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Carbon dioxide utilisation for carbamide production by application of the coupled UCG-Urea process

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Abstract

World-wide coal reserves can supply the global demand for primary energy for several centuries. However, low thickness and structural complexity may constrain the economic exploitation of many coal deposits. Taking into account these circumstances, underground coal gasification (UCG) can offer an economical and sustainable approach for coal exploitation and subsequent feedstock generation from the syngas. The UCG process produces a high-calorific synthesis gas mainly consisting of methane, hydrogen and carbon dioxide, which can be used for electricity generation or feedstock production at the surface. Considering the latter, the Urea process can be applied to establish the nitrogen based fertilizer carbamide (CH4N2O). The required feedstock for carbamide production in the Urea process can be supplied by UCG syngas. The aim of the present study was the development of an integrated carbon utilisation concept based on the coupled UCG-Urea process. A significant amount of carbon dioxide from the UCG synthesis gas is required for carbamide production in the Urea process, while the excessive carbon dioxide can be re-injected into the cavities resulting in the coal seams and surrounding strata after the gasification process. Thus, a new approach for utilisation of carbon dioxide resulting from coal combustion was developed to provide a coupled technology also comprising geological storage of excessive carbon dioxide. A theoretical feasibility study considering UCG-Urea process economics and potentials of UCG and carbon dioxide storage in the gasified strata was

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conducted for a selected study area in northern Bangladesh revealing the high competitiveness of the combined technology on the international feedstock markets.

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1. Introduction

Global coal reserves can provide primary energy supply for several centuries. Conservative calculations considering the available coal resources in Europe reveal different feasible scenarios of coal based energy supply. Nevertheless, low thickness and structural complexity of coal deposits may constrain their economic exploitation. In addition, relatively high greenhouse gas emissions of conventional coal based energy generation are causing reservations against this method of coal utilisation. Considering these circumstances, underground coal gasification (UCG) can offer an economical and sustainable approach for coal utilisation to generate feedstock. The UCG process is based on the development of coal deposits using directional in-seam drillings maintained at the coal seam base to establish a connection between injection and production wells in low-permeable seams. Subsequently, the coal seam is ignited by the injection of an oxidant (in general a mixture of oxygen and water vapour) which triggers and controls an in situ sub-stoichiometric combustion process. This gasification process produces a high-calorific synthesis gas mainly consisting of methane, hydrogen and carbon dioxide, which can be used for electricity generation or feedstock production at the surface.

Taking into account the UCG syngas production of fertilizers based on hydrogen, nitrogen and carbon dioxide is reasonable in industrial and emerging countries world-wide. For that purpose, the high pressure and temperature based Urea process can be applied to establish the nitrogen based fertilizer carbamide (CH₄N₂O). Carbamide is known to provide the lowest transportation costs compared to any other solid fertilizer due to its high content of nitrogen of about 46 w/w %. After gasification, hydrogen, nitrogen and carbon dioxide are required as input for the Urea process. Hydrogen and carbon dioxide can be separated from the synthesis gas with low energy demand as a result of the high initial process pressure. Nitrogen required for the Urea process is generated at the air separation unit which is applied anyway to separate oxygen from air in order to establish the necessary constitution of the oxidant for the UCG process.

The present study targets the development of an integrated carbon utilisation concept based on the coupled UCG-Urea process. A significant amount of carbon dioxide from the UCG synthesis gas is required for carbamide production in the Urea process, while the excessive carbon dioxide can be re-injected into the resulting cavities in the coal seams and surrounding strata after the gasification process. The volume of these cavities depends on UCG process control and offers remarkable storage ratios of more than 100% of the carbon dioxide originally generated during the UCG process. A theoretical feasibility study considering UCG-Urea process economics and potentials of UCG and carbon dioxide storage in the gasified strata was conducted for a selected study area in northern Bangladesh revealing the high competitiveness of the combined technology on the international feedstock markets.

2. Principle of the coupled UCG-Urea-CCS process

In this example, the UCG process uses 25 directional drillings with a spacing of 40 m and one production well as plotted in Figure 1 to develop a coal deposit of 1 km² size by in situ gasification using the CRIP (Controlled Retracted Injection Point) technology [1], [2]. This technology is based on utilisation of a liner located in the in-seam parts of the drillings to control the point of oxidant injection, and therewith the location of the gasification reactor. After exhaustion of a gasification reactor the liner is burned using an in-well burning device to develop a new reactor at the desired location in the seam [1]. The coupled UCG-Urea process in combination with carbon dioxide capture and its geological storage (CCS) in the UCG void volumes is illustrated in Figure 2.

An oxidant is produced in an air separation unit (ASU) providing oxygen for a subsequent mixing with water vapour. The sub-stoichiometric gasification process is then triggered and further on controlled by injection of the oxidant into the coal seam producing a high-calorific syngas mainly consisting of methane, hydrogen, carbon dioxide and carbon monoxide and extracted at the production well.
Tars, ash and sulphur are removed from the syngas stream at the surface. Subsequent to drying, carbon monoxide is combusted in the CO-shift process and CO\(_2\) is captured using one of the standard methods. Cryogenic rectification is applied to retrieve methane from the syngas and subsequent gas calibration separates hydrogen and nitrogen resulting from the UCG process which is then mixed with nitrogen separated at the ASU.

![Figure 1: Schematic view of directional drilling design as required for coal seam development considering the commercial-scale UCG process modified from [2].](image1.png)

![Figure 2: Schematic view of the coupled UCG-Urea-CCS process.](image2.png)
Carbamide is generated using the high P/T Urea process (about 14 MPa and 180°C). For that purpose, carbon dioxide is compressed in a multi-step compressor until achieving synthesis pressure while ammonia pressure (NH3) is increased using a high-pressure pump resulting in an exothermic condensation to ammonia carbamate (CH6N2O2). Followed by endothermic drying carbamate is converted into Urea (CH4N2O). This process is repeated using the CO2 strip process to consume the remaining ammonia. The products of Urea process are similar to the stoichiometric composition of ammonia and carbon dioxide, and thus cannot be further reduced.

3. Study area

Bangladesh was chosen as study area, since its significant shortage in fertilizer and electric power supply can be addressed by the coupled UCG-Urea-CCS technology. Bangladesh’s total fertilizer demand is increasing and additional Urea plants are not available. Installed capacity of the seven national Urea fertilizer plants sums up to 2.895 Mt Urea providing 2.480 Mt Urea while 0.749 Mt Urea (23.2%) had to be imported in 2006. Bangladesh immediately needs additional production capacities of 1.122 Mt Urea/y to cover its fertilizer demand [3]. In addition, the lack of electric power supply is responsible for the insufficient national fertilizer production which is consuming about 24% of the energy supplies. Total energy supply sums up to 3,800 MW/d, while total energy demand is about 6,000 MW/d. [4].

Economical application of UCG requires a continuous coal deposit with multiple coal seams without any conflicting interests with conventional coal mining. The Jamalganj coalfield located in north-western Bangladesh (Figure 3a) was discovered in 1962 and is suitable for development using the combined UCG-Urea-CCS technology. This suitability is mainly related to the deposit depth ranging from 640 to 1,158 m not relevant for conventional mining under prevailing economical considerations. Furthermore, the deposit is located close to the town of Jamalganj (Figure 3b), and thus close to the western branch of the north-south broad-gauge railway line [5].

The Jamalganj coal belongs to the Gondwana group mainly consisting of compacted and low permeable sandstones with coal layers and few shales. The Gondwana group is divided into Lower and Upper Gondwana. Lower Gondwana sequence holds seven coal seams with a cumulative thickness of 64 m located at depths between 640 and 1,158 m. Seam thicknesses vary from less than 2 m to about 46 m. Jamalganj coal was classified as high-volatile bituminous coal with a Vitrinite reflectance between 0.66 to 0.84%. The coalfield holds a total coal resource of 1.054 billion tons [5], [6], [7].

Figure 3 a) Gondwana coalfields of Bangladesh from [5]. b) Elevations of the top of the Jamalganj coalfield modified after [6], [7].

Figure 3b shows the locations of the exploration wells and the elevations of the top of the Lower Gondwana formation. According to the evaluation of the data provided by [5], the study area (as indicated in Figure 3b) located between the cities of Dhananjoypur in the North-West and Debral in the East is most suitable as pilot site due to the
4. Economical assessment

Any economical evaluation of the UCG-Urea-CCS technology requires detailed knowledge of the single processes involved. The calculation model developed by [2] was applied in this study and adapted to account for the Urea process. According to [1] 2,700 Sm³ of UCG syngas/t of coal can be produced with an average H₂ content of about 30% using the H₂-oriented UCG process as described by [8]. Based on an operating time of the UCG-Urea-CCS plant of 30 years, the N₂ required for the Urea process (118.7 t N₂/d) can be completely supplied by N₂ generated during oxygen production in the ASU (about 2,180 t N₂/d). Furthermore, a mass flow of 25.44 t H₂/d and 186.6 t CO₂/d are required for operation of a 250 t/d Urea plant. Remaining gas components not required for Urea production are 2,619 t N₂/d and about 814 t CO₂/d. CO₂ consumption of the Urea process amounts to 18.6% of the CO₂ produced during underground coal gasification. This excess CO₂ can be injected into the cavities resulting from underground coal gasification. The amount of injectable CO₂ depends on the total vertical influence of the gasification process. Gasification of a seam with low thickness (3 m) involving a resulting impact height of 3 m (equals the seam thickness without any roof influence) yields CO₂ storage potentials of about 14.2% of the CO₂ produced but not consumed for Urea production. This is equivalent to 11.6% of total CO₂ produced in the UCG-Urea process.

In the present study, a single seam with an average thickness of 3 m and an area of 1 km² is required to supply the necessary syngas to operate the 250 t/d urea plant. This involves the drilling of 25 injection wells and one production well resulting in a total of 63,804 meters to be drilled considering a seam inclination of 0.05° into a westerly direction. Costs of drilling and completion sum up to 191.4 M€ assuming average costs of 3,000 €/meter drilled. Regarding oxidant preparation 4 €/t O₂ are accounted for ASU operation (about 8 Mt O₂ are produced during the entire lifetime of the urea plant). Syngas processing (sulphur, ash and tar separation as well as drying) and CO₂-Shift sum up to 10 €/Sm³ syngas (9.27 M€ for entire life time of the urea plant). Costs for Urea production from the CO₂ capture process and storage in the gasified coal seams were assumed with 12.5 €/t CO₂. H₂ production in the UCG process equals to about 721 €/t H₂. Separation of H₂ from the syngas in the gas calibration process is accounted with 40 €/t H₂. Benefits of methane production and the cryogenic rectification process are neglected within this study. NH₃ production in the Urea process and its conversion to carbamide is accounted with 190 €/t.

Considering all components of the coupled UCG-Urea-CCS process costs for Urea production sum up to about 318 €/t Urea (dehydrated). These costs mainly depend on the impact of H₂ production which is mainly related to coal seam thickness and syngas composition.

5. Discussion

Regarding the calculation example presented with this study, the coupled UCG-Urea-CCS process can provide dehydrated Urea at production costs of about 318 €/t which is above the market price of 224 €/t reported by [3]. However, UCG process optimisation with regard to utilisation of coal seams with high thickness could significantly improve UCG-Urea-CCS economics, e.g. utilisation of a 15 m thick coal seam would decrease the costs to roughly 262 €/t Urea. In addition, drilling and completion costs have been assumed very conservatively in order to address uncertainties during the technological realisation of linking injection and production wells. A cost reduction can very likely be achieved with increasing amount of drilling targets related to the size of a UCG-Urea-CCS project. Furthermore, production of about 0.45 Mt of CH₄ was not considered in the economical evaluation of the coupled process. A sensitivity analysis on the calculated costs is currently being undertaken to determine cost variation based on uncertainties related to the UCG process as well as the Urea process, transport and Urea utilisation.

Based on the present calculation twelve UCG-Urea-CCS plants consuming about 45.9 Mt of coal would be required to provide the necessary 1.122 Mt Urea/year for 30 years in Bangladesh. Thus, the Jamalgonj coal field alone could provide Urea for about 690 years by application of the coupled UCG-Urea-CCS process. For this, a coal deposit with an areal size of at least 1 km², coal resources of more than 5.5 Mt and a cumulative seam thickness of 36 m would be required. CO₂ utilisation of about 18.6% and additional storage potential of at least 11.6% in the remaining coal gasification cavities underground (not taking into account vertical impact of the gasification and CO₂ sorption on the gasification residues as well as the surrounding rocks) have to be considered, when addressing up dip trend of the Lower Gondwana in westerly direction and the absence of major faults. Furthermore, the selected area is accessible using the available roadway infrastructure.
greenhouse gas mitigation combined with competitive fertilizer production technologies. Thus, the total CO₂ emission reduction potential is at least 30.2% due to its utilisation and underground storage.

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7. References

[1] Hewing, G.; Hewel-Bundermann, H.; Krabiell, K.; Witte, P.: Post-1987 R&D studies of underground coal gasification. Research Association for Second-Generation Coal Extraction, Essen, Germany, 1988, 55 pp.
[2] Kempka, T.; Nakaten, N.; Schlüter, R.; Azzam, R.: Economic viability of in-situ coal gasification with downstream CO₂ storage, Glückauf Mining Reporter, 1, VGE, Essen, 2009, p. 43-50.
[3] Quader, A.K.M.A.: Strategy for developing fertilizer sector in Bangladesh for sustainable agriculture, Chemical Engineering Research Bulletin 13, 2009, p. 39-46.
[4] Munim, J.M.A.; Hakim, M.M., Abdullah-Al-Manum, M.: Analysis of energy consumption and indicators of energy use in Bangladesh, Econ Change Restruct, Springer, 2010, 28 pp.
[5] Imam, M.B.; Rahman, M.; Akhter, S.H.: Coalbed methane prospect of Jamalganj coalfield, Bangladesh, The Arabian Journal for Science and Engineering, Volume 27, Number 1A, 2002, 17-27.
[6] Krupp Rohstoffe: Jamalganj Coal Project, Final Report, 2nd phase, Development and Mining Scheme Feasibility submitted to East Pakistan Industrial Development Corporation, 1966, 146 pp.
[7] Holloway, S.; Baily, H.E.: Coal Bed Methane Pre-Feasibility Study - NW Bangladesh, Technical Report Wc/95/59 R, British Geological Survey, Overseas Geology Series, 1995, 65 pp.
[8] Rogut, J.: EU-RFCS project Hydrogen oriented UCG in Europe, pers. comm., 2009.