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Postprint: Experimental Study on Solar Enrichment of Low-Concentration Methane

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Abstract

During production, coal mines directly discharge substantial quantities of gas containing low-concentration methane into the atmosphere, causing severe energy waste and aggravating the greenhouse effect. This paper addresses the limitations of conventional gas enrichment technologies and proposes employing the Soret effect for separating mixed gases in coal mine gas and enriching methane. Through theoretical analysis, the initial methane concentration and temperature difference are identified as the key parameters influencing the enrichment process; through experiments and numerical simulation, the patterns by which these parameters influence enrichment performance are investigated.

Full Text

Preamble

Experimental Study on Enrichment of Low-Concentration Coal Bed Methane with Solar Energy

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Abstract

During coal production, large quantities of low-concentration methane gas are emitted directly into the atmosphere, resulting in serious energy waste and exacerbating the greenhouse effect. Aiming to address the shortcomings of conventional coal mine methane enrichment technologies, this paper proposes utilizing the Soret effect for separating gas mixtures and enriching methane in coal mine gas. Through theoretical analysis, the initial methane concentration and temperature difference were identified as the primary parameters affecting the

enrichment process. The influence patterns of these parameters on enrichment performance were investigated through experiments and numerical simulations.

Keywords: Thermal diffusion; Soret effect; Coal mine methane enrichment

1. Principle of Methane Enrichment via the Soret Effect

The Soret effect, also known as thermal diffusion, refers to the phenomenon where components in a uniform fluid mixture migrate toward the hot and cold walls of a container under the influence of a non-uniform temperature field, forming concentration gradients. First discovered and named by Swiss scientist J. Soret in 1879, this separation and enrichment process is influenced only by temperature differences and the physical properties of the mixture to be separated. It offers advantages including simple separation equipment, low investment, and high operational flexibility, and has been applied in fields such as isotope separation, separation of gas products from controlled thermonuclear reactors, and separation of isomeric organic compounds, demonstrating broad application prospects.

The temperature difference required for thermal diffusion can be provided by solar energy, which reduces system operating costs and energy consumption. According to research by Shen Wenming et al. [?], coating applications and transparent cover plates can help pipes absorb solar energy and maintain internal temperatures at 150°C. In this study, temperature-controlled heating plates were used to heat the experimental pipes within this temperature range to simulate solar heating effects.

Research on thermal diffusion shows that when a temperature gradient is applied to a uniformly concentrated binary system, thermal diffusion mass transfer occurs and establishes a stable concentration gradient [?]. The thermal diffusion coefficient can be positive, zero, or negative. A positive value indicates that a component concentrates in the cold region, with the temperature gradient driving mass transfer from high to low temperature; a negative value indicates concentration in the hot region, with mass transfer from low to high temperature. For a binary gas mixture of methane and nitrogen, the concentration equation for each component is:

$$\frac{\partial c_i}{\partial t} + \text{div}(c_i \mathbf{U}) = D_{12} \text{div}(\nabla \cdot c_i) \pm D_T \text{div}(\nabla \cdot \ln T) \quad (1)$$

where c_i is the molar concentration of component i ($\text{mol} \cdot \text{m}^{-3}$), with $i = 1, 2$ representing the heavy and light gases in the mixture, respectively; t is time (s); \mathbf{U} is velocity ($\text{m} \cdot \text{s}^{-1}$); D_{12} is the gas diffusion coefficient ($\text{m}^2 \cdot \text{s}^{-1}$); and $D_T = D_{12} \cdot k_T$ is the thermal diffusion coefficient ($\text{m}^2 \cdot \text{s}^{-1}$), where k_T is the thermal diffusion ratio. The heavy gas component takes the “+” sign, while the light gas component takes the “−” sign.

Given that the gas mixture in our experiments consists of methane and air, and air's main component is nitrogen with a similar relative molecular mass, the actual mixture was reasonably approximated as a binary methane-nitrogen mixture for theoretical analysis. According to Equation (1), under a temperature gradient, methane in the methane-nitrogen mixture transfers from the cold surface to the hot surface, while nitrogen transfers from the hot surface to the cold surface. Simultaneously, due to Fickian diffusion effects, diffusive mass transfer also occurs because of the resulting concentration gradient. Equation (1) shows that at any moment, the mass fluxes of the two gases at any cross-section are equal in magnitude and opposite in direction. For either gas, when the mass transfer rates of thermal diffusion and Fickian diffusion become equal, the concentration field reaches dynamic equilibrium.

Equation (1) indicates that factors affecting thermal diffusion mass transfer include component molar concentration, time, temperature, gas diffusion coefficient, and thermal diffusion ratio. The latter two are determined by the gas's physical properties and are constants for our research object. This study focuses on enrichment performance under steady-state conditions, with experiments allowing sufficient time for the system to reach steady state, thus time effects were not investigated.

Based on this physical effect, we designed a pipe with hot and cold surfaces, enabling methane in the methane-nitrogen mixture to achieve higher concentrations at the hot surface and nitrogen at the cold surface. If multiple such devices are cascaded—after sufficient heat and mass transfer time in the previous stage, gas with higher methane concentration is extracted from the hot side of the previous stage and fed into the next stage—the methane concentration in the gas extracted from each stage will be higher than that fed into it, thereby effectively enriching methane in the mixture.

2. Experimental Investigation

2.1 Experimental System

The experimental system used for methane enrichment is shown in [Figure 1: see original paper]. In the experiments, gas mixtures prepared from methane and air simulated coal mine gas. The flow rates of each gas were controlled via valves on methane cylinders and regulators on air pumps to match the designed experimental conditions for gas proportions and total flow rate. After entering the pipe, the mixture was rapidly homogenized through a flow straightener, and the internal temperature was measured by thermometers. The pipe's upper surface was covered with a temperature-adjustable heating plate, while the lower surface was cooled with an ice-water mixture, creating a substantial temperature difference across the pipe height. The pipe was 2 meters long with a rectangular cross-section of 150 mm width and 75 mm height.

Considering the Soret effect, the laminar-flowing gas mixture simultaneously moved along the pipe length while experiencing temperature gradients in the

height direction, causing the two components to diffuse toward the hot surface (upper surface) and cold surface (lower surface) according to Equation (1). At the pipe outlet, a flue gas analyzer extracted gas at different heights to investigate the methane concentration distribution along the height direction.

2.2 Experimental Results

The methane concentration in the figures refers to the molar concentration of methane in the gas mixture fed into the pipe. The temperature difference is the measured difference between gas temperatures at the hot wall and cold wall surfaces inside the pipe. The concentration ratio is defined as the ratio of methane concentration at the hot wall surface to that at the cold wall surface.

Given that methane is a flammable and explosive gas, experimental conditions were selected with methane concentrations below the explosion limit, and experiments were conducted with good ventilation to prevent methane accumulation. To ensure sufficient thermal diffusion mass transfer, the gas velocity inside the pipe was maintained low. Based on theoretical calculations and considering the actual pipe length, an average velocity of 0.001 m/s was selected. Due to equipment limitations, the achievable temperature difference within the pipe did not exceed 100°C. Experimental results are presented in [Figure 2: see original paper], [Figure 3: see original paper], and [Figure 4: see original paper].

[Figure 2: see original paper] shows that the concentration ratio, which characterizes enrichment performance, increases with temperature difference. For safety reasons, experiments at higher temperature differences were not conducted. [Figure 3: see original paper] and [Figure 4: see original paper] indicate that when the methane concentration of the inlet gas mixture increases, the concentration ratio at the same temperature difference also improves. This may be because increased methane concentration raises the absolute concentration difference under the same temperature difference, thereby eliminating or reducing the impact of negative factors that interfere with experimental results.

A blank experiment was conducted at 1.31% methane molar concentration without any temperature difference, yielding a concentration ratio of 0.998—approximately 1/5 to 1/15 of that under temperature difference conditions. This result falls within the measurement error range of the experimental instruments. Therefore, we conclude that temperature difference is a necessary condition for producing enrichment effects.

The concentration ratios calculated from experimental data show consistent trends with theoretical calculations, though discrepancies exist, particularly when temperature difference and inlet methane concentration are varied, causing significant fluctuations in the concentration ratio.

Under known conditions of hot surface temperature, cold surface temperature, and initial methane concentration, the single-stage theoretical concentration ratio for this system can be derived from Equation (1) as:

$$\eta_{th} = \frac{(1 + A)c + 2A}{(A - 1)c + 2}$$

where c is the methane mole fraction in the mixture; A is an intermediate variable, $A = e^{\alpha\Delta T}$, where α is the thermal diffusion constant (determined by physical properties and independent of temperature), T_h is the hot surface temperature (K), and T_c is the cold surface temperature (K).

Comparisons reveal that all experimentally obtained concentration ratios are lower than theoretical predictions. The differences between theoretical and experimental values, along with fluctuations in experimental data, are attributed to the following factors:

1. The principle of methane enrichment using temperature difference relies on a constant temperature field causing light and heavy molecules to diffuse toward hot and cold ends, respectively. In this experiment, the heating plate significantly affected the temperature at the cold wall surface, causing the temperature difference to decrease over time and preventing the establishment of a truly constant temperature field.
2. Theoretical calculations assumed adiabatic boundary conditions on both side walls of the transport pipe, whereas experimental boundaries were not adiabatic.
3. The pipe height was relatively small and gas flow rate was low; the flue gas analyzer's gas extraction during measurement affected the concentration and temperature field distributions.
4. A three-way device was installed at the analyzer probe for measurement purposes, and the flow meter readings controlling the extraction rate significantly impacted methane concentration measurements, introducing measurement errors.
5. The limited pipe length resulted in insufficient mixing of gases, leading to differences from theoretical separation limits.

3. Conclusions

1. Under temperature differences, coal mine gas undergoes thermal diffusion, with the light component methane transferring toward the hot surface and the heavy component nitrogen toward the cold surface until dynamic equilibrium is reached with Fickian diffusion. This physical phenomenon can be utilized to enrich methane in coal mine gas.
2. The parameters significantly affecting enrichment performance are initial methane concentration and temperature difference.
3. Under laminar flow conditions, the enrichment process of gas mixtures in pipes is strongly influenced by the temperature difference between upper

and lower surfaces; with other conditions constant, the maximum concentration difference of each component in the pipe increases with the maximum temperature difference across the cross-section.

4. At a given temperature difference, increasing pipe length or reducing gas velocity (both increasing gas residence time) can improve the enrichment concentration ratio.
5. Considering solar heating capacity, the temperature difference in practical systems is limited. Since the concentration ratio of a single-stage pipe is constrained by temperature difference, multiple such devices can be cascaded to increase methane concentration in the final product gas. After the previous stage reaches its maximum concentration ratio under the given temperature difference, gas with higher methane concentration is extracted from its hot side and fed into the next stage. The methane concentration in gas extracted from each stage will be higher than that fed into it, theoretically enabling the final product methane concentration to be increased to the required value under modest temperature differences.

4. Significance and Future Work

This system was originally conceived to address the energy waste and greenhouse effect caused by venting coal mine gas. Considering methane's importance as an energy and chemical feedstock, enriched coal mine gas can be further processed and purified for applications in energy, chemical, and other fields. This not only achieves greenhouse gas emission reduction but also recovers substantial chemical energy contained in coal mine gas, embodying positive significance for energy conservation, emission reduction, and waste valorization.

The work presented herein primarily proposes a new coal mine gas enrichment technology, verifying its technical feasibility and viability under certain natural conditions. Through experimental research and numerical simulation, the main factors affecting this process and their basic influence patterns have been identified. However, current conclusions remain fundamental, with significant gaps to industrial application. Further research may proceed in the following directions:

1. Construct larger-scale experimental pipes to identify and address potential issues when applying this technology to industrial practice.
2. Employ high-performance numerical simulation workstations for larger-scale simulations. As this system represents an industrial application of a micro-scale mass transfer process spanning multiple orders of magnitude, existing computational limitations have restricted simulation scales, warranting further work in this area.
3. Due to inherent limitations of solar energy (low energy flux density, inability to operate continuously throughout the day), practical implementation for coal mine gas enrichment requires large gas storage tanks, increasing

construction costs and land occupation while complicating system layout. Consider integrating this system with thermal storage devices to enable round-the-clock operation.

4. Since the concentration ratio of single-stage pipes is limited by temperature difference, solar collectors could be used to further increase temperature differences, achieving greater concentration ratios and better enrichment effects with the same pipe length.

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