLECULAR SPECTRA, R. W. B. PEARCE AND A. G. GAYDON, CHAPMAN AND HALL LTD., 1950, p. 47.

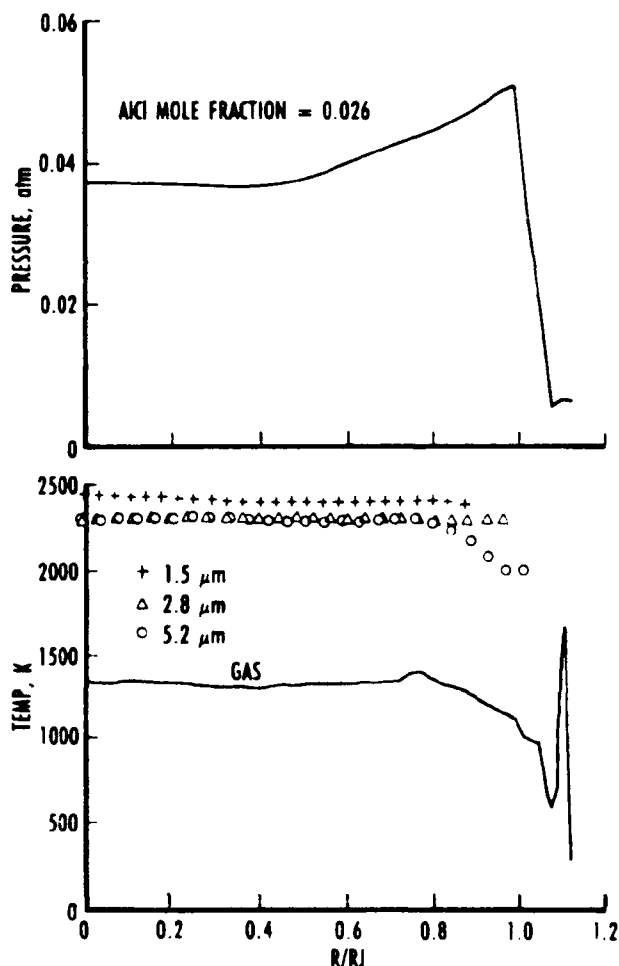


Fig. 4. Predicted radial profiles of temperature and pressure for solid-propellant rocket motor at axial location where UV spectra were measured (standard plume flow-field code using three particle groups).

The molecular constants of the two electronic states ($A^1\Pi$ and $X^1\Sigma$), measured by several investigators and reported critically by Huber and Herzberg⁵, are given in Table 2. The values are for $Al^{35}Cl$, although $Al^{37}Cl$ also naturally occurs; these two isotopes occur in the ratio 75.5:24.5. The relatively small vibrational constant of the ground state (480 cm^{-1}) leads to a vibrational-rotational fundamental at a wavelength of about $21\text{ }\mu\text{m}$. (Observation of this LWIR emission band from rocket plumes has not been reported.) The constants of the $X^1\Sigma$ state have been re-measured recently in the infrared bands, and improvements in accuracy have been reported. These data will be introduced subsequently. The rotational constants for this closely packed molecule are also quite small ($\sim 0.254\text{ cm}^{-1}$), and the overlapping branches make resolution of the lines near impossible.

Table 2. Molecular Constants for the Ground and Resonant States of $Al^{35}Cl$ (cm^{-1})

	$X^1\Sigma^+$	$A^1\Pi$
T_e	0	38254.0
ω_e	481.30	449.96
$\omega_e x_e$	1.95	4.37
$\omega_e y_e$	...	-0.216
B_e	0.24393	0.259
α_e	0.001611†	0.006
D_e	2.503×10^{-7}	...
β_e	-0.0053×10^{-7}	...

* BASED UPON Q-BRANCH BANDHEADS

† $4.697 \times 10^{-6}(\nu + 1/2)^2 - 4.7 \times 10^{-9}(\nu + 1/2)^3$

The vibrational correction to the rotational constant given as footnotes in Ref. 5 is a necessity for proper modeling of this band. Also, because of the small values of B_e , rotational quantum numbers out to at least 100 must be used at the temperature of interest.

Radiative Transfer Model

The preliminary model adopted for this study is illustrated in Fig. 5. It is supposed that the core of solid particulates provides a continuum source for transmission through a layer of gas containing the absorbing $AlCl$. The continuum radiation from the particulates was curve fit by joining the spectra at both extremes of the absorption feature, and served as a source for the absorption calculations. The gas layer was considered uniform for these preliminary calculations, with temperature and number density as parameters. The absorption spectra were modeled as Voigt lines with the spectrometer triangular slit function considered to be much greater than a line width.

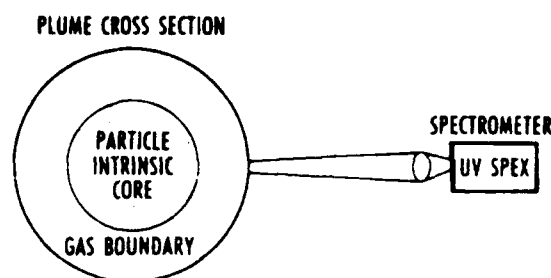


Fig. 5. Physical model of solid-propellant rocket motor plume UV spectral measurement experiment.

A complete solution to the monochromatic radiative transfer equation is desired. The equation is written

$$\frac{dI_\nu}{dx} = S_E N_{TOT} L_\nu - S_A N_{TOT} L_\nu I_\nu \quad (1)$$

where

N_{TOT} is the total number density of emitter/absorber.

L_ν is the line shape function

S_E is the molecular emission strength

S_A is the molecular absorption strength

I_ν is the spectral radiance. (i.e., $\text{W}/\text{sr}\cdot\text{cm}^2\cdot\text{cm}^{-1}$) at wavenumber ν

x is the spatial coordinate within the media

The boundary condition at $x = 0$ is given in this case by the fit to the continuum spectra, neglecting the $AlCl$ absorption feature; i.e., the continuum background.

Conceptually, the calculation involves 3 steps:

1. Determine the positions of all spectral lines. Determine the $AlCl$ emission and absorption strengths for each line, based on a rotational and vibrational temperature. This yields a spectral line atlas of line strengths versus line center wavenumber for all lines.

2. Given S_A and S_E , and N_{TOT} , solve Eq. (1). Here, only one spatial zone is considered so that integration of Eq. (1) is simplified.

3. Convolve the result of 2 for each line with spectrometer response function. In this case a triangular slit function suffices since the actual line width is much smaller than the instrument monochromatic response function.

Part 1

The strength of each emission line is given by:

$$S_E = \frac{hc\nu}{4\pi} A_{\nu\nu''} \frac{S_{J'J''}}{2J'+1} \chi(A; \nu'; J') \quad (2)$$

where

$A_{\nu\nu''}$ are the Einstein coefficients.⁷
 $S_{J'J''}$ are Honi-Londons⁸
 ν wavenumber
 $\chi(A; \nu'; J')$ is the fractional population of the $A^1\Pi$ state for ν' and J' level, normalized by the partition function, i.e.

$$\chi(A; \nu'; J') = \frac{g_{A\nu J'} \exp\left[-\frac{hc}{kT} E(A; \nu', J')\right]}{\sum_i g_i \exp\left[-\frac{hc}{kT} E_i\right]} \quad (3)$$

where g_i is the degeneracy factor of the given state.

The absorption strength is similarly given by

$$S_A = h\nu B_{\nu\nu''} \frac{S_{J'J''}}{2J''+1} \left\{ \chi(X; \nu'', J'') - \chi(A; \nu', J') \right\}$$

with the Einstein B coefficient as

$$B_{\nu\nu''} = \frac{1}{8\pi hc\nu^3} A_{\nu\nu''} \quad (5)$$

The band strength (or lifetime) of the AlCl $A^1\Pi \rightarrow X^1\Sigma$ (0,0) transition has been measured⁶ to be 6.4×10^{-4} sec, where no effort was made to separate the two isotopic states. The relative band strengths were calculated by Langhoff and Bauschlicher⁷ from first principles. They are given as Einstein coefficients in Table 3. Note the relatively strong values for the $\Delta\nu = 0$ sequence, indicating that these will be the most evident in the structure, as also reported in the experimental analyses.³ These calculations of band transition probabilities were made for Al³⁵Cl with the assumption that the results are the same for Al³⁷Cl.

Energies of individual states are given by

$$E_i = T_i + E_{V_i} + E_{R_i} \quad (6)$$

where T_i is electronic state energy, E_{V_i} is the vibrational state energy, and E_{R_i} is the rotational

Table 3. Einstein Coefficients for the $A^1\Pi - X^1\Sigma^+$ transition in AlCl (Ref. 7)

$\nu \backslash \nu''$	0	1	2	3	4	5	6	7	8
0	1933	...	...	...	...	...	...	...	...
1	...	1890	...	...	...	...	...	...	...
2	...	7.92	18.32	...	...	...	...	...	...
3	...	...	3.3	1745	12	16.6	...	...	...
4	...	...	...	92	1578	37	31	...	...
5	...	...	...	...	200	1344	77	54	...
6	...	...	...	...	...	366	1028	121	87
7	...	...	...	...	...	...	574	658	149
8	...	...	...	...	...	...	18	766	301

ALL VALUES SHOULD BE MULTIPLIED BY 10^5 sec^{-1} (REF. 6)
 VALUES NOT GIVEN ARE LESS THAN 0.5 PERCENT OF VALUE AT (0,0)

energy of quantum. Now, in terms of the vibrational quantum number ν , the rotational quantum number J , and the electronic angular momentum quantum number Λ ,

$$E_{V_i} = \omega_e \left(\nu + \frac{1}{2}\right) - \omega_e x_e \left(\nu + \frac{1}{2}\right)^2 + \omega_e y_e \left(\nu + \frac{1}{2}\right)^3 \quad (7)$$

$$E_{R_i} = B_v [J(J+1) - \Lambda^2] - D_v [(J(J+1) - \Lambda^2)^2] \quad (8)$$

with

$$B_v = B_e \alpha_e \left(\nu + \frac{1}{2}\right) + \alpha_e' \left(\nu + \frac{1}{2}\right)^2 + \alpha_e'' \left(\nu + \frac{1}{2}\right)^3$$

$$D_v = D_e + \beta_e \left(\nu + \frac{1}{2}\right)$$

For mass Al³⁷Cl all constants are modified by the ratio $\rho = \sqrt{35/37}$

$$B_e^{(37)} = B_e^{(35)} \rho^2$$

$$\alpha_e^{(37)} = \alpha_e^{(35)} \rho^3 \text{ etc.}$$

The molecular constants for the $A^1\Pi$ state were taken from Huber and Hertzberg⁵ and are listed in Table 2. For the ground electronic state the latest reported measurements⁸ use the Dunham expression⁹ for the sum of the vibrational and rotational energy,

$$T_{vJ} = \sum_{l,m} Y_{lm} \left(\nu + \frac{1}{2}\right)^l [J(J+1)]^m \quad (9)$$

The values of Y_{lm} were given to the author by Bernath⁸ and appear in Table 4.

Table 4. Constants for Dunham Level Expression of Energy Levels of the $\chi^1\Sigma$ state of AlCl

Molecular Constants for AlCl($\chi^1\Sigma$) used in Term Energy Expression: $T_{v,J} = \sum_{l,m} Y_{lm}(v + 1/2)^l J(J+1)^m$			
		Y_{lm}	
l	m	Al ³⁵ Cl	Al ³⁷ Cl
0	0	0.00000000000E+00	0.00000000000E+00
0	1	2.439300631168E-01	2.381908103047E-01
0	2	-2.501623484131E-07	-2.384688355076E-07
0	3	-4.247846717816E-14	-5.631834454137E-14
0	4	9.082196640775E-19	8.257101917000E-19
1	0	4.817765661053E+02	4.760760032685E+02
1	1	-1.611082122620E-03	-1.554551027648E-03
1	2	4.398230380075E-10	4.161398270080E-10
1	3	0.000000000000E+00	0.000000000000E+00
2	0	-2.101817969356E+00	-2.052568958832E+00
2	1	4.692056013898E-06	4.472534718003E-06
2	2	7.160332258952E-12	8.149446348886E-12
3	0	6.638164756386E-03	6.420867608319E-03
3	1	-5.291195847775E-05	-4.985420578000E-05
4	0	-2.023833735279E-05	-1.929715135000E-05

VALUES ARE IN UNITS OF CM⁻¹ (REF. 8)

With the transition energies defined, line positions are obtained by subtraction

$$\nu = E(A, v', J') - E(X, v'', J'')$$

A line atlas is built up from these expression for v' , v'' out to 8 and J' , J'' out to 200 for both isotopes.

Part 2

The line-by-line model then solves Eq. (1) for each line in the atlas. A Voigt profile for each line is assumed. The S_E and S_A are calculated at a minimum of ten spectral frequencies between spectral line centers by summing contributions of all relevant lines at that position. Here v' , v'' out to 8 and J' , J'' out to 200 are used to generate a large number of stored numbers. Thus, for 20,000 spectral lines, at least 200,000 radiances and transmittances are generated. The program also does spatial integration, although for this use (1 zone), this is trivial.

Part 3

The spectrometer is modeled by a triangle function of 2Å full width at half maximum. The convolution of the integral

$$H(\nu) = \int I(\nu - \nu') G(\nu') d\nu'$$

is done numerically using the trapezoid rule, where $H(\nu)$ represents the instrument reading at ν and $G(\nu')$ represents the instrument function,

Results of Modeling

The approach taken was to vary the vibrational and rotational temperatures, the column density, and the column length in Eq. (2) until the closest fit to the experimental data was achieved. The temperature and pressure were varied about the predicted averages along the column, (see Fig. 4) and the predicted mole fraction of AlCl was used initially. It was first necessary to calculate the constants accurately for each vibrational state of each of the isotopic molecules. This proved to be very sensitive in obtaining a fit to the vibrational bandheads. Then, the number of J values necessary to fit the absorption level was determined.

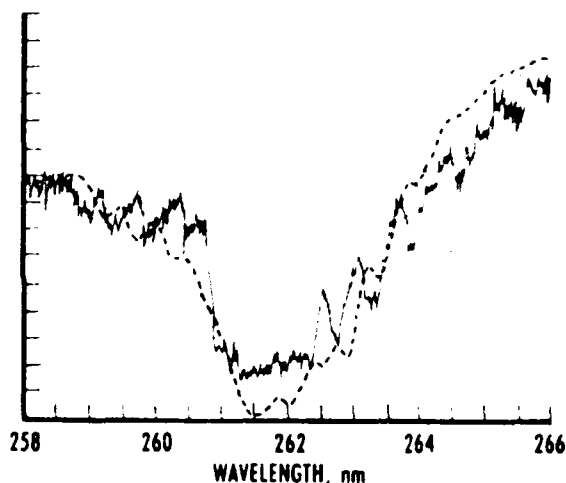


Fig. 6. Comparison of computed with measured spectra of SRM plume radiation at predicted plume conditions.

Only two results will be shown: First, the result using the predicted temperature and AlCl density, Fig. 6, and the final result, the best fit, Fig. 7. Referring to Figs. 4 and 5, the parameters used for the baseline calculations were

Vibrational Temperature, $T_v = 800$ K
 Rotational Temperature, $T_r = 800$ K
 Number Density, $n = 2.36 \times 10^{16}$ cm⁻³
 Absorber Path Length, $\ell = 5$ cm

Quite clearly from Fig. 6, the fit of the calculation to the data is poor. The structure is much smoother than the data, although the same spectral resolution was used in the model as for the spectrometer. In the process of iterating on temperature and density,

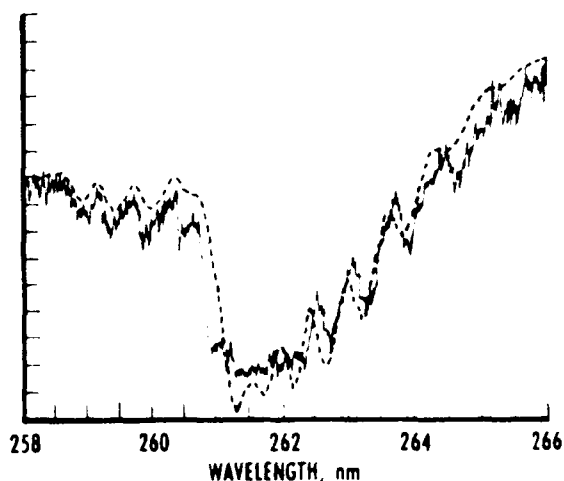


Fig. 7. Comparison of computed with measured spectra of SRM plume radiation at "best fit" conditions.

a better fit could be found in the central portion of the absorption, but the structure in the wings became poorer. It was this lack of spectral fit that led to use of the more accurate data for the $X^1\Sigma$ state. Finally, since the molecular data for the upper state are somewhat dated and some uncertainty exists in the relative values of the rotational constant for the two isotopes, the vibrational and rotational constants of the $A^1\Pi$ state were also iterated. Since this process could go on and on, a final fit, Fig. 7, was produced. The final parameters are as follows:

Vibrational Temperature, $T_v = 1,000$ K
 Rotational Temperature, $T_r = 1,000$ K
 Number Density, $n = 2.36 \times 10^{16} \text{ cm}^{-3}$
 Absorber Path Length, $l = 5 \text{ cm}$

and the molecular constants were also shifted

ω_e 450 $\rightarrow$ 430 cm^{-1}
 $\omega_e x_e$ 4.37 $\rightarrow$ 4.1 cm^{-1}
 B_e 0.259 $\rightarrow$ 0.249 cm^{-1}

Here, the spectral match is fair and the overall absorption matches pretty well, so that continued iteration appears unwarranted.

The final result is reasonable. Because of the high particle temperature, it is not surprising to have a gas temperature somewhat larger than predicted (1,000 K versus 800 K predicted). Also, because of small particles in the outer gas flow it is not surprising that the radiation from the gas layer masks the absorption at the deepest absorption frequency. One might expect that an arbitrary shift of the wavelength for the experimental data would produce a better fit. However, attempts to make this match were only marginally successful. The simple physical

model of Fig. 5 cannot be correct, since the radiation is obviously extended and the AlCl exists all through the plume. However, the modeling brought assurance that the absorber was AlCl, and only a slight variation of temperature brought the model results very near the measured data.

Concluding Remark

Clearly, if the AlCl feature is to be used for diagnostics, laboratory work using a heated absorption cell will be required to reach a better fit between calculated and measured spectra. In a heated cell experiment the temperature and pressure parameters would be well known, leaving only the spectrometer wavelength calibration and the molecular constants unknown. The issues raised above can then be resolved.

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