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Addressing the CO₂ emissions of the world's largest coal producer and consumer: lessons from the Haishiwan Coalfield, China

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Abstract

China is now the world's largest user of coal, and also has the highest greenhouse gas emissions associated with the mining and use of coal. In the mining sector, the interests of workforce safety coincide with those of greenhouse gas (GHG) management. While the traditional approach to ensuring workforce safety in coal mines was simply to vent the hazardous gases to the atmosphere, thus increasing GHG emissions, recent innovations have seen elements of carbon capture and storage (CCS) being used to simultaneously ensure workforce safety and minimization of GHG emissions. The Haishiwan Coalfield represents a particularly challenging environment for applying this approach, as the coal-bearing strata host both oil shales and a naturally-occurring CO₂ reservoir, disturbance of which could both imperil workers and lead to elevated GHG emissions. A low-carbon, CCS-based model of gas management developed in the Haishiwan Coalfield offers attractive lessons for application to other coal mines, within and beyond China. This approach achieves multiple benefits: energy production, enhanced workforce safety and minimization of GHG emissions. Given the extreme nature of the Haishiwan case, it ought to be even easier to implement these approaches elsewhere.

Keywords: CCS model; Haishiwan coalfield; oil shale; gas hydrate

1. The challenge of greenhouse gas emissions in China

1.1. Introduction

Emissions of greenhouse gases (GHG) are widely accepted to be the principal agent of anthropogenic climate change[1]. The atmospheric concentrations of the principal greenhouse gases carbon dioxide (CO₂) and methane (CH₄) were 391 ppm and 1803 ppb respectively in 2012, exceeding pre-industrial levels by about 40% and 150% respectively[2]. Coal use is responsible for about 40% of global electricity generation as well as 40% of greenhouse gas emissions. It is present in some seventy nations, with the United States, Russia, and China possessing the largest reserves. Coal emits far more CO₂ per unit of energy produced than other fossil fuels: about 30% higher than that of crude oil, and about 70% more than natural gas. Without the implementation of pollution controls, increased coal usage will inevitably result in serious environmental impacts, both in terms of global climate change and through release of other contaminants that have more localized impacts.[3] The greatest potential for reducing the GHG emissions from coal is carbon capture and storage (CCS), which offers CO₂ emissions reductions of 80-90% per unit of energy produced[4]. CCS is being considered as one element of a wider strategy for stabilizing atmospheric CO₂ concentrations. This plan requires that billions of tonnes of CO₂ must be captured worldwide each year, and concentrated and

stored to prevent it entering the atmosphere for hundreds to thousands of years.[5]

1.2. The case of China

Since China opened its doors to the world in 1978, it has not only been the world's fastest-growing large economy, but also an outstanding exporter and a large recipient of foreign direct investment (FDI). China has now become the "workshop of the world", with demand for its good from all around the world spurring its burgeoning manufacturing sector: China's primary, secondary and tertiary manufacturing industries accounted for 10.1%, 46.8% and 43.1% of its GDP in 2010,[6] and manufacturing depends on an adequate supply of affordable energy. Market pressures mean that energy must be cheap and account for only a small proportion of the total cost of manufacturing. This has translated into a huge demand for coal. China has become the world's largest energy producer and consumer. As a primary energy source, in 2012, coal accounts for 68% and 52% of Chinese production and consumption respectively. In 2013, China was consuming 52% of all coal used worldwide, the vast majority of which was produced within China. According to predictions to 2040, coal will continue to dominate energy production and consumption in China for the foreseeable future[7, 8].

According to International Energy Agency (IEA) estimates, China's CO₂ emissions in 2011 were 7.95 gigatonnes (Gt)[9]. Coal-related CO₂ emissions are not restricted to final combustion: the process of mining and beneficiation of coal is also CO₂ intensive. In terms of CO₂ emissions per tonne of coal delivered to power stations, China emitted four times as much as the USA, and twice as much as the UK[10]. Fugitive emissions of methane from coal mines further exacerbate the GHG impacts of coal mining *per se*. Global methane emissions from coal mines were estimated to account for approximately 8% of worldwide anthropogenic methane emissions in 2010, and these emissions were projected to rise by 15% over the following 10 years[11]. China's estimated methane emissions from coal mines that same year were more than 295 MtCO₂e, greatly exceeding those of the second-greatest emitter, the USA, which released slightly over 68 MtCO₂e. Controlling CO₂ emissions without hindering economic development is a major challenge not only for China, but for the many economies worldwide that rely on its goods[12]. As part of a wider initiative to rein-in carbon emissions in the coal-to-energy chain, China clearly needs to reduce the direct and indirect GHG emissions from the process of mining coal. This paper explores how this is currently being approached, using a particularly challenging example to illustrate the feasibility of the approach.

2. The interdependency of gas emissions and mine safety

2.1 The health and safety imperative in Chinese mining

In the early 21st Century, Chinese coal production increased significantly, from 1299 million tonnes (Mt) in 2000 to 3050 Mt in 2009, an annual growth rate of 10%[13]. Unlike many other countries, where opencast mining now predominates, underground mining accounts for 95%, of Chinese coal production. Although the death rate per million tonnes of coal produced has been decreasing steadily, the coal industry remains the most dangerous sector, recording more than 2000 deaths every year[14]. The depth of coal mining is typically 600-800m with an average rate of face advance of 20 m per year. With increasing burial depth of the coal seams, gas pressures and concentrations also increase, which leads to ever greater risks of coal and gas 'outbursts' as mines deepen further. An outburst is defined as a violent, simultaneous release of gases and comminuted rock material into a working face or the interior of a shaft. Outbursts in coal mines represent considerable hazards. Apart from impact injuries, the most immediate hazard, and certainly the most perilous, is the unexpected inundation of the ventilation systems with asphyxiating volumes of gas. When methane is the released gas, an explosive hazard can be created, possibly exacerbated by ejected coal dust[15]. To date, coal and / or gas outbursts have been recorded in more than 1040 mines, scattered across most of the mining provinces in China. Out of 2433 Chinese coal mine deaths in 2010, 623 (25.6%) were due to gas-related accidents[16].

Outbursts are only one manifestation of the dangers posed by coal mine methane (CMM), which is a general term for all methane released during and after mining operations. CMM may have formed biogenically or thermogenically, though most methane in deep strata turns to be thermogenic in origin. Methane is primarily stored in coal by adsorption onto the coal surface; thus it is pore surface area that determines the maximum gas holding potential of a coal seam[17]. CMM has long been considered a danger in underground coal mining due to its explosion risk, which poses a serious threat to worker safety and thus productivity. One of the most important duties of ventilation in underground coal mines is to keep methane levels well below the explosive limit by diluting methane emissions that occur during mining. Methane entering a mine can create a localized zone of high concentration in an area of low air velocities and quantities. The concentration of methane in these zones may pass through a range between 5% and 15%, known as the explosive range. In this range, methane can be ignited easily by any ignition source to create a violent methane explosion that may propagate beyond the

methane-affected zone by explosion of suspensions of combustible coal dust. In addition to proper ventilation practices, removal of coal mine methane from the mining environments prior to, during, and after coal production by using various in-seam and surface-to-mine borehole designs, has been the key component to alleviate the outburst and explosion threat in mining operations[18]. Pre-drainage of methane is preferable from a GHG emissions reduction perspective, as the methane is obtained in more concentrated form, which is more amenable to separation, use and storage of the CO₂ derived from its use.

Some coal seams contain little methane (e.g. in certain areas of the Illawarra coal measures in New South Wales, Australia), with the predominant coal seam gas being carbon dioxide[19]. Where a mine atmosphere becomes saturated with CO₂, so that the proportion of oxygen drops below the respirable limit, this also poses a mortal hazard to miners[20]. Thus the management of gases for worker safety in coal mines addresses precisely the same gases which are of concern in the context of GHG emissions. Historically, CMM and mine-derived CO₂ have simply been diluted with air and vented to the atmosphere; clearly this approach needs to be amended if a reduction in GHG emissions from mines is to be achieved.

2.2. Coal use and options for minimising CO₂ emissions

Coal fuels almost 40% of the world's electricity and in many countries this figure is much higher: Australia, China, India and South Africa, for example, use their large indigenous supplies of coal to generate most of their electricity. For China, particularly given its large indigenous reserves, continuing use of coal for power generation seems inevitable in coming decades, with coal-fired generation still expected to amount to 70% of total electricity supply in 2030[21]. This is daunting, as increased coal use has dominated the growth in GHG emissions for energy production: between 1990 and 2011, growth in GHG emissions from power generation was overwhelmingly due to coal use. Beyond the power sector, there are other significant uses of coal in the iron and steel industry and in the cement sector. These industries also need to address coal-related GHG emissions whilst keeping costs under control. It seems clear that any further coal usage must be accompanied by carbon capture and storage (CCS).

The greatest potential is offered by CCS which can reduce CO₂ emissions to the atmosphere by 80-90%. CCS technologies enable emissions of CO₂ to be stripped out of the exhaust stream from coal combustion or gasification and stored in geological

formations so that they do not enter the atmosphere. There are three main CO₂ capture processes under development for power generation: pre-combustion capture systems (applicable where coal is first gasified before final combustion of the resultant synthesis gas); post-combustion capture systems; and oxyfuel combustion capture systems. Geological features generally being considered for CO₂ storage fall into three categories: deep saline formations, depleted oil and gas fields and unmineable coal seams[22]. To these may be added deep voids produced by coal mining or by underground coal gasification[23, 24]. There are various methods for capturing the naturally occurring gas in coal seams to prevent it entering mine airways, most notably the use of pre-drainage boreholes to remove gas before the mine workings reach the strata, as well as post-drainage techniques for coal seams already disturbed by the extraction process.

3. Case study: Haishiwan coalfield

3.1 Introduction

We have chosen a particularly challenging example where there is a natural CO₂ reservoir associated with the coal, so that mining can unleash CO₂ not even related to coal use. In this paper, we use the Haishiwan coalfield in western China as an example of a closed-circuit resource utilisation model for coalbed gas drainage, drained gas separation and storage, and processing of associated oil-shales (Fig.1). The goal is to develop a low-carbon model for simultaneous satisfaction of energy production, coal mine safety assurance and environmental protection imperatives.

[FIGURE 1 PLACEHOLDER]

[FIGURE 1 PLACEHOLDER]

The No. 2 seam of Haishiwan is a high-volatile bituminous coal of high calorific value, with vitrinite reflectance values ranging from approximately 1 to 1.5%. The thickness of the seam varies from 10 to 40 m. CO₂ outbursts are a significant hazard in the No. 2 seam. The maximum gas content is 40 m³/t, and the maximum gas pressure is 7.3 MPa. The gas mixture in the No. 2 coal seam comprises CO₂ (>80%) and CH₄. The native permeability of the No. 2 coal seam ranges from 0.0004 to 0.0005 mD. For mining safety purposes, the overlying oil-shale (Fig. 2) is first extracted, as this relieves the overburden pressure (geostress) on the No. 2 seam, resulting in expansion of fracture (cleat) pore apertures in

the coal seam, thus increasing permeability, which makes gas drainage much easier to achieve. During oil-shale mining, the gas mixture containing high CO₂ concentrations is drained by 'cross-measures' boreholes (i.e. boreholes that are drilled across the adjoining non-coal strata). This procedure was but the first step in the evolution of an elaborate series of gas control measures which have placed the Haishiwan Coalfield in the vanguard of GHG emissions reduction in the Chinese (and worldwide) coal sector, as described below.

[FIGURE 2 PLACEHOLDER]

[FIGURE 2 PLACEHOLDER]

3.2 Outline of geology and mining in Haishiwan

The Haishiwan Coalfield is located in the western margin of Minhe in the Gansu province of western China, comprising strata of middle Jurassic age, in which coal seams are interbedded with sandstones, mudstones, conglomerates and oil-bearing sandstones and oil-shales (Fig.3a).

[FIGURE 3 PLACEHOLDER]

[FIGURE 3 PLACEHOLDER]

In the Haishiwan coalfield, small-scale coal mining dates back to the 13th century, and modern large-scale mining has occurred since 1958. The oil-shale and No. 2 coal seam, separated by a distance of 40 m, are the primary mined seams. The generalised stratigraphy of the Haishiwan coalfield is shown in Fig.3b. The oil-shale is derived from a lacustrine transgressive sedimentary system, ranging from emergent to submerged (high stand) conditions, which resulted in lake-swamp, medium-depth and deep water sedimentary facies. Oil-shales in the Minhe basin are in the early and early-to-medium diagenetic stages. The average thickness of the oil-shale is 4 m, and the oil content is 10%. The reflectance (R_o) is generally between 0.4 and 1.0%, and the thermal evolution of oil-shale organic substances has reached the immature to low-maturity stage.

Naturally high CO₂ concentrations have been observed in the No 2 coal seam of the Haishiwan coalfield. Dynamic-thermal metamorphism associated with the F19 Fault, which is actually a ductile-brittle shear zone, is believed to have resulted in the release of CO₂ from basement Proterozoic marble formations (Fig.4)[25]. Thus large volumes of CO₂

have been naturally stored in the coal seams of the Haishiwan coalfield for >15 Ma. The process of CO₂ injection into the coal seam by dynamic metamorphism via faults and replacement of pre-existing CH₄ can be considered as a natural analogue of CO₂-enhanced coalbed methane (ECBM) and CO₂ storage pilot projects [26, 27].

[FIGURE 4 PLACEHOLDER]

[FIGURE 4 PLACEHOLDER]

3.3 Adapting mining and allied activities to optimize safety and CO₂ emission minimization

Draining gas from coal seams using boreholes is not only an effective means of capturing gas but is also an effective method to reduce the gas pressure and thus eliminate outburst hazards in coal seams. In view of the naturally low permeability of coal seams, permeability enhancement is necessary to improve the results of borehole drainage.

3.3.1 Simultaneous extraction of coal and gas

(i) Coalbed gas flow

Coal can be considered a dual-porosity system comprising micropores within the solid coal matrix, and fracture pores (cleat). In the process of coalification, gas is stored on the surfaces of the micropores and cleat, with an adsorptive volume ranging up to dozens of cubic metres per tonne. Diffusion and seepage are the main modes of gas transport within the coal seams. Once the pressure or temperature changes, gas will desorb from the micropore walls and pass through the micropore system in a diffusional process, controlled by concentration gradients in accordance with Fick's Law of diffusion. The diffusive flux subsequently enters the cleat porosity, where it tends to become subject to advection (i.e. Darcian seepage driven by pressure differences) (Fig.5)[28]. The velocity of gas transport is a fundamental control on the extractability of coal seam gas, and is thus commonly determined in laboratory tests.

[FIGURE 5 PLACEHOLDER]

[FIGURE 5 PLACEHOLDER]

Flow is controlled by the pressure difference, length and cross-sectional areas of coal

samples, as shown by the equation:

$$q = \frac{kA}{\mu L} \Delta P \quad (1)$$

where q is the flow rate (m^3/s), ΔP is the pressure difference across the sample (Pa), L is the length of the sample (m), μ is viscosity of the fluid ($\text{Pa}\cdot\text{s}$), and A is the cross-sectional area of the sample (m^2). However, given that fluid viscosity is an independent parameter, the only way to control the flow rate is to alter the permeability of porous media. Previous work has demonstrated that the magnitude of permeability (k) dominates the seepage process and can be used to quantitatively illustrate the connectivity of the fracture system. Because ground stress and gas pressure affect the internal connectivity of porous media, permeability is certain to be related to these two parameters. The coal permeability model that considers the effect of the effective stress and coal matrix deformation (ESMD model) is given as[29]:

$$\frac{k}{k_0} = \underbrace{\left\{ 1 - \frac{\alpha}{\phi_0 K} [(\bar{\sigma} - \bar{\sigma}_0) - (P - P_0)] \right\}}_{\text{Effect of effective stress}} \underbrace{\left\{ \frac{f_m}{\phi_0} \left(\frac{\varepsilon_{\max} P}{P + P_L} - \frac{\varepsilon_{\max} P_0}{P_0 + P_L} \right) \right\}^3}_{\text{Effect of coal matrix deformation}} \quad (2)$$

Where, $\bar{\sigma}$ and $\bar{\sigma}_0$ (both in MPa) are the mean stress and initial mean stress, respectively; K is coal bulk modulus (MPa); α is Biot's coefficient (dimensionless); ϕ_0 is initial coal matrix porosity (dimensionless); ε_{\max} is maximum adsorption strain (dimensionless); P and P_0 are gas pressure and initial gas pressure, respectively (MPa); P_L is Langmuir's pressure (MPa); and f_m is effective coal matrix deformation factor (dimensionless).

The permeability of Chinese coal seams is usually in the magnitude of 10^{-4} - 10^{-3} mD, which is four orders of magnitude lower than typical seams in the U.S. and three orders of magnitude lower than those in Australia[30]. The low permeability makes extraction of the gas from underground much more difficult in China than in these other countries. Therefore, the permeability in coal seams should be improved in order to improve the ease of extraction. Fortunately, many engineering methods have been used to solve this problem, including the use of dense boreholes, hydraulic fracturing, loose blasting and protective coal seam mining. All these measures, especially protective coal seam mining, are effective at reducing ground stress and improving coal seam permeability.

(ii) Stress distribution under the influence of mining

“Protective coal seam mining” involves the prior extraction of one seam to improve the permeability, and thus gas extractability, of another seam in the same sequence of strata. Typically, the seam targeted for prior extraction will have low gas contents with no danger of outbursts. Protective coal seam mining can improve the movement and deformation of coal seams and rock seams, creating an enhanced fracture system. As the stress in the roof and floor of the mining goaf declines and the coal seam swells, the permeability is increased by a factor of hundreds to thousands, greatly aiding the gas extraction process[31].

In the case of the Haishiwan coalfield, the ‘protective horizon’ that needs to be extracted first is actually the oil shale seam. With a thickness of 3 m extracted, the stress in the coal and enclosing rock layers in the mining area decreases markedly. The magnitude of the pressure-relief effect varies with distance from the oil shale seam, as well as with the mechanical strength and the gas pressure distribution. Using numerical modelling, the pressure-relief effect is estimated for a mining length of 200 m in the upper oil shale seam. When the vertical distance from the oil shale seam is 50 m, the roof stress is decreased by 20.71 MPa from the initial value. The stress in the floor of the No. 2 coal seam, which is 90 m away from the oil shale seam, decreases by 15 MPa. At different separation distances, the vertical stress experiences different magnitudes of relief, generally resulting in a symmetrical reverse-wedge shape (Fig.6).

[FIGURE 6 PLACEHOLDER]

[FIGURE 6 PLACEHOLDER]

After the exploitation of the upper protective seam, coal and rocks begin to swell on the floor of the goaf (i.e. the zone of settling where all coal has been removed). The stress-sensitive permeability in the No 2 coal seam initially increases significantly due to the stress relief offered by protective oil shale seam mining. Subsequently, permeability gradually decreases again as the oil shale goaf settles and the stress field recovers[32]. The tensile stress of the rock mass under the oil shale goaf exceeds its tensile strength, leading to an obvious fractured appearance (Fig.7)[33]. Many experiments have demonstrated that there is a positive relationship between fracture occurrence and stress-relief effects[34-36].

[FIGURE 7 PLACEHOLDER]

[FIGURE 7 PLACEHOLDER]

(iii) Field characterisation

In naturally fractured formations such as coal, permeability is highly sensitive to changes in stress or pore pressure. The undisturbed permeability of the No. 2 coal seam ranges from 0.00039 mD to 0.00051 mD, but it increases gradually due to the unloading of vertical stress. The actual maximum measured value is 0.042 mD, which corresponds to 45 days since the mining of the oil shale in the underground coal mine (Fig. 8) and is approximately 100 times that of the pre-development permeability.

[FIGURE 8 PLACEHOLDER]

[FIGURE 8 PLACEHOLDER]

Thus through the use of protective coal seam mining, the permeability is enhanced by as much as two orders of magnitude, and the seepage rate consequently also increases to an acceptable value, supporting gas removal via boreholes drilled through the seam. The boreholes, with 93mm diameters and spaced every 15 to 20m, need to be drilled in the tunnel under the workface through the whole No.2 coal seam so that the gas flow can be easily transported into the extraction system.

[FIGURE 9 PLACEHOLDER]

[FIGURE 9 PLACEHOLDER]

This cross-seam extraction system obtained a total of 40.5 million cubic metres (Mm^3) of gas from December 2008 to June 30th, 2010 (30 Mm^3 of CO_2 and 10.5 Mm^3 of CH_4). The average concentration of extracted gas (methane and CO_2) was 70%. Every day, approximately 69.9 thousand tonnes of gas was extracted (55.6 thousand tonnes of CO_2 and 14.3 thousand tonnes of CH_4). The quantity of flow in a single borehole reached an average 0.8 m^3/min , with a maximum value of 2.0 m^3/min (Fig.9). The high gas concentrations indicate that this is a highly valuable extraction system for use in mines. However, the different gas ingredients make it difficult for it to be used as a fuel or fertiliser. Therefore, we need introduce a gas separation process to expand the applicability of the method.

The gas extraction rate of the protected seam is over 75%. The residual gas pressure is decreased to below 0.5 MPa, and the residual gas content falls below 10 m³/t. These results show that a seam that initially had a high risk of gas outburst has been transformed into a seam with zero outburst risk, thus allowing the safe and efficient mining of the over 3,000 tonnes produced daily in the working coal mine.

Every year, the mine yields 1.2 million tonnes of oil shale, assuring the production of 2 million tonnes of coal resources from the No. 2 seam. In addition, approximately 140 million cubic metres of gas are extracted each year. With the method of stress-relief mining, the outburst danger was reduced and the safety conditions were improved. Additionally, a high concentration of gas was captured, and the greenhouse effect was diminished.

3.3.2 Responsible use of Haishiwan oil shale resources

Since the oil shale has a low heating value (ranging from 10 MJ/kg to 13 MJ/kg), voluminous solid waste and greenhouse gases would be generated during conventional combustion of this material. Fortunately, the oil shale in the Haishiwan Mine has a high oil ratio (>10 %) which can be extracted for further use. This means that the oil shale here is not so much a liability to be avoided, but a resource which can be exploited responsibly, that is to say in a manner most consistent with CCS. Various facets of this exploitability are briefly summarised here.

(i) Compositional analysis

The Haishiwan oil shale has a dark brown, greasy lustre and planar quasi-schistose lamination. Scanning electron microscopy (SEM) results show the bedded nature and pore structure of the oil shale (Fig.10). The average oil content of Haishiwan oil shale is 10%. Tables 1 and 2 give typical analyses. The organic matter in the the oil shale is a kerogen (a complex combination of carbon, hydrogen, sulphur) that cannot be extracted with organic solvents. Oil shale ash is an inorganic byproduct of combustion of oil shale and retorted shale. Since it can cause environmental problems, attempts have been made to put these materials to beneficial use, especially in the construction industry.[37]

[FIGURE 10 PLACEHOLDER]

[FIGURE 10 PLACEHOLDER]

[TABLE 1 PLACEHOLDER]

[TABLE 1 PLACEHOLDER]

[TABLE 2 PLACEHOLDER]

[TABLE 2 PLACEHOLDER]

(ii) Pyrolysis characteristics

Pyrolysis is a complex suite of reactions, involving changes in both physical and chemical properties, by means of which solid organic matter (kerogen) is converted to shale oil, hydrocarbon gases, water vapour and char by steadily heating the raw material to temperatures of 500 – 550°C. The oil shale characteristics, pre-treatment conditions and other case-specific factors have significant impacts on the pyrolysis process. [37]

[FIGURE 11 PLACEHOLDER]

[FIGURE 11 PLACEHOLDER]

Fig.11a shows the yields of oil, gas and water produced from Haishiwan oil shale at pyrolysis temperatures ranging from 350 °C to 550 °C. Oil yield increased with increasing temperature and reached a maximum at 525 °C. When the temperature exceeded 530 °C, the yield of shale oil decreased. The rate of decomposition was highest at temperatures between 400 °C and 525 °C, indicating the degradation of kerogen. The yield of retorting gas always increased in the temperature range from 350 °C to 550 °C, which was due to the desorption of adsorbed gas from the surfaces and micropores of the oil shale samples, as well as to the decarboxylation and decomposition of carbonate and organic compounds. The content of H₂ gradually increased, presumably due to the aromatisation and condensation of organic matter in spent shale.

As seen from Fig.11, the pyrolysis temperature ranged from 350 °C to 550 °C. Figure 11b demonstrates that CH compound release increased in tandem with temperature, due to the cracking of macromolecules or side chains, up to an inflection point at 475 °C. At higher temperatures, the yield of CH compounds began to decrease, while that of hydrogen continued to climb, reflecting progressive condensation and aromatisation reactions. While CO₂ yield initially declined, due to exhaustion of options for decarboxylation reactions, after the peak of CH yield the previous decline in CO₂ yield was arrested somewhat. CO yield changed very little over the observed temperature range. It was thus concluded that maximisation of oil yield from these shales would be favoured by pyrolysis temperatures of 490-520 °C.

(iii) Retort subsystem

Shale oil was produced by destructive distillation using SJ-IV low-temperature carbonisation with internal combustion heat provided by a continuous vertical square furnace. Oil shale particles with diameters of 6-80 mm were used as the raw materials. Approximately 195,000 tonnes of oil shale were processed every year, and the oil recovery rate was 84-88%. Semi-coke and retorting gases are by-products. The semi-coke, which has a high calorific value, is not burned and thus can be used for other purposes. The retorting gas is captured and used as the heating gas in the pyrolysis process. The carbonisation gas does not contain oxygen, so the oil recovery rate can be improved by using an electric capture device.

The overall process is as follows: The raw materials are first transported by belt conveyor and are loaded into the stock bin in the upper part of the reactor. They are then loaded into the retort furnace through the feed port and auxiliary tank. The massive shales loaded into the furnace move downwards, contacting the heating gas (i.e. the hot flue gas generated from combustion of retort gas in air). The heating gas is sent into the furnace and is gradually heated, producing oil gas and semi-coke by pyrolysis. Retorting oil gas is derived from the furnace roof through the riser tube. The roof temperature should be maintained at 80-120°C, and the distillation section temperature should be 550-700 °C. After retorting, the oil shale semi-coke is cooled to approximately 80°C and is then passed through the discharge coke box to the quenching coke tank in the lower part of the furnace. The semi-coke is continuously discharged through the coke-pushing and scraper machines into the buffer position. It is then transported to the coke field by belt conveyor for storage.

The steam generated from raw materials during drying, the shale oil and carbonisation gas produced during the dry distillation, the smoke produced by the carbonisation gas during the burning of the lower portion, and the steam produced from semi-coke are mixed together. The mixtures are collected by the gas-collecting hood at the top of the reactor. The mixtures are then fed into the condensate recovery system through the ascending pipes. The condensate recovery system contains a Venturi tube tower, a swirl board scrubber and an electrostatic oil-trap. The retorting process is shown in Fig.12.

[FIGURE 12 PLACEHOLDER]

[FIGURE 12 PLACEHOLDER]

The retorting of Haishiwan oil shale produces low-sulphur shale oil, with the characteristics shown in Table 3.

[TABLE 3 PLACEHOLDER]

[TABLE 3 PLACEHOLDER]

As previously noted, Haishiwan oil shale retorting produces semi-coke and retorting gases as by-products.. The semi-coke has a high calorific value due to its combustion in air-free conditions, as shown in the analytical data in Table 4. The oil shale fuel mixture, which consists of semi-coke, No.2 coal and fine oil shale (0-6mm), is fed to the circulating fluidised bed boilers to burn in order to generate high-pressure steam. The steam is used to supply heat and generate electricity using a traditional Rankine cycle process.

[TABLE 4 PLACEHOLDER]

[TABLE 4 PLACEHOLDER]

The retorting gas consists mainly of N_2 , H_2 , CH_4 , CO_2 and CO , as shown in Table 5. The heating value of the gas is 5.02 MJ/m^3 . Following chemical absorption-wet oxidation desulfurisation, the retorting gas is burned in a gas engine to generate electricity, and the flue gas waste heat is recovered by a heat recovery boiler, which is used to co-generate heat and electricity.

[TABLE 5 PLACEHOLDER]

[TABLE 5 PLACEHOLDER]

Retort ash from the Haishiwan oil shale consists mainly of SiO_2 , Fe_2O_3 and Al_2O_3 with minor K_2O , Ti_2O , Na_2O , and CaO components, as shown in Table 2. This shale ash can be used in the Portland cement production process, which comprises three unit processes: material preparation (grinding, drying and mixing), clinker production, and clinker crushing.

3.3.3 Gas Hydrate Separation and CO_2 storage

(i). Gas Hydrate Separation

The main components of the gas obtained through the pressure-relief drainage process in the Haishiwan coal mine are CO₂, CH₄, and N₂. Due to the inhomogeneity of gas drainage, the highest CO₂ concentration is above 70%, while the concentration of CH₄ is approximately 10%. As CO₂ and CH₄ are both greenhouse gases, the mixed gas must be separated for effective gas use and underground storage. At present, the internationally available methods for mixed gas separation include pressure swing adsorption, low temperature processing, and separation membranes, all of which have their limitations.[38, 39] Therefore, we propose the method of gas hydrate separation[40, 41]. Gas hydrates are cage-type, crystalline, non-stoichiometric compounds resembling ice. They are generated by water (the 'subject' molecule) and gas (the 'guest' molecule) at certain temperatures and pressures. Gas hydrates have three crystal structures:

type I (8M·46H₂O)

type II (24 M·136H₂O), and

type H (6 M·34H₂O)

where 'M' represents the guest molecule.

The main components of high-CO₂ concentration gas, including CO₂, CH₄ and N₂, can all form hydrates at certain temperatures and pressures. However, the phase equilibrium pressures of CO₂, CH₄ and N₂ hydrates vary greatly at the same temperature (e.g. at 0°C, the phase equilibrium pressures of CO₂, CH₄ and N₂ hydrates are 1.24 MPa, 2.56 MPa and 14.30 MPa, respectively). Therefore, by controlling the build-up pressure, CO₂ easily undergoes a phase change (from gaseous state to solid state) and forms hydrates. Through the process of multilevel gas-solid transformation, CO₂ separation and purification in mixed gas can be realised. The principle of hydrate separation in high-CO₂ concentration gas is shown as Fig.13.

[FIGURE 13 PLACEHOLDER]

[FIGURE 13 PLACEHOLDER]

At present, few experimental data exist for hydrate separation of high-CO₂ concentration gas. We developed our own jacket-cooling-type high-pressure reaction to separate high-CO₂ concentration gas. We use micro-confocal Raman Spectrometry in the visible range to observe the products of gas hydrate separation. The gas samples used in the experiment are 75% CO₂, 11% CH₄ and 14% N₂.

[FIGURE 14 PLACEHOLDER]

[FIGURE 14 PLACEHOLDER]

A CO₂-CH₄-N₂ mixed gas hydrate is compounded in a high-pressure vessel with a 1.5-cm diameter sapphire window of 3 ml volume. We inject deionised water into the vessel and flush it two to three times by gas replacement. We then reduce the temperature of the experimental vessel to 1°C, then hold temperature constant. We subsequently inject gas into the vessel at 5 MPa. After about 6 minutes, white and granular hydrate first appears on the gas-fluid contact surface inside the experimental vessel (Fig.14a). The pressure is then reduced to 4.17 MPa. As the reaction continues, the amount of white hydrate increases and keeps growing upwards through the reaction still and around the sapphire window (which is gradually obscured, so that the visibility of the vessel interior decreases; Fig.14b). The total time for the experiment including the hydrate reaction is 75 min. The hydrate is white and snow-like and eventually fills the entirety of the experimental vessel (Fig.14c). The gaseous phase pressure does not change at all. After the hydrate reaction reaches equilibrium, hydrate production ends.

[FIGURE 15 PLACEHOLDER]

[FIGURE 15 PLACEHOLDER]

In order to determine the types of guest molecules and analyse the microscopic properties of the compound gas hydrate, we use Raman spectroscopy to observe the gas hydrate separation process. Fig.15. shows the Raman spectrogram of gas hydrate separation. Only CO₂ and CH₄ molecules fill the hydrate crystal cage during gas hydrate separation; N₂ molecules are not a part of the hydrate separation process[42]. Based on comparison with known Raman spectra[43, 44], we conclude that the fundamental structure of the experimental hydrate is type I[45].

We determined the elemental composition and crystal form of the gas hydrate through microanalysis. For the initial experimental conditions of 1°C and 5 MPa, only CO₂ and CH₄ participate during the hydrate reaction of the high-CO₂ concentration mixed gas. Because the phase equilibrium conditions for N₂ separation are not yet reached, N₂ molecules are not involved in the hydrate separation reaction. Therefore, by controlling

the experimental conditions of hydrate separation, we can separate CO₂ from the mixed gas.

(ii) CO₂ storage in longwall mining goaf and abandoned coal mines.

Fires from the spontaneous combustion of coal seams significantly impact coal safety production in China; more than 56% of the coal mines in China are in danger of spontaneous combustion. Fires in abandoned mines and waste banks are a relatively common occurrence in coal-producing areas.[46] The injection of inert CO₂ into coal goaf could reduce oxygen concentrations, decreasing the risk of spontaneous combustion in coal seams. Gas adsorption capacity increases with increasing CO₂ content and decreases with increasing CH₄ content (Fig.16). The CO₂ Langmuir volume of the No. 2 coal seam reaches a value of 33.34 m³/t, which is approximately 1.24 times greater than that of CH₄. The relative adsorption capacities of gas on coal vary thus: CO₂>CH₄>CO>N₂. CO₂ adsorbed on coal hinders oxygen adsorption and thus prevents spontaneous combustion. At the same time, the mining goaf includes substantial remains of coal, but has a far greater porosity than unmined seams; hence a large amount of CO₂ could be stored in the goaf aquifer through adsorption in the free phase and dissolved state[23, 47, 48].

There are many abandoned deep coal mines in China, and these potentially offer an interesting option for CO₂ storage. Gas in a coal mine can be stored in three modes: as a free gas, in the voids and mining-induced fractures; adsorbed onto the remaining coal and other dispersed organic matter; and in the dissolved state in the mine water[49, 50]. The theoretical CO₂ storage capacity depends on:

- (a) the volume of voids,
- (b) the remaining reserves of coal and its sorption capacity and
- (c) the pressure, temperature and chemical composition of mine water.

Many factors have to be considered in a simulation of CO₂ sequestration in abandoned coal mines. These include depth, coal reserves, sealing, mine condition, mine water, and existent faults. As the degree of mine sealing, mine condition, mine water circulation and fault hydrology are usually well constrained from mine water studies[51], the most important factors are likely to be depth and total coal reserves. Depth governs the phase state of CO₂: under supercritical conditions, the CO₂ combines the compressibility of a gas

with the density of a liquid, which permits efficient use of underground reservoirs for storage. The critical point where CO₂ enters the supercritical phase is defined as 31.1 °C and 7.38 MPa, can be achieved at a depth of only 756 m under normal hydrostatic pressure. This is within the range of many abandoned deep coal mines in China. ,

To implement such a storage method, the risk assessment of CO₂ storage in abandoned coal mines will be necessary. This requires a wide range of data quantifying the geology, mine geometry, mine water hydrology (including any natural and induced fracture permeability) and the stress state of any faults intersected by the mine. It is also necessary to determine the properties of the rock mass strength characteristics and in particular any data on the gas permeability of coal seams and the enclosing rock mass, plus investigations of any freshwater features (e.g. potable water aquifers) that may have been affected by mining activities. Only a sub-set of all abandoned mines are likely to be suitable for secure CO₂ storage; however, these represent very large reservoirs, with internal permeabilities that can exceed those of natural saline aquifers by as much as three orders of magnitude[51]. This new CO₂ geological storage prospect merits further investigation alongside the current attention being paid to shale gas development without CCS [52-54].

[FIGURE 16 PLACEHOLDER]

[FIGURE 16 PLACEHOLDER]

3.4 Resource recycling in the Haishiwan coal mine

A method of resource recycling has been established for the Haishiwan coal mine. In this method, the coal is safely mined, and the CMM / GHG gases are captured. The oil shale seam mining in the Haishiwan coal mine provides a vivid demonstration of this method. Because of the oil shale mining, a highly-concentrated CO₂ source is drained and collected, and the risk of outburst in the No.2 coal seam is eliminated. Based on the characteristics of the available oil shale, a new comprehensive utilisation system was analysed for shale oil production, electricity generation, oil shale ash processing, economical efficiency and environmental protection. The system involves three subsystems: the retort subsystem, where coarse oil shale (6–80 mm) is retorted to produce shale oil, hydrocarbon gases and semi-coke; the combustion subsystem, where

the fuel mixture of oil-shale, semi-coke and coal are burnt in a circulating fluidised bed (CFB) furnace to generate high-pressure steam, which is used to supply heat and generate electricity; and the ash processing subsystem, where ash from the CFB furnace is utilised to produce cement. The hydrate method achieves CO₂ separation from the mixture drainage gas, which is easy to store and transport on a large scale. CO₂ can be injected into the coal mine goaf to prevent spontaneous combustion of coal and sequester CO₂ (Fig.17). The by-product and greenhouse gases reduce with resource recycling in the Haishiwan coal mine every year as show Tab.6. The solutions of coalbed gases capture and resource recycling eliminate gas problem and casualties, which promote mining safety and reduce greenhouse emissions. Coupling coal extraction/preparation with power generation facilities would improve the economics of “zero-emission” power plants due to the proximity of all the value-chain facilities.[55]

[FIGURE 17 PLACEHOLDER]

[FIGURE 17 PLACEHOLDER]

[TABLE 6 PLACEHOLDER]

[TABLE 6 PLACEHOLDER]

4. Implications for CO₂ management worldwide

4.1 Implication for other Chinese coalfields

There are 43 mining areas in China, and some 200 coal mines reach depths of 800m or more; 47 coal mines are deeper than 1000m[56]. As coal mining depths have increased, accompanying increases in geostress and coal seam gas pressure and content have been observed. However, the permeability of coal seams appears to decrease with depth, which has contributed to a growing number of coal and gas outburst and gas explosion disasters. Although the processing of coal is an ancient problem and has been practiced for centuries, the constraints posed on today’s coal conversion processes are unprecedented, and utmost innovations are required for finding solutions to the problem[57]. Coal mine methane (CMM) is considered first and foremost as a safety issue, as it has resulted in numerous accidents in China’s mining history. Yet CMM is also a GHG and a valuable fossil fuel. Recovery of CMM by borehole drainage prior to coal mining reduces atmospheric emissions and increases safety. The process is greatly enhanced by increasing coal bed permeability. The selection of a suitable CMM gas

drainage method in China is mainly determined by factors including the source of methane, the type of coal, the coal extraction method and the geological conditions. In this manner, high-concentration gas is captured, with a methane content of 30% to 80%. The main use for medium-concentration CMM is power generation, which decreases GHG emissions due to the much lower global warming potential of CO₂ compared with methane. To reduce these CO₂ emissions, CO₂ capture systems (e.g. using the hydrate process described above) can be coupled with high-efficiency combustion and advanced power generation technologies.

CO₂ could be stored by adsorption on residual coal and organic matter, by accumulation in porosity in goaf areas and damage zones of abandoned coal mines and the surrounding bed rock, with both coal mine safety and GHG storage benefits. The injection of inert CO₂ into coal goaf could reduce oxygen concentrations, decreasing the risk of spontaneous combustion in coal seams. The goaf areas and abandoned coal mines represent a significant void volume for CO₂ storage in China. It is not realistic to propose a single, generic model combining safe coal mining, clean coal use and CCS applicable to all coal mines. Site-specific, comprehensive recycling models for energy, safety mining and GHG reduce should be decided upon according to the nature of each coal mine. However, the Haishiwan example provides clear indicators of the kinds of possibilities that can be identified.

4.2 Implication for other major coal-producing countries

The largest coal producing countries are not confined to one region of the world: the top five producers are China, the USA, India, Australia and South Africa. Much coal tends to be used in the country in which it is mined. The other coal producing countries should take actions to reduce CO₂ by a range of methods outlined above, including:

- 1) Gas drainage mainly using surface and underground pre-drainage before conventional mining, which could capture high-concentration coalbed methane and carbon dioxide to enhance worker safety, support power generation and facilitate CCS.
- 2) Underground coal gasification combined with CCS, which offers an extra degree of freedom or an added level of flexibility in reducing the overall carbon footprint of a cluster of carbon-intensive industries[23].
- 3) Comprehensive recycling models for energy, safety mining and GHG reduce should be decided upon according to the nature of each coal mine. Plans for resource

circulation and sustainable development in all the coal mines should be established[58].

5. Conclusions

China is the world's largest coal producer and consumer, and with the biggest CO₂ emissions arising from coal mining and use. The Haishiwan coalfield set an example for a CCS low-carbon model for coal mining and coal use under Chinese conditions. The high CO₂ content of the coal seam has posed a threat to miners' safety, as well as a potent source of GHG emissions. Depressurisation of the coal seam by prior extraction of the overlying oil shale reduces in-situ stress, which increases the permeability with about 100 times of the coal seam in Haishiwan coal mine. Thus, highly concentrated CO₂ gas mixtures can be captured by boreholes and transport systems, which are based on permeability and Darcian gas flow processes. The quantity of flow in a single borehole reached an average 0.8 m³/min, and the average concentration of extracted gas (methane and CO₂) was 70%.

In the Haishiwan case, depressurisation was achieved by prior mining of overlying oil shale, which through retorting yields shale oil, retorting gas and semi-coke, supporting power production using a gas engine and circulating fluidised bed combustion with CO₂ capture. The quality of coal, shale oil, cement, electricity could reach to 2.0 Mt, 0.1Mt, 0.6Mt, 170 million kWh every year respectively, and the annual greenhouses reduction of methane and carbon dioxide is 6650 tons, 70110 tons in the Haishiwan coal mine, respectively. Furthermore, the solid combustion residue can be applied to cement manufacturing and backfilled into coal mine goaf.

In other mine settings, the same depressurisation could be achieved by sequential down-sequence mining of subjacent coal seams, or by means of staged underground coal gasification.

There are many CO₂ separation methods: this paper presents a gas hydrate separation method for CO₂ capture, which is convenient for storage and mass transit. Our experimental results indicate that CO₂ and CH₄ could be separated and purified under near-ambient temperatures and pressures. CO₂ injection into coal mine goaf or abandoned coal mine voids allows CO₂ to be stored in the free phase, adsorbed on the remaining coal or dissolved in mine water. The amount of storage could be significant depending on geological characteristics of mines and engineering design parameters. Further research to quantify such sequestration capacity, and to assess the attendant risks, is underway in China. Similar approaches have global applicability.

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Figure captions:

Fig.1 Summary of the integrated system of underground coal and oil shale mining, surface resources utilization within which carbon capture and storage strategies are being developed for the Haishiwan Coalfield, China.

Fig.2. The mined stratal sequence in the Haishiwan Coalfield, western China, showing the No 2 coal seam, and the overlying oil-shale seam, which is extracted before the coal is worked to facilitate gas drainage from the latter. The strata above the oil-shale are sandstones; between the oil shale and the No 2 coal seam, the strata are carbonaceous clastics, while underlying the coal seam are siltstone, conglomerate and silty fine-grained sandstone. as well as the.

Fig.3. (a) Generalized stratigraphy features of the Haishiwan coalfield. (b) Seam floor depth contours for the No. 2 coal seam in the Haishiwan coalfield, showing locations of major geological faults, exploration boreholes, and selected mined panels of coal discussed in the text.

Fig.4. Formation of the CO₂ gas pool in the No 2 coal seam by migration of CO₂ from deeply-buried marble via the F19 fault zone. Note that this fault zone consist of a central core and a damage zones: the fault core itself is of low permeability; the damage zone is the primary route of CO₂ migration.

Fig.5. Schematic summary of gas migration in coal seams across the range of scales

Fig.6. Distribution of stress in the roof and floor of the No.2 coal seam due to mining of the shown panel in the overlying oil shale seam 'protective layer'.

Fig.7. CT reconstruction of damage and fractures development during coal sample unloading stress

Fig. 8. (a) Evolution of the permeability of the No. 2 coal seam with stress relief. The results are calculated for the borehole datums. (b) The relationship between the flow rate and pressure of the relief boreholes (drilled up from the floor of No.2 coal seam) in the lower protected layer and the progress of longwall mining in the overlying 'protective layer' (i.e. the oil shale horizon).

Fig. 9. Evolution of gas drainage in the floor tunnel over time, in response to stress-relief induced by prior extraction of the overlying oil shale 'protective layer'. (a) Methane; (b) carbon dioxide.

Fig.10 . Photograph and SEM image of Haishiwan oil shale. (a) Oil shale develops flat bedding and has a heavy asphaltic odour. (b) Macropores and mesopores within an intimate mixture of amorphous kerogen (organic matter) and platy clay minerals.

Fig. 11 (a) Yields of pyrolysates at different pyrolysis temperatures. (b) The composition of retorting gas at different pyrolysis temperatures.

Fig.12. Schematic diagram of the comprehensive utilization system for oil-shale at Haishiwan. .

Fig.13. Hydrate separation principle for gas with high CO₂ concentrations.

Fig.14. Representative pictures of the hydrate growth process

Fig. 15. Raman spectrogram of hydrate separation production in the gas sample.

Fig.16. Low-pressure binary gas (CO₂ and CH₄) adsorption on the dry Haishiwan coal at 30 °C

Fig.17. The low-carbon model of safe coal mining, resource cycling and environmental protection developed for the Haishiwan coal mine.

Table 1. Proximate and ultimate analyses of Haishiwan oil shale (on an air-dried base)

Proximate analysis (wt%)		Ultimate analysis (%)	
Moisture	1.31	C	79.38
Volatile matter	58.84	H	7.81
Ash	54.30	O	8.76
Fixed carbon	18.76	N	1.78
TOC	27.8	S	1.17

Table 2. Ash analysis of Haishiwan oil shale, wt%

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃	K ₂ O	Na ₂ O	TiO ₂
64.5	19.46	8.73	1.26	1.48	0.14	1.36	0.55	1.37

Table 3. Basic properties of Haishiwan shale oil

Analysis item		Results
Calorific value (MJ/tonne)		49.26
Moisture (%)		0.76
Ash (%)		0.0077
Density (kg/m ³)		913.60
Kinematic viscosity(50°C) (mm ² /s)		10.51
Sulphur content (°C)		0.52
Carbon Residue (%)		1.20
Flash point (°C)		115.00
Pour Point (°C)		27.00
Solidifying point (°C)		24.00
Mechanical impurity (%)		0.04
Four components	saturated hydrocarbon (%)	47.94
	Aromatic hydrocarbon	22.36
	Resin (%)	26.38
	Asphaltene (%)	3.33
Distillation range	Initial Boiling Point(°C)	149.60
	Gasoline fraction (%)	2.50
	Diesel fraction (%)	48.10
	Heavy oil fraction (%)	49.40

Table 4. The property of Haishiwan semi-coke derived from oil shale

Sample	Moisture (%)	Ash (%)	Volatile matter (%)	Fixed carbon (%)	Calorific value kJ/kg
Semicoke	3.9	84.79	4.1	7.21	4274

Table 5. The bulk composition of retorting gas produced at Haishiwan (%)

Composition	Volume fraction	Composition	Volume fraction
N ₂	56.99	C ₂ H ₂	0.45
CO	11.56	C ₃ H ₈	0.01
CO ₂	15.20	C ₂ H ₄	0.05
CH ₄	2.34	C ₄ H ₈	0.22
H ₂	12.78	n-butane(C ₄ H ₁₀)	0.11
C ₂ H ₆	0.13	i-butane(C ₄ H ₁₀)	0.05

Table 6. The products and greenhouses gases reduce during resource recycling every year in the Haishiwan coal mine.

Coal	Shale oil	Cement	Electricity	CH ₄ extraction	CO ₂ emission reduction
2.0 million tons	0.1 million tons	0.6 million tons	170 million kWh	6650 tons	70110 tons

Density of methane and carbon dioxide is taken as 0.716 kg/m³, 1.964 kg/m³, respectively, a density in standard condition.