A Hydrogen Sensor based on Acoustic Rotational Relaxation Spectra

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Abstract

As an energy carrier and a colorless hazardous and flammable gas, hydrogen has a wide range of applications. In this paper, we propose a new model based on acoustic rotational relaxation spectra for hydrogen sensor. The simulation results show good agreements between our model and experiments. It has a potential to be used for natural gas detection with blending hydrogen.

Keywords: hydrogen sensor, acoustic relaxation spectra, gas detection

Introduction

Hydrogen is a useful, hazardous and colorless flammable gas. It has been a popular research topic for over 100 years. There are approximately eight types in the main existing and emerging hydrogen sensing technologies [1]. Ultrasonic acoustic is ongoing and less well-developed technology of hydrogen sensors, which has many advantages such as very wide detection range, low power consumption, on-line measurement.

There are many publications reported for gas sensors based on acoustic spectra such as Yi Hu and Ming Zhu [2]. But few paper research H_2 based on acoustic rotational relaxation spectra. In this paper we provide a hydrogen sensor based on acoustic rotational relaxation spectra. There are usually three methods of hydrogen detection in acoustic relaxation research. Takayanagi and Kishimoto [3], Montero and Pérez-Ríos [4] had calculated the rotational heat capacity for H_2 using the quantum-mechanical method of distorted waves. Rhode [5], Bauer and Bass [6] had developed an efficient method to relate the transition rates for rotational energy transfer to the acoustic dispersion and absorption using irreversible thermodynamics. Geide [7], Sluijter, Knaap and Beenakker [8] had obtained the data of rotational absorption by experiments.

The present paper is organized as follows. In Sec. II we introduce the analytical model of acoustic rotational relaxation spectra for hydrogen. In Sec. III we simulate the model and discuss the result at different ambient temperatures. In Sec. IV we conclude this paper.

Basic Principles

According to the definition of enthalpy and the Amagat law, the enthalpy of hydrogen is the sum of translation, rotation and vibration of hydrogen molecules. The enthalpy of ideal gas mixtures can be expressed as:

$$H = H_{trans}(T) + H_{rot}(T) + H_{vib}(T) \quad \text{(Eq. 1)}$$

Where H is the sum of total enthalpy, $H_{trans}(T)$ is the translation enthalpy, $H_{rot}(T)$ is the rotational enthalpy, $H_{vib}(T)$ is the vibrational enthalpy.

Compared with rotation mode, the vibration mode of hydrogen molecules is so small that it can be ignored. Thus Eq. (1) can be approximately obtained as:

$$H = H_{trans}(T) + H_{rot}(T) \quad \text{(Eq. 2)}$$

Deriving Eq. (2), we obtain

$$dH = C_p dT$$

(Eq. 3)

Using Eq. (2) and Eq. (3), the following is the total expression of the hydrogen rotational and translational relaxation:

$$C_p^{rot} = \sum_{j=0}^{n} \frac{E_j dN_j}{dT^{trans}} \quad \text{(Eq. 4)}$$

$$C_p^{eff} = C_p^{tr} + C_p^{rot} \quad \text{(Eq. 5)}$$

Where C_p^{eff} is the sum of all heat capacity, $C_p^{tr} = 3R/2$ is the translational heat capacity, C_p^{rot} is the rotational effective heat capacity. According to Eq. (5), we can get the value of C_p^{eff} .

$$c_e^2 = \frac{P_0}{\rho_0}\gamma^{eff} = \frac{P_0}{\rho_0}\frac{C_p^{eff}}{C_p^{eff} - R} \quad \text{(Eq. 6)}$$

Where c_e is the effective thermodynamic sound speed, P_0 is the equilibrium pressure, ρ_0 is the equilibrium density. R is the universal gas constant, γ^{eff} is the effective isochoric heat ratio.

Because the relationship between c_e and k_e, we can also obtain the expression in terms of

$$k_e = \frac{\omega}{c} - i\alpha_r = \frac{\omega}{c_e} \qquad \text{(Eq. 7)}$$

Where k_e is the effective acoustic wave number, $\omega = 2\pi f$ is the acoustic angular frequency, $i = \sqrt{-1}$, c is the sound speed depend on frequency, α_r is the molecular relaxation absorption coefficient.

From the relationship between acoustic formula of thermodynamics and the effective wave number, Eq. (6) and Eq. (7), the relaxation absorption of hydrogen can be obtained. Derived from the above formulas, we can draw a conclusion that hydrogen absorption process mostly caused by rotational absorption.

Simulations and Discussion

We firstly consider acoustic rotational absorption in a pure hydrogen gas at different temperatures. Natural hydgrogen gas is a mixture of three parts orthohydgogen to one part parahydgrogen. Parahydrogen have two rotational relaxation processes corresponding to $J = 0 \rightarrow 2$ and $J = 2 \rightarrow 4$ transitions when orthohydrogen have two $J = 1 \rightarrow 3$ and $J = 3 \rightarrow 5$ transitions. Some parameters of natural hydgrogen are shown in Table 1. Sigma is the rotational relaxation cross section and Bulk_visc is the volume viscosity. The proportion and energy of rotational transitions in natural hydgrogen at 296 K are shown in Table 2.

Table 1. Some parameters for natural hydgrogen at room temperature.

T	Sigma	Bulk_visc
300	0.0982	266
296	0.0978	263
275	0.0968	257

Table 2. The proportion and energy of rotational transitions in natural hydgrogen at 296 K.

Transition	% of total	Energy (cm^{-1})
0--2	18.7	349
2--4	9.3	801
4--6	0.3	1292
1--3	66.4	586
3--5	5.2	1062
5--7	0.3	1530

Fig. 1 and Fig. 2 present acoustic absorption and sound speed spectra for hydrogen gas at different temperatures. From the pictures, we can see our acoustic absorption curves agree very well with the experiment values at low frequencies at T=296 K and T=873 K. The predicted vales of the model are slightly higher than experiment data at T=296 K, while they are slightly lower at T=873 K. The reason for this phenomenon is that the relaxation process of hydrogen at high temperature is more active. The experiment values of sound speed in hydrogen at low frequencies are high than our model values at T=296 K.

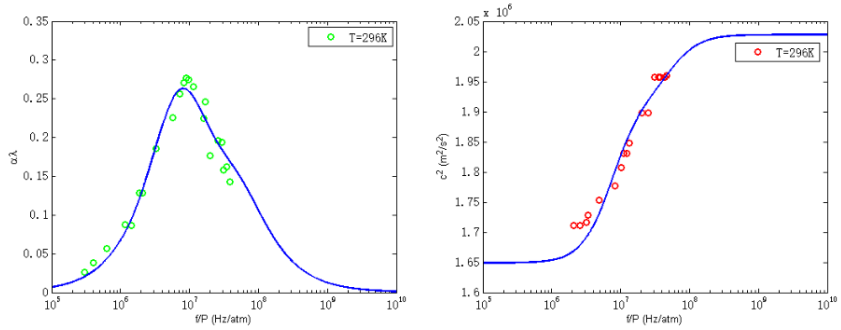

Fig.1. (a) Acoustic absorption in hydrogen. (b) Sound speed in hydrogen. T=296 K, blue curve represents the spectrums predicted by the model of this work at the same temperature, green and red circles represent the experimental data from the literature [9].

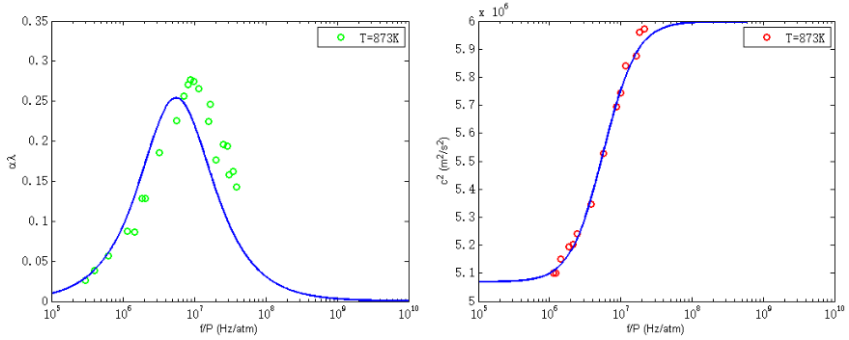

Fig. 2. (a) Acoustic absorption in hydrogen. (b) Sound speed in hydrogen. T=873 K, blue curve represents the spectrums predicted by the model of this work at the same temperature, green and red circles represent the experimental data from the literature [9].

Conclusion and outlook

In this paper, we present a model of hydrogen sensor based on acoustic relaxation spectra. It provides deep insight into the relation between the entropy of hydrogen and its rotational relaxation. At T=296 K and T=873 K, the hydrogen relaxation spectra based on the proposed model are consistent with experimental data quite well. Those comparisons verify our model. However, the relaxation processes of methane, nitrogen and many other gases are mainly dominated by vibrational relaxation process. Taking into account the difference between other gases and hydrogen, it can be further used to detect composition and concentration in gas mixtures such as hydrogen compression natural gas.

Acknowledgements

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