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EQUILIBRIUM SPEED OF SOUND IN A DISSOCIATING GAS

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An expression is obtained for the speed of sound in dissociating nitrogen tetroxide with consideration of the nonideality of the gas.

Dissociating nitrogen tetroxide shows promise as a heat-exchange agent and working substance in nuclear energy equipment.

To derive an expression for the equilibrium speed of sound in this system (gas), we will use the formula

$$a=\sqrt{\left(\frac{\partial p}{\partial \rho}\right)_s}, \tag{1}$$

where s indicates isentropic conditions. Expressing a in terms of the specific volume, we obtain

$$a^2=-v^2\left(\frac{\partial p}{\partial v}\right)_s. \tag{2}$$

For the partial derivative in Eq. (2) we will use the formula [1]

$$\left(\frac{\partial p}{\partial v}\right)_s=-\frac{C_p}{C_v}\frac{\left(\frac{\partial p}{\partial T}\right)_v}{\left(\frac{\partial v}{\partial T}\right)_p}. \tag{3}$$

There is a relationship between the partial derivatives of the function of two independent variables:

$$\left(\frac{\partial v}{\partial p}\right)_T\left(\frac{\partial p}{\partial T}\right)_v\left(\frac{\partial T}{\partial v}\right)_p=-1. \tag{4}$$

Defining $(\partial p/\partial T)_v$ from this expression and substituting in Eq. (3), we obtain

$$\left(\frac{\partial p}{\partial v}\right)_s=\frac{C_p}{C_v}\frac{\left(\frac{\partial p}{\partial T}\right)_v}{\left(\frac{\partial v}{\partial p}\right)_T}. \tag{5}$$

We will now use the differential equation [1]

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TABLE 1. Coefficients a_{ij} of Interpolation Equation of State [3] for Eq. (14)

j	a_{1j}	a_{2j}	a_{3j}
0	6,724331	—1,920155	4,22444
1	—13,12774	—0,2206247	—5,990096
2	—4,561842	1,044964	—8,061043
3	21,69903	10,70347	16,69250
4	—10,85284	—9,995447	—6,835001

$$C_p - C_v = T\left(\frac{\partial p}{\partial T}\right)_v \left(\frac{\partial v}{\partial T}\right)_p \tag{6}$$

and the equation of state of dissociating nitrogen tetroxide in the form

$$pv = Z_{\text{ef}} \frac{R}{\mu_{N_2O_4}} T, \tag{7}$$

where the coefficient Z_{ef} considers both dissociation and the nonideal nature of the gas. Substituting in Eq. (6) the value of $(\partial p/\partial T)_v$ from Eq. (3), and considering the resulting equation together with Eqs. (2), (7), we obtain

$$a^2 = -\frac{1}{\left(\frac{\partial v}{\partial p}\right)_T + \frac{T}{C_{p\text{ef}}}\left[\left(\frac{\partial v}{\partial T}\right)_p\right]^2}\left(Z_{\text{ef}} \frac{R}{\mu_{N_2O_4}} \frac{T}{p}\right)^2. \tag{8}$$

On the basis of Eq. (7), we express the partial derivatives appearing in Eq. (8) as follows:

$$\left(\frac{\partial v}{\partial p}\right)_T = -\frac{RT}{\mu_{N_2O_4} p^2}\left[Z_{\text{ef}} - p\left(\frac{\partial Z_{\text{ef}}}{\partial p}\right)_T\right], \tag{9}$$

$$\left(\frac{\partial v}{\partial T}\right)_p = \frac{R}{\mu_{N_2O_4} p}\left[Z_{\text{ef}} + T\left(\frac{\partial Z_{\text{ef}}}{\partial T}\right)_p\right]. \tag{10}$$

With consideration of Eqs. (9), (10) and introduction of the notation

$$\eta = Z_{\text{ef}} - p\left(\frac{\partial Z_{\text{ef}}}{\partial p}\right)_T, \tag{11}$$

$$\omega = Z_{\text{ef}} + T\left(\frac{\partial Z_{\text{ef}}}{\partial T}\right)_p \tag{12}$$

Eq. (8) takes on the form

$$a = \sqrt{\frac{R}{\mu_{N_2O_4}} T}\,\frac{Z_{\text{ef}}}{\sqrt{\eta - \frac{R}{\mu_{N_2O_4} C_{p\text{ef}}}\omega^2}}. \tag{13}$$

For our purposes the most suitable equation of state for dissociation nitrogen tetroxide will be that presented in [2]:

$$p = \frac{RT(1 + \alpha_{10} + \alpha_{10}\alpha_{20})}{v\mu_{N_2O_4}}\left(1 + \sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}\right), \tag{14}$$

where $\tau = T/T_{cr}$; $\pi = p/p_{cr}$; $T_{cr} = 431.35$°K; $p_{cr} = 102.492$ bar.

Comparing Eqs. (17) and (14), we obtain

$$Z_{\text{ef}} = (1 + \alpha_{10} + \alpha_{10}\alpha_{20})\left(1 + \sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}\right). \tag{15}$$

From Eqs. (13) and (15) we find

$$a = (1 + \alpha_{10} + \alpha_{10}\alpha_{20})\left(1 + \sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}\right)\sqrt{\frac{RT}{\mu_{N_2O_4}\left(\eta - \frac{R}{\mu_{N_2O_4}C_{p\text{ef}}}\omega^2\right)}}. \tag{16}$$

TABLE 2. Equilibrium Speed of Sound in Dissociating Nitrogen Tetroxide

T, K	a_{exp}	a_{calc}	$\frac{a_{\text{exp}}-a_{\text{calc}}}{a_{\text{exp}}}100\%$
$p = 1$ bar		$T_s = 294$ °K	
350	246,1	243,9	0,89
360	256,6	257,1	—0,19
370	266	266,9	—0,33
380	274,6	273,8	0,29
390	282,4	280	0,84
$p = 10$ bar		$T_s = 352$ °K	
400	261,7	258,6	1,18
410	271,9	268,7	1,17
420	281,9	279,5	0,85
$p = 50$ bar		$T_s = 405$ °K	
450	274,3	273,2	0,40

The degree of dissociation in the first and second stages of the reaction is usually determined by simultaneous solution of two equations expressing the relationship between the equilibrium constants and the unknown degree of dissociation. This relationship was presented in [2].

For further operations on Eq. (15) we introduce the notation

$$\varepsilon = 1 + \alpha_{10} + \alpha_{10}\alpha_{20}, \tag{17}$$

$$Z = 1 + \sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}, \tag{18}$$

i.e.,

$$Z_{\text{ef}} = \varepsilon Z. \tag{19}$$

We will write an expanded expression for the coefficient η. From Eqs. (17), (19)

$$\left(\frac{\partial Z\text{ef}}{\partial p}\right)_T = \varepsilon\left(\frac{\partial Z}{\partial p}\right)_T + Z\left(\frac{\partial \varepsilon}{\partial p}\right)_T$$

or

$$\left(\frac{\partial Z\text{ef}}{\partial p}\right)_T = (1 + \alpha_{10} + \alpha_{10}\alpha_{20})\left(\frac{\partial Z}{\partial p}\right)_T + Z\left[(1 + \alpha_{20})\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T + \alpha_{10}\left(\frac{\partial \alpha_{20}}{\partial p}\right)_T\right]. \tag{20}$$

Substituting this expression in Eq. (11), we obtain

$$\eta = (1 + \alpha_{10} + \alpha_{10}\alpha_{20})\left(1 + \sum_{i=1}^{3}\sum_{i=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}\right) - p\left\{(1 + \alpha_{10} + \alpha_{10}\alpha_{20})\left(\frac{\partial Z}{\partial p}\right)_T + \right.$$
$$\left. + \left(1 + \sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^i}\right)\left[(1 + \alpha_{20})\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T + \alpha_{10}\left(\frac{\partial \alpha_{20}}{\partial p}\right)_T\right]\right\}. \tag{21}$$

Expanding Eq. (18) and taking the derivative, we have

$$\left(\frac{\partial Z}{\partial p}\right)_T = \frac{1}{p}\left[\pi\left(a_{10} + \frac{a_{11}}{\tau} + \frac{a_{12}}{\tau^2} + \frac{a_{13}}{\tau^3} + \frac{a_{14}}{\tau^4}\right) + 2\pi^2\left(a_{20} + \frac{a_{21}}{\tau} + \right.\right.$$
$$\left.\left. + \frac{a_{22}}{\tau^2} + \frac{a_{23}}{\tau^3} + \frac{a_{24}}{\tau^4}\right) + 3\pi^3\left(a_{30} + \frac{a_{31}}{\tau} + \frac{a_{32}}{\tau^2} + \frac{a_{33}}{\tau^3} + \frac{a_{34}}{\tau^4}\right)\right]. \tag{22}$$

The coefficients a_{ij} are presented in Table 1.

We now derive expressions for the partial derivatives $(\partial\alpha_{10}/\partial p)_T$ and $(\partial\alpha_{20}/\partial p)_T$. We use the formula [1]

$$\left(\frac{\partial \ln K_{N_0}}{\partial p}\right)_T = -\sum \nu_i \frac{1}{p}, \tag{23}$$

where K_{N_0} is the equilibrium constant expressed in terms of molar fractions; $\sum \nu_i$ is the change in number of moles due to the reaction. Since

$$K_{p0} = K_{N_0} p^{\Sigma \nu_i}, \tag{24}$$

the relationship for K_{p_0} is known, the equilibrium constant K_N can be written as

$$K_{N_{10}} = \frac{4\alpha_{10}^2 (1-\alpha_{20})^2}{(1-\alpha_{10})(1+\alpha_{10}+\alpha_{10}\alpha_{20})} p^{1-(\Sigma\nu_i)_1}, \tag{25}$$

$$K_{N_{20}} = \frac{\alpha_{10}\alpha_{20}^3}{(1-\alpha_{20})^2(1+\alpha_{10}+\alpha_{10}\alpha_{20})} p^{1-(\Sigma\nu_i)_2}. \tag{26}$$

We find the partial derivative

$$\left(\frac{\partial K_{N_{10}}}{\partial p}\right)_T = \left(\frac{\partial \alpha_{10}}{\partial p}\right)_T \frac{4\alpha_{10}(1-\alpha_{20})^2(2+\alpha_{10}\alpha_{20})}{(1-\alpha_{10})^2(1+\alpha_{10}+\alpha_{10}\alpha_{20})^2} - \left(\frac{\partial \alpha_{20}}{\partial p}\right)_T \frac{4\alpha_{10}^2(1-\alpha_{20})(1-\alpha_{10})(2+3\alpha_{10}+\alpha_{10}\alpha_{20})}{(1-\alpha_{10})^2(1+\alpha_{10}+\alpha_{10}\alpha_{20})} -$$

$$-\frac{4\alpha_{10}^2(1-\alpha_{20})^2\beta_1}{(1-\alpha_{10})(1+\alpha_{10}+\alpha_{10}\alpha_{20})} p^{\beta_1-1}, \tag{27}$$

where

$$\beta_1 = 1-\left(\sum \nu_i\right)_1, \ \beta_2 = 1-\left(\sum \nu_i\right)_2. \tag{28}$$

Dividing Eq. (27) by Eq. (25) and considering Eq. (23), we obtain

$$\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T \frac{2+\alpha_{10}\alpha_{20}}{(1-\alpha_{10})(1+\alpha_{10}+\alpha_{10}\alpha_{20})\alpha_{10}} - \left(\frac{\partial \alpha_{20}}{\partial p}\right)_T \frac{2+3\alpha_{10}+\alpha_{10}\alpha_{20}}{(1-\alpha_{20})(1+\alpha_{10}+\alpha_{10}\alpha_{20})} + \frac{\beta_1}{p} = -\frac{1}{p}\left(\sum \nu_i\right)_1. \tag{29}$$

Taking the partial derivative of Eq. (26), after corresponding transformations we obtain

$$\left(\frac{\partial K_{N_{20}}}{\partial p}\right)_T = \left[\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T \frac{\alpha_{20}^3(1-\alpha_{20})}{(1-\alpha_{20})^3(1+\alpha_{10}+\alpha_{10}\alpha_{20})^2} + \left(\frac{\partial \alpha_{20}}{\partial p}\right)_T \frac{\alpha_{10}\alpha_{20}^2(3-\alpha_{20}+3\alpha_{10}+\alpha_{10}\alpha_{20})}{(1-\alpha_{20})^3(1+\alpha_{10}+\alpha_{10}\alpha_{20})^2}\right] p^{\beta_2} +$$

$$+\frac{\alpha_{10}\alpha_{20}^3\beta_2}{(1-\alpha_{20})^2(1+\alpha_{10}+\alpha_{10}\alpha_{20})} p^{\beta_2-1}. \tag{30}$$

Dividing Eq. (30) by Eq. (26) and considering Eq. (23), we have

$$\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T \frac{1}{\alpha_{10}(1+\alpha_{10}+\alpha_{10}\alpha_{20})} + \left(\frac{\partial \alpha_{20}}{\partial p}\right)_T \frac{3-\alpha_{20}+3\alpha_{10}+\alpha_{10}\alpha_{20}}{(1-\alpha_{20})\alpha_{20}(1+\alpha_{10}+\alpha_{10}\alpha_{20})} + \frac{\beta_2}{p} = -\frac{1}{p}\left(\sum \nu_i\right)_2. \tag{31}$$

We have now obtained two equations, i.e., Eqs. (29), (31), with two unknowns. We solve this system for $(\partial\alpha_{10}/\partial p)_T$ and $(\partial\alpha_{20}/\partial p)_T$:

$$\left(\frac{\partial \alpha_{10}}{\partial p}\right)_T = -\frac{\alpha_{10}(1-\alpha_{10})}{6p}(1+\alpha_{10}+\alpha_{10}\alpha_{20})(3+\alpha_{20}), \tag{32}$$

$$\left(\frac{\partial \alpha_{20}}{\partial p}\right)_T = -\frac{\alpha_{20}(1-\alpha_{20})}{6p}(1+\alpha_{10}+\alpha_{10}\alpha_{20}). \tag{33}$$

Substituting in Eq. (21) the values of the partial derivatives from Eqs. (32), (33), we have

$$\eta = (1+\alpha_{10}+\alpha_{10}\alpha_{20})\left(1+\sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^i}\right) - p\left\{(1+\alpha_{10}+\alpha_{10}\alpha_{20})\left(\frac{\partial Z}{\partial p}\right)_T\right.$$

$$-\left(1+\sum_{i=1}^{3}\sum_{j=0}^{4}\frac{a_{ij}\pi^i}{\tau^j}\right)\left[(1+\alpha_{20})\frac{\alpha_{10}(1-\alpha_{10})}{6p}(1+\alpha_{10}+\right.$$

$$\left.\left.+\alpha_{10}\alpha_{20})(3+\alpha_{20}) + \frac{\alpha_{10}\alpha_{20}(1-\alpha_{20})}{6p}(1+\alpha_{10}+\alpha_{10}\alpha_{20})\right]\right\}. \tag{34}$$

To derive the ω which appears in Eq. (16), it is necessary, according to Eq. (12), to first obtain an expression for the partial derivative $(\partial Z_{ef}/\partial T)_p$. From Eq. (19)

$$\left(\frac{\partial Z_{ef}}{\partial T}\right)_p = \varepsilon \left(\frac{\partial Z}{\partial T}\right)_p + Z \left(\frac{\partial \varepsilon}{\partial T}\right)_p$$

and we have

$$\omega = (1 + \alpha_{10} + \alpha_{10}\alpha_{20}) \left(1 + \sum_{i=1}^{3} \sum_{j=0}^{4} \frac{a_{ij}\pi^i}{\tau^i}\right) + T\Bigg\{(1 + \alpha_{20} + \alpha_{10}\alpha_{20}) \left(\frac{\partial Z}{\partial T}\right)_p +$$

$$+ \left(1 + \sum_{i=1}^{3} \sum_{j=0}^{4} \frac{a_{ij}\pi^i}{\tau^j}\right) \left[(1 + \alpha_{20}) \left(\frac{\partial \alpha_{10}}{\partial T}\right)_p + \alpha_{10} \left(\frac{\partial \alpha_{20}}{\partial T}\right)_p\right]\Bigg\}. \tag{35}$$

The partial derivatives $(\partial\alpha_{10}/\partial T)_p$ and $(\partial\alpha_{20}/\partial T)_p$ were obtained in [5]:

$$\left(\frac{\partial \alpha_{10}}{\partial T}\right)_p = \frac{\alpha_{10}(1 - \alpha_{10})}{6RT^2} [\Delta H_{p1}(3 + 3\alpha_{10} - \alpha_{20} + \alpha_{10}\alpha_{20}) + \alpha_{20}\Delta H_{p2}(2 + 3\alpha_{10} + \alpha_{10}\alpha_{20})], \tag{36}$$

$$\left(\frac{\partial \alpha_{20}}{\partial T}\right)_p = \frac{\alpha_{20}(1 - \alpha_{20})}{6RT^2} [\Delta H_{p1}(2 + \alpha_{10}\alpha_{20}) - \Delta H_{p2}(1 - \alpha_{10})], \tag{37}$$

where ΔH_{p1} and ΔH_{p2}, i.e., the thermal effects of the first and second stages of the chemical reaction, are equal to [3]; $\Delta H_{p1} = 623.4$ kJ/kg in the temperature range 300-450°K; $\Delta H_{p2} = 1225.9$ kJ/kg in the range 400-1300°K. If in Eq. (37) we take the value $R = 8314$ J/mole·deg, then $\Delta H_{p1} = 623.4 \cdot 10^3 \cdot 92$ J/kmole, $\Delta H_{p2} = 1225.9 \cdot 10^3 \cdot 92$ J/kmole.

Expanding Eq. (18) and taking the derivative, we obtain an expression for the partial derivative $(\partial Z/\partial T)_p$, appearing in Eq. (34) in the form

$$\left(\frac{\partial Z}{\partial T}\right)_p = -\frac{1}{T}\left[\pi\left(\frac{a_{11}}{\tau} + \frac{2a_{12}}{\tau^2} + \frac{3a_{13}}{\tau^3} + \frac{4a_{14}}{\tau^4}\right) + \pi^2\left(\frac{a_{21}}{\tau} + \frac{2a_{22}}{\tau^2} + \frac{3a_{23}}{\tau^3} + \frac{4a_{24}}{\tau^4}\right) + \right.$$

$$\left. + \pi^3\left(\frac{a_{31}}{\tau} + \frac{2a_{32}}{\tau^2} + \frac{3a_{33}}{\tau^3} + \frac{4a_{34}}{\tau^4}\right)\right]. \tag{38}$$

The coefficients a_{1j}, a_{2j}, and a_{3j} are taken from Table 1.

Thus, to find the value of ω in Eq. (35) we first define the value of $(\partial Z/\partial T)_p$ with Eq. (38). Substituting in Eq. (35) the value of the partial derivatives from Eqs. (36), (37), we obtain

$$\omega = Z(1 + \alpha_{10} + \alpha_{10}\alpha_{20}) + T\Bigg[(1 + \alpha_{10} + \alpha_{10}\alpha_{20}) \left(\frac{\partial Z}{\partial T}\right)_p + \left(1 + \sum_{i=1}^{3} \sum_{j=0}^{4} \frac{a_{ij}\pi^i}{\tau^j}\right) \Bigg\{(1 + \alpha_{20}) \frac{\alpha_{10}(1 - \alpha_{10})}{6RT^2} [\Delta H_{p1}(3$$

$$+ 3\alpha_{10} - \alpha_{20} + \alpha_{10}\alpha_{20}) + \alpha_{20}\Delta H_{p2}(2 + 3\alpha_{10} + \alpha_{10}\alpha_{20})] + \frac{\alpha_{10}\alpha_{20}(1 - \alpha_{20})}{6RT^2} [\Delta H_{p2}(2 + \alpha_{10}\alpha_{20}) - \Delta H_{p1}(1 - \alpha_{10})]\Bigg\}\Bigg]. \tag{39}$$

After finding η with Eq. (34) and ω with Eq. (39) at specified values of p, T, and $C_{p_{ef}}$, we finally use Eq. (13) to determine the equilibrium speed of sound a.

Table 2 shows values of the equilibrium speed of sound in the gaseous phase as obtained by the formula derived herein, at certain pressures and temperatures also presented in [2] and taken from the study of Nikolaev and Timofeev (Vests. Akad. Nauk BSSR, Ser. Fiz. Energ. Navuk, No. 3 (1974)). Values of isobaric heat capacity for the calculations were taken from [4].

We see that even near the saturation line, at temperatures exceeding the saturation temperature by about 50°, the deviation is less than 1%, and only reaches 1.17 and 1.18% at two points. This indicates the accuracy and reliability of the formula obtained for the speed of sound in the gas, which can be used with experimental data on isobaric heat capacity.

NOTATION

p, v, T, pressure, volume, and temperature; R, universal gas constant; $\mu_{N_2O_4}$, molecular mass; C_V, C_p, isochoric and isobaric specific heats; Z, compressibility coefficient; $C_{p_{ef}}$ effective specific heat at constant pressure; Z_{ef}, effective coefficient considering effects of dissociation and nonideality; K_{p10}, K_{p20}, temperature dependent equilibrium constants for first and second reaction stages for an ideal gas mixture; α_{10}, α_{20}, degree of dissociation of first and second reaction stages in ideal gas state.

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CHARACTERISTICS OF A GAS-DISCHARGE CO-LASER IN GENERATION ON AN OVERTONE

1. STEADY-STATE REGIME

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The work presents the parametric dependences of the energy and spectral characteristics of the gas-discharge CO-laser in steady-state regime in generation on the first overtone on the gas temperature, the pumping power, and the cavity Q-factor of the resonator. The article examines questions of the selection of the individual vibrational and rotational lines. Comparisons with experimental results are made.

At present there is considerable interest in lasers on vibrational overtone of the carbon monoxide molecule [1-4]; this is due to the potentially high effectiveness of such a laser close to the IR range and the possibility of using it for problems of laser chemistry, isotope separation, etc. We will also demonstrate that the investigation of this laser makes it possible to elaborate on the kinetics of CO molecules, especially at high vibrational levels. Knowledge of the theoretical parametric dependences obtained on the basis of numerical modeling makes it possible to predict the expected characteristics of such a laser, and it stimulates and facilitates its experimental investigation. The present work consists in the theoretical investigation of the characteristics of the gas-discharge CO-laser operating in steady-state regime of generation on the first overtone. We analyze the parametric dependences of the energy and spectral characteristics on the gas temperature, the pumping power, and the cavity Q-factor of the resonator. We also examine questions of the selection of individual vibrational and rotational lines.

Previously, we substantiated the possibility of obtaining generation on vibrational overtones of carbon monoxide molecules with pumping in a stationary and pulsed electric discharge [1]. An analysis of the population density of vibrational levels of CO molecules indicated that it is possible to effect partial inversion at the transitions $J-1$, $v \to J$,

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