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The mechanisms of thermal diffusion and baro-diffusion effects on thermacoustic mixture separation

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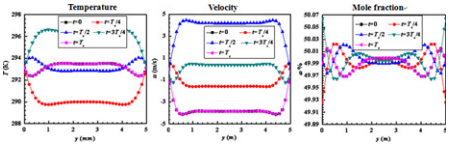
HIGHLIGHTS

- A new numerical method has been developed to study thermacoustic separation.
- The difference between the numerical and experimental values is no more than 5.8%.
- It is shown that only the radial gradient can cause separation of the mixture.
- Thermal diffusion performs as main driving force of thermoacoustic separation.
- The detailed process of thermoacoustic mixture separation is revealed.

GRAPHICAL ABSTRACT

Temperature, velocity and mole fraction distribution along the radial direction in a cycle.



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ABSTRACT

Thermoacoustic separation is a totally new method for separating gas mixtures and is also considered as an effective mean of separating even isotopes and azeotropes. In this paper, a two-dimensional numerical method based on the compressible SIMPLE algorithm has been developed to study the mechanisms of thermal diffusion and baro-diffusion effects on thermacoustic mixture separation. It is found that the numerical results are in good agreement with experimental data and theoretical predictions from published literatures. Furthermore, contributions of thermal diffusion and baro-diffusion to thermoacoustic separation are studied, respectively. It is shown that only the radial gradient can cause separation of mixture and thermal diffusion caused by radial temperature gradient performs as the main driving force of thermoacoustic separation. Thus, temperature, velocity, and concentration distributions along the radial direction have been studied to investigate the detailed process of thermoacoustic separation in the resonator. Under the combined action of sound wave and thermal diffusion, the heavy component is driven into pressure node during the expansion half cycle and the light component is driven into velocity node during the compression half cycle, making the mixture separated in the axis direction.

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1. Introduction

In 1777, Higgins enclosed a hydrogen diffusion flame on a large tube and found an organ-pipe oscillation (Putnam and Dennis, 1956). This is the first recorded observation of thermoacoustic effect. Based on the discovery of the thermoacoustic effect, great attention has been attracted from researchers in relating fields all over the world. Wheatley and Cox (1985) invented the first practical thermoacoustic heat engine, which is named "natural engine". After that, thermoacoustic refrigerator is also invented by Hofler (1986, 1988, 1993). Thermoacoustic heat engines and refrigerators realize the transformation between thermal energy and acoustic power. Here comes a question: Can we use sound wave to pump mass? The answer is absolutely affirmative.

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In recent years, researchers have developed a new mixture-separation process via sound wave. This method is referred to as thermoacoustic separation (Spoor and Swift, 2000).

Mixture separation remains an active field of chemical engineering and related industries (Zeng et al., 2011; Liu et al., 2011). Because of the wide range of applications for separation, no single technique meets all possible demands. Geller and Swift (2002b) showed that when separating mixtures of isotopes or azeotropes, the efficiency of thermoacoustic mixture separation was actually of the same order of magnitude as that of other widely used separation processes, such as conventional thermal diffusion (Clusius and Dickel, 1938; Yeh, 2001) and gaseous diffusion (Villani, 1979; Bilous and Doneddu, 1986). Compared with conventional thermal diffusion, thermoacoustic separation has three potential advantages. First, the temperature difference in thermoacoustic separation, which is due to the adiabatic compressions and expansions of sound wave, is far lower than the absolute mean temperature of the gas; in thermal-diffusion columns, though, the temperature difference between the walls is generally comparable to the absolute mean temperature (Jones and Furry, 1946); therefore, the efficiency of thermoacoustic separation has much room to be improved. Second, the mole fraction difference occurs due to acoustically driven in thermoacoustic separation, instead of being driven by natural convection as in thermal-diffusion columns, allowing thermoacoustic separation to work with any orientation, or even in the absence of gravity (Geller and Swift, 2009). Third, since thermoacoustic separation has potential advantage due to its mechanical simplicity, it is quite applicable to combine thermoacoustic separation with conventional thermal diffusion, gaseous diffusion or centrifugal separation (Willems et al., 2008) to further improve the separation efficiency. Thermoacoustic mixture separation, which holds the merits of having no moving parts, simple structure, high reliability, ability to operate at ambient temperature and pressure, and independence of gravity, has potential application prospects to become competitive, especially in the separation of hydrogen isotopes, uranium isotopes and stable isotopes for biological and medical use (Hanson, 2002).

Additionally, in thermoacoustic heat engines and refrigerators, using mixture working gases, which have relatively lower Prandtl numbers, is a quite useful method to improve the performance and efficiency (Giacobbe, 1994; Belcher et al., 1999; Tijani et al., 2002; Shen et al., 2009; Hu et al., 2010). However, the gas mixture will be partially separated when a sound wave propagates through the thermoacoustic engine, creating axial concentration gradients of the components. The densities, resonance frequencies and Prandtl numbers of the mixture gases change with the concentration gradually in the separation process. In this case, the operation stability, reliability and efficiency of thermoacoustic engines will be affected. In fact, thermoacoustic separation was firstly recognized when an anomalous difference (up to 3%) was observed in the resonance frequencies of the two resonators when using a He–Xe mixture (Swift and Spoor, 1999). And what's more, the amplitude of sound wave will be larger and larger to improve the power and efficiency of thermoacoustic engines for practical applications (Dai et al., 2007; Yu et al., 2007; Sun et al., 2008; Ke et al., 2010). With the increase of amplitude, thermoacoustic separation effect will be much more significant. Consequently, it is of great theoretical interest and application value to study the mechanism and influence factors of thermoacoustic separation process.

Separation of gas mixtures using an acoustic wave was firstly systematically researched by Passau (1948), who performed experiments to separate hydrogen–carbon dioxide mixture using traveling wave. Passau supposed this separation was due to the influence of pressure gradient caused by traveling wave. Under the influence of baro-diffusion, the gas molecules are pushed in the propagation direction of the wave, the lighter molecules are pushed farther ahead of the wave front, leaving behind a mixture enriched with the heavier molecules. Thus, with repeated cycles, the mixture will be separated gradually along the wave-propagation direction. Eyraud (1956) carried out experiments to study the separation of a mixture of hydrogen and nitrogen and noted partial separation of the components. Noble et al. (1964) measured the velocity of sound wave in a binary mixture and observed a deviation from the expected theoretical value. He attributed it to a partial separation of the gas mixture. Tikhomirov et al. (1967) added an ultrasonic wave into a tube filled with a mixture of nitrogen and hydrogen. The resonant frequency was 670 kHz. After three hours, a mole fraction difference of 20% was measured at the end of the tube. All these early separations were attributed to the influence of the pressure gradient caused by ultrasonic waves. Thus, this sort of separation process of gas mixture is referred to as baro-diffusion separation.

An experiment was firstly conducted to investigate the possibility of separating gas mixture by using a standing wave in an acoustic resonator by Dykhne et al. (1982). The resonant frequency ranged from 500 to 550 Hz for an 86 cm resonator. The working gases were mixtures consisted of helium, argon, carbon dioxide, and xenon. Experimental results showed that the separation of the heavier component was about 10%. Then, they executed a series of experiments for mixtures of CO, CO_2, CF_2HCl, SF_6, and CF_3I (Dykhne et al., 1985). Their experimental results showed the separation rate is a function of the pressure gradient. However, the experimental separation rates were approximately ten times larger than the theoretical calculations. They guessed that the pressure gradient was not the only factor for the separation of gas mixtures in this acoustic resonator. The separation of gas mixtures in an acoustic resonator was attributed to both baro-diffusion and thermal diffusion. After that, Bozhdankevech et al. (1986) carried out experiments to investigate the effect of thermal diffusion using a similar experimental setup as described above. The results showed that baro-diffusion and thermal diffusion had the opposite effects in the separation of gas mixtures in an acoustic resonator.

Swift and Spoor (1999) observed an anomalous difference in the resonance frequencies in acoustically coupled acoustic resonators when using a He–Xe mixture. They concluded that the sound wave separated the He and Xe in the acoustic coupler. Then, they performed experiments of a He–Ar mixture in a thermoacoustic apparatus. The result showed a mole-fraction difference of 7% arised across the end of the duct in about 3 h (Spoor and Swift, 2000). This separation process is firstly referred to as thermoacoustic separation. During the past decade, Swift, Spoor and Geller have done much work to understand its consequences and its possible practical implementation of thermoacoustic separation both experimentally and theoretically (Geller and Swift, 2002a, 2004; Swift and Geller, 2006). They considered this separation approach in boundary-layer approximation could be an effective methodology of separating even isotopes by coupling the diffusion caused by thermal gradients at boundaries and transportation by the acoustic waves. Acoustic separation in ternary gas mixtures had been described by Howell (1988). Thermoacoustic condensation and evaporation of one component in a gas mixture had been also studied by Raspet et al. (1999), with oscillating mass diffusion of the condensing component.

Although thermoacoustic separation has attracted much attention of researchers around the world for many years, the theory itself is not very complete and much work remains to be done to understand the dynamics and mechanism of the separation process. Furthermore, no comprehensive model has been developed to

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