



Gas hydrate technology: A sustainable approach for comprehensive processing of coal mine water and coal mine methane

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ABSTRACT

For countries whose economies and industries are heavily dependent on the mining sector (e.g., the USA, PRC, Ukraine), the environmental issues associated with coal mining are particularly acute and pertinent. This study focuses on a sustainable approach for the integrated utilization of two of the most widespread types of coal mining waste: coal mine drainage and mine gas.

1. First aspect – mine drainage

Underground mining operations generate considerable volumes of mine, pit, and drainage water, collectively referred to as mine water (MW). In Ukraine alone, mining enterprises collectively produce approximately one billion cubic meters of MW annually, consuming ~0.4 billion m³ of drinking and technical water (Hasii and Gasii, 2024). Given the average volume of 3–4 t of pumped water per 1 t of mined coal, processing these highly mineralized streams represents a significant challenge in achieving the SDGs.

The deterioration of the mining and geological conditions of coal seams increases the volume of MW and its mineralization. In view of the mounting scarcity of potable water worldwide, the problem of processing MW into technical water is of paramount importance. Regrettably, MW are still employed only to a limited extent in the water supply of mining industries, with the majority being discharged into water bodies. Furthermore, MW processing can yield valuable chemical products, such as edible salt (NaCl), sodium sulphate (Na₂SO₄) and various calcium and magnesium compounds.

Known methods of MW purification (distillation, osmosis, electro-dialysis, etc.) either require considerable energy expenditure or have limitations regarding the degree of mineralization of desalinated water (Dastgheib et al., 2016). Despite advancements in desalination techniques, they remain inadequate for addressing this challenge on an

industrial scale. For example, evaporation desalination has a degree of thermodynamic perfection (exergy efficiency) of $\eta_E = 1\%$, electro-dialysis has $\eta_E = 3\%$, and reverse osmosis has $\eta_E = 10\%$. However, the issue of brine disposal arises. Gas hydrate desalination exhibits greater efficiency, with an exergy efficiency of 12–26% (Liang et al., 2024). N-butane, propane, and chlorofluorocarbons (CFCs) have been used as hydrate-forming gases. Nevertheless, owing to the limited availability and high toxicity of these gases, the use of mine gases is a promising avenue for investigation.

2. Second aspect – mine gas

The current state of underground coal deposit development is characterized by increasing depth and growing complexity of geological conditions. In particular, the gas content of mine workings increases, reaching 25–30 m³ per 1 t of coal (Zhang et al., 2022). The emission of methane into the mine environment can impede the rate of coal production, increase production costs, and seriously compromise miner safety.

The concentration of methane in the extracted gas-air mixture varied considerably depending on the degassing method employed. During downhole degassing, the methane-air mixture exhibited considerable fluctuations (from 0.5% to 80%). Currently, many countries employ only a limited portion of the extracted mine gas, with the remainder

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being emitted into the atmosphere, resulting in significant air pollution and accelerating the greenhouse effect.

Depending on the methane content, mine gas can be expediently utilized in the following processes (Liang et al., 2021):

- methanol synthesis (85–98 % CH₄);
- motor fuel production (CH₄ ≥ 95 %);
- combustion in boilers (CH₄ ≥ 30 %);
- combustion in steam generators (CH₄ ≥ 9 %);
- in internal combustion engines (CH₄ ≥ 6 %);
- combustion in furnaces (methane-air mixture with 2 % CH₄).

The reorientation of degassing methodologies towards the facile utilization of mine gas would enhance the operational efficiency and economic viability of coal deposit exploitation while substantially ameliorating environmental impacts. Consequently, it appears most judicious to develop technologies for mine gas utilization that are not strictly contingent on fluctuations in the flow rate and concentration of CH₄ in the gaseous mixture. From this perspective, the use of the gas hydrate concentration technique is particularly promising.

3. Gas hydrates – a sustainable solution

A principal characteristic of gas hydrate technologies is their inherent multifunctionality, which refers to their capacity to yield a broad spectrum of products within a single framework. One unit of methane hydrate contains approximately 170 units of gas, whereas the storage of methane in hydrate form requires 5–8 times lower pressure than that in the free state (2–3 MPa versus 16 MPa, respectively) (Yamamoto and Nagakubo, 2021). The substantial thermal energy associated with the phase transitions of hydrates (> 500 kJ/kg) can be effectively utilized in thermal, cryogenic, and mass storage systems (Toghyani et al., 2024).

When conducting hydrate formation on mine gas using MW, it is necessary to consider the degree of mineralization, as it requires higher pressures and lower temperatures than when using tap water. Hydrates are composed of hydrate-forming gases (e.g., methane, ethane, propane and CO₂) and water molecules. Dissolved salts and dispersed impurities remain in solution because their molecules are too large to fit into the cavities formed by the water molecules. This effect serves as a basis for the practical implementation of gas hydrate demineralization of MW (Jalihal and Venkatesan, 2019). This process facilitates the separation of impurities, thereby enabling methane purification.

Pressure and temperature parameters determine the overall efficiency of the technology (speed, completeness and kinetics of the process), which affects the cost of desalinated and electricity produced from methane. The efficiency of the process depends largely on the initial water mineralization. Studies on the formation of mine gas hydrates with methane concentrations of 13–40 vol% confirmed that the most appropriate temperature for hydrate formation is the zone from 0 to 4°C. In that range the hydrate formation pressure increase (ΔP) for each 1°C increase is minimal. For example, in the range from 1 to 4°C, $\Delta P = 1$ MPa, and in the temperature range above 4°C, $\Delta P = 3$ MPa or more. This is probably due to the different physical state of water at different temperatures.

It should be noted that the reduction of methane concentration in mine gas up to 14 % during the isothermal process of hydrate formation leads to the appearance of loose film structures of hydrate crystals, which quickly decompose after pressure reduction below the equilibrium. Such hydrate structures are of little use in technologies that require hydrate accumulation and storage but are very effective for desalination, where rapid decomposition is required.

Increasing the mineralization of MW causes the equilibrium temperature of hydrate formation to shift to lower temperatures. In the study of MW with mineralization from 1 to 10 g/L, we found that an increase in mineralization of each 1 g/L leads to a decrease in the

equilibrium temperature of ~ 0.2 °C.

The incorporation of constituents from the methane-air mixture and MW as gas hydrate precursors will allow for the simultaneous conditioning of mine gas and desalination of MW, ultimately yielding purified methane, freshwater, and other valuable chemicals (Fig. 1). The first two products can also act as precursors for the production of green fuels, such as hydrogen, through processes such as methane pyrolysis or water electrolysis (Supplementary Information).

The desalinated water recovery coefficient increases with increasing pressure in the reactor. For MW with different mineralization the dependence of fresh water extraction coefficient on pressure is different. Therefore, for MW with a mineralization of 1.7 g/L, when the pressure in the reactor was increased from 10 to 14 MPa, the desalinated water extraction coefficient increased by ~ 0.2 , and for MW with a mineralization of 10 g/L, when the pressure of the hydrate formation process was increased from 11 to 15 MPa, it increased by 0.35. The higher the mineralization of inflow water, the higher hydrate formation pressure is required for the desalinated water obtaining at constant recovery factor. Studies also proved that each value of mineralization of MW corresponds to a limiting pressure of the hydrate formation, which allows the obtaining of desalinated water with a given mineralization, without washing hydrate crystals with fresh water from salt.

Thus, to obtain the required quantity and quality of fresh water, it is sufficient to pass MW through a multi-stage gas hydrate process. To obtain desalinated water with salinity less than 1 g/L, a unit with a small (2–6) number of stages is required.

The cost of 1 m³ of desalinated water depends mainly on the cost of raw materials. Because the raw materials (mine gas and MW) are associated, their cost is determined only by the cost of energy expenditure required to carry out the gas hydrate treatment. According to our calculations, to obtain 1 m³ of desalinated water under the proposed technology it is necessary to spend 20–25 kWh of electricity. That can be partly covered by the cogeneration plant working on mine methane, directly on mine site. For each individual country the electricity tariff is different, so the cost of one cubic meter of treated MW will vary.

Consequently, gas hydrate technology for the integrated utilization of MW and methane has the potential to promote cost-effective advancement of coal deposit resources while markedly diminishing the harmful impact of mining operations on the environment.

4. Conclusions and outlook

The issue of MW recycling has not yet been adequately addressed. As long as conventional approaches do not constitute a sustainable strategy, the treatment of MW via the gas hydrate method, utilizing mine gas as a hydrate generator, necessitates extensive investigation to substantiate the economic viability of this initiative. This challenge was effectively addressed in the early 1990s across multiple coal mining operations in Eastern Ukraine. The trials demonstrated the feasibility of generating mine gas hydrates at a semi-industrial facility, thereby prompting the development of fundamental principles for gas hydrate technology. The evaluation of the pilot facility confirmed the practical possibility of the hydrate formation process within the technologically viable parameters of pressure and temperature.

Consequently, it is posited that gas hydrate technology will yield an additional source of freshwater and mitigate contamination of water bodies. Further dissemination of the proposed strategy is feasible in both active coal mining operations and regions where mining activities have ceased. Stakeholders may encompass coal companies, while the potential benefits extend to other sectors within the mining industry. The practical application of the outlined technological process in any coal-mining region globally will not only facilitate the effective management of mining waste but also generate additional revenue through the commercialization of valuable products (desalinated water, purified methane, dry salts, etc.).

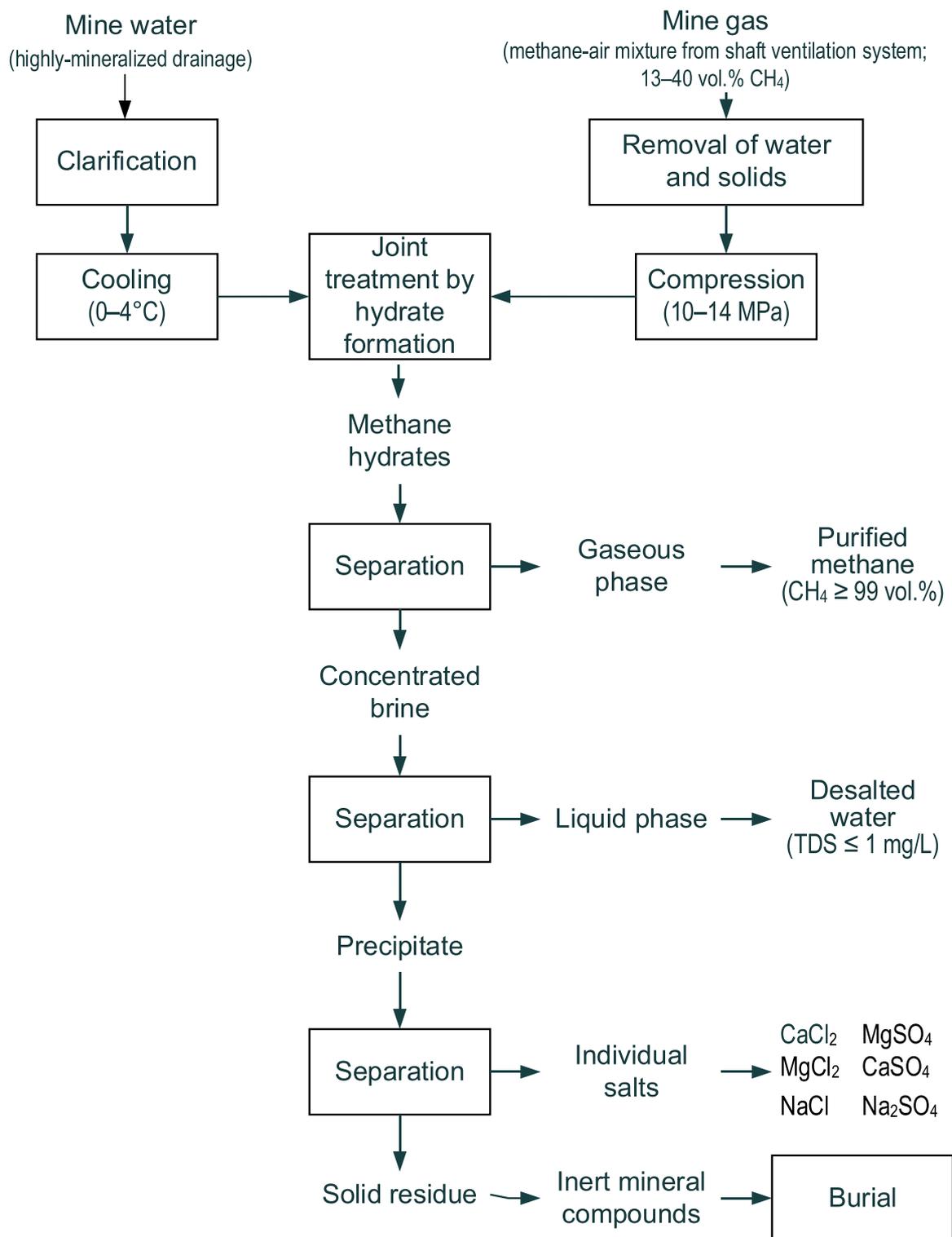


Fig. 1. Flowsheet of integrated technology for mine water and gas utilization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the

online version at [doi:10.1016/j.clwas.2025.100267](https://doi.org/10.1016/j.clwas.2025.100267).

Data availability

Data will be made available on request.

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