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RFCS PROJECT METHENERGY+ METHANE RECOVERY AND HARNESSING FOR ENERGY AND CHEMICAL USES AT COAL MINE SITES

RFCS PROJEKT METHENERGY+ ZAJEM METANA IN NJEGOVA ENERGETSKA TER KEMIČNA IZRABA V PREMOGOVNIŠTVU

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Keywords: ventilation air methane, abandoned mine methane, coalmining, methane recovery and harnessing, thermal or chemical upgrading, adsorption-based technologies, materials development, thermal and catalytic regenerative oxidizers, methanol, greenhouse gases.

Abstract

Ventilation Air Methane emissions (VAM) from coal mines lead to environmental concern because of their high global warming potential and the loss of methane (CH_4) resources. How to tackle methane harnessing and its use was studied and analysed in the scope of the RCFS project, which was performed from 2017 till 2020, and coordinated by the University of Oviedo in Spain within the scope of an international consortium of eleven entities from Poland, Spain, the United Kingdom, Czechia, Greece, Slovenia and Sweden, combining universities, research institutions and industry (mostly Polish mines and the Slovenian Velenje mine). The main challenge tackled

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in the project was the use of methane released from both operating and abandoned mines, which is an environmental and safety hazard and also a useful source of energy. Therefore, the effective extraction of methane, its enrichment, purification, separation, thermal or chemical upgrading, and its use, considering coal mine site specifics, was assessed. Despite good operational results, after in-depth economic analysis of the integration, CAPEX and OPEX calculation, there turned out to be a high economic dependence on the cost of adsorbent, since adsorption was the most promising technology for concentrating the methane in these emissions. Therefore, the economic viability depends on the development of materials that meet a minimum cost and performance. Within the scope of the project, a lot of activities were carried out in order to widen and exploit the results.

Povzetek

Emisije odzračenega metana iz premogovnikov (t.i. VAM) imajo vpliv na okolje zaradi visokega toplogrednega učinka metana, po drugi strani pa pomenijo izgubo energetskega vira. Kako in na kakšen način izkoristiti metan, je bilo preučeno in analizirano v okviru RCFS projekta, ki se je izvajal od leta 2017 do leta 2020. Koordinator projekta je bila Univerza v Oviedo iz Španije, projektni mednarodni konzorcij pa je sestavljalo enajst partnerjev iz Poljske, Španije, Združenega kraljestva, Češke, Grčije, Slovenije in Švedske. Konzorcij je združeval univerze, raziskovalne ustanove in industrijo (večinoma Poljske premogovnike in Slovenski Premogovnik Velenje). Glavni izziv projekta je bila preučitev možnosti zajema in uporabe metana, emitiranega iz delujočih in zaprtih premogovnikov, saj je le-ta škodljiv za okolje in predstavlja varnostno tveganje, po drugi strani pa koristen energetski vir. Vsled tega se je preučila učinkovita ekstrakcija metana, njegova obogatitev, očiščenje, separacija, toplotna ali kemična nadgradnja in njegova uporaba ob upoštevanju specifik posameznih premogovnikov. Kljub dobrim operativnim rezultatom, poglobljeni ekonomski analizi integracije, izračunom investicijskih in obratovalnih stroškov, se je pokazala velika odvisnost ekonomike od stroškov adsorbenta, saj je bila adsorpcija najbolj obetavna tehnologija za koncentracijo metana glede na velikost emisij. Zaradi tega je ekonomska opravičenost pogojena z razvojem materialov, ki bodo imeli nizke stroške in istočasno dobro učinkovitost. V okviru projekta se je opravilo veliko aktivnosti z namenom razširjanja in eksploatacije rezultatov.

1 INTRODUCTION

Coal mine Premogovnik Velenje (PV) participated in the EU's METHENERGY+ project, cofounded by the Research Fund for Coal and Steel (RFCS) programme. RFCS is an EU funding programme supporting research projects in the coal and steel sectors. The RFCS has its own legal basis and stands outside the Multiannual Financial Framework. It is funded via the revenues generated by the European Coal and Steel Community (ECSC) in liquidation assets, which are exclusively devoted to research in the sectors related to the coal and steel industries.

The project with title and subject "Methane recovery and harnessing for energy and chemical uses at coal mine sites", was performed from 2017 till 2020, and was coordinated by the University of Oviedo in Spain within the scope of an international consortium of eleven entities from Poland, Spain, the United Kingdom, Czechia, Greece, Slovenia and Sweden, combining universities, research institutions and industry (mostly Polish mines and the Slovenian Velenje mine).

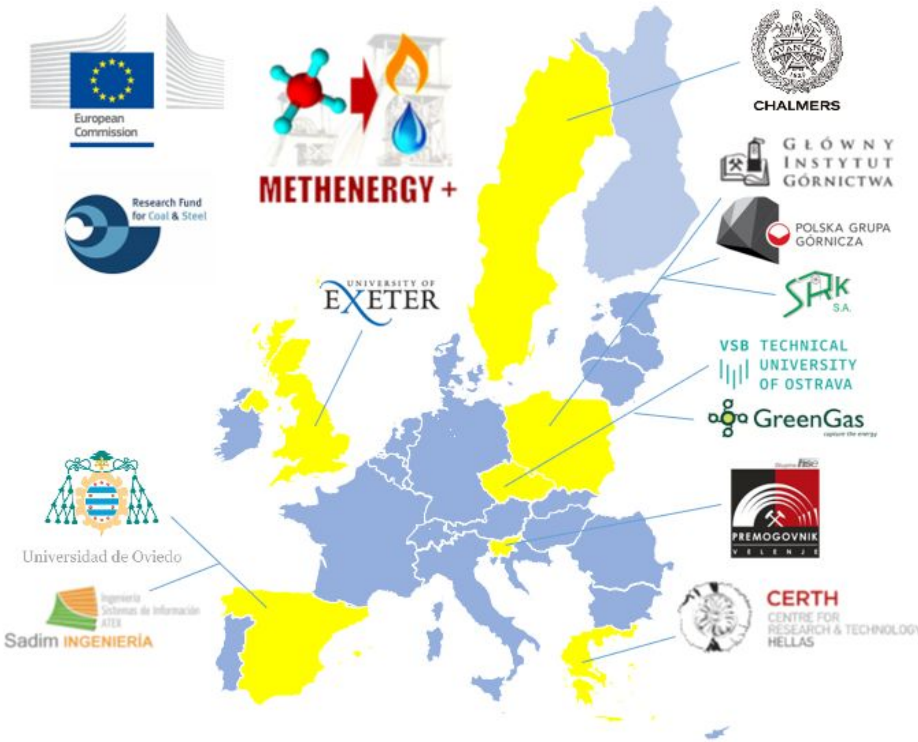

Figure 1: *Project METHENERGY+ partners*

The main challenge tackled in project was the use of methane released from both operating and abandoned mines, which is an environmental and safety hazard and also a useful source of energy and raw material for manufacturing chemicals. Therefore, effective methane extraction, its enrichment, purification, separation, thermal or chemical upgrading, and its use, considering coal mine site specifics, was assessed.

2 SOURCE OF METHANE

Coal production releases CH_4 trapped in coal seams and surrounding strata, and which can be categorised into three types:

- CH_4 drained from the seam before mining (60–95% CH_4),
- CH_4 drained from worked areas of the mine (30–95% CH_4), and
- CH_4 diluted through ventilation fans (0.1–1.5% CH_4) while extracting coal.

Therefore, there are mainly three different types of mining emissions containing methane:

- coal bed methane (CBM),
- abandoned mine methane (AMM) and

- ventilation air methane (VAM).

The first two usually contain high and medium purity methane (>30%), and are easily exploited with the well-known harnessing technologies available.

VAM has a very low concentration, so it has very low energy efficiency, and is in most cases vented into the atmosphere via underground coal mine fan stations.

In the case of Premogovnik Velenje in Slovenia, the maximum allowed gas concentration at fan stations is 0.5%, which is carefully monitored due to safety reasons. What also needs to be highlighted are the widely fluctuating concentrations resulted from underground mining activities. Major influence factors include the intensity of mine face production and also mining method, mine geology and type of coal extracted. In the project, concentrations and parameters from partner mines where average VAM concentrations in the range 0.1–0.3% CH_4 with very high total flow rates (even as high as 200 Nm^3/s) were considered.

Therefore, based on the average results of several authors and measured concentrations in project partner mines, a stream with fair to moderate methane concentration and air flow rate values has been selected for the analysis made: 0.57% CH_4 and 4.4 Nm^3/s, respectively [2].

3 SCOPE OF THE PROJECT

The scope of the project was:

1) Optimisation of methane recovery (methane concentration and flow rate) at operating and flooded coal mines, considering the geological and operational features of the shafts.

2) Development of adsorption-based technologies for concentrating methane from ventilation air methane (VAM) and abandoned mine methane (AMM) emissions, including materials development and process design and simulation. Different approaches will be used for preparing the potential adsorbents, including the development of high-performance tailored materials or the use of waste materials from other processes as adsorbents.

3) Development of a membrane-based technology for the separation of methane from VAM and AMM emissions, including the development of perm-selective membranes using nano-structured materials, as well as the modelling and simulation of these units at industrial scale.

4) Explore the possibility of using thermal and catalytic regenerative oxidizers for the efficient and environmentally safe combustion of these emissions (with and without previous methane enrichment).

5) Explore the possibility of transforming the methane contained in the studied emissions (with or without further enrichment) into other valuable chemicals, such as methanol or hydrogen.

6) Evaluate the application of the above technologies for methane enrichment and utilisation, both to operating mines and to mines where coal mining activity has ceased.

3.1 Methane recovery at deep coal mine sites within the whole life cycle of a mine

Exhaustive research and analytical work was carried out to demonstrate the impact of various factors on the possibility of collecting and flowing water and methane in mining excavations, with particular emphasis on mine closure. On the basis of a review of the methods of capacity assessment of underground water reservoirs and filtration properties in the rock mass, using the methods of mining hydrogeology, a description of the process of water and gas accumulation and the conditions of their flow in mining excavations and in goafs was prepared. A database was prepared on selected components of mine gases for 15 active and 1 closed mine, with a total of 32 shafts in the analysis. The results of measurements of methane concentration, temperature, humidity and air composition for all available levels were presented. A total of approximately 50,000 datapoints were compiled and analysed. Moreover, a catalogue of the main natural and technical factors determining the flow and accumulation of methane and water in the workings of active and liquidated mines was developed. An outline of the recommended methodology for estimating the amount of gas (methane) emitted during the flooding of mining excavations is presented in the example of a liquidated test mine. The geological, mining and hydrogeological conditions and the impact of their changes on the methane potential of the mines on the scale of the coal basin were verified. The impact of these conditions on the possibility of considering scenarios and models of mine closure in terms of methane recovery was assessed. Further development of the scenarios was carried out.

3.2 Adsorption technologies for methane recovery in VAM

Research work was done on adsorption technologies for methane recovery in VAM to establish the potential of adsorption processes for the separation of methane from ventilation gases, to gather relevant data on the most appropriate adsorbents and adsorption technologies, depending on the characteristics of the emissions, and to model on the lab scale in order to provide useful information for implementation at the mine site. More materials were considered and compared, including porous carbon materials from zeolite, zeolites prepared from fly-ashes, and MOFs[1] (two different families, Basolites and IRMOFs). Adsorption processes were rigorously modelled, both taking into account adsorption and desorption steps. Modelling codes were experimentally validated for different materials considered in this project. A process based on the use of temperature swing adsorption (TSA) was proposed and designed.

3.3 Tailored materials for methane recovery using membrane processes

Tailored materials for methane recovery using membrane processes were assessed and studied to:

- establish the potential for the separation of methane from ventilation gases using membrane technologies
- determine the optimum materials and procedures for the preparation of membranes for methane concentration in VAM

[1] Metal-Organic Framework.

- To model the lab scale results in order to provide useful information for implementation at mine sites

Mass transfer has been modelled using the Maxwell-Stefan multicomponent surface diffusion model. The model performance has been validated using the limited literature data available for this type of mixture. The application of this model has been extended by simulating the concentration profiles along the length of the membrane module using the plug flow model.

3.4 Combustion technologies

There was a systematic comparison of thermal (TFRR or RTO) and catalytic (CFRR or RCO) flow reversal reactors for methane abatement in mine gases. Attention was also devoted to designing a reactor for mine implementation in order to reduce VAM and AMM emissions and to optimise the design of control strategies for this kind of reactor. Formation of secondary pollutants (NOx), which are present especially in the case of CFRR, was determined.

Methane concentration in ventilation air of the different mines analysed ranges from 0.10% to 0.25% (vol.) in most situations. These are very low values, which can be handled by RCO (minimum concentration of 0.18%), but barely with RTO (minimum concentration of 0.36%). In the latter case, only the use of high efficiency regenerative oxidizers may lead to autothermal operation, e.g. for a methane concentration of 0.25% the thermal efficiency should be higher than 93%.

Environmental and safety issues associated with these technologies were deeply analysed. Secondary environmental impacts of these technologies are negligible. Concerning safety constraints, these are also manageable.

The recovery of the energy associated with methane from coal mine ventilation air emissions can be done using lean-burn gas turbines (EDL, CSIRO, IR, etc.). These types of turbine are especially suited to work with low methane concentrations and are able to produce work (electricity) directly. Lean-burn turbines are not widely used for the harnessing of VAM, because they require a minimum methane concentration of 1% [7], which is rather high for most VAM emissions. Hence, a prior methane concentration step is needed. A combination of adsorption processes with gas turbines has been proposed in this work as a realistic strategy for upgrading these emissions.

3.5 Chemical upgrading of VAM

The objectives of this work package were the following:

To select catalytic technologies for the conversion of methane at the conditions encountered both in mine methane emissions and in methane-enriched emissions.

To select and test under real conditions the catalysts and reactors useful for methane upgrading in mine methane emissions.

To study the influence of reactor configurations and operating variables on the performance of the system.

The most promising upgrading approach for both enriched VAM and AMM are based on the direct oxidation to methanol. Among the different approaches suggested in the literature, gas phase heterogeneous oxidation using metal loaded zeolites seems to be the most feasible alternative.

Different Cu and Fe-loaded zeolites were prepared and tested for the direct oxidation of methane, the best results being obtained with Cu-loaded mordenite.

A preliminary design of a process based on this approach has been accomplished. The upgrading of diluted methane emissions into valuable products can be accomplished at low temperatures (200°C) by the direct partial oxidation of methanol over copper-exchanged zeolite catalysts. The reaction has been studied in a continuous fixed-bed reactor loaded with a Cu–mordenite catalyst, according to a three-step cyclic process: adsorption of methane, desorption of methanol, and reactivation of the catalyst. The purpose of the work is the use of methane emissions as feedstocks, which is challenging due to their low methane concentration and the presence of oxygen. Methane concentration had a marked influence on methane adsorption and methanol production (decreased from 164 µmol/g Cu for pure methane to 19 µmol/g Cu for 5% methane). The presence of oxygen, even in low concentrations (2.5%), reduced methane adsorption drastically. However, methanol production was only affected slightly (average decrease of 9%), concluding that methane adsorbed on the active centres yielding methanol is not influenced by oxygen [4].

It has been demonstrated that the desired reaction can be accomplished even in the presence of air. For this purpose a cyclic operation was proposed, treating the catalyst with the methane containing gas and with steam in successive cycles. These cycles have been optimised during this period.

Evaluation and integration

A methodological approach for determining methane recovery from both operating and abandoned coal mines has been developed. The approach takes into account hydrogeological modelling of the coal basins.

Integrated processes for the harnessing of coal mine ventilation air methane has been proposed, designed and economically evaluated. The concentration of methane in the ventilation air is a critical parameter for the selection and design of an adequate harnessing technique. This concentration may differ from mine to mine, and at different times during mine operation. However, during the mine closure and flooding processes, this concentration may increase for some mines. The mine case study considered a ventilation air flow rate of 4.41 Nm^3/s with methane concentration of 0.57%.

The use of a prior concentration stage based on fixed-bed adsorption has been proposed to raise methane concentration before harnessing. In this context, two integrated schemes have been analysed and proposed:

Adsorption + Combustion in regenerative oxidizer. The fixed-bed adsorption unit is filled with Norit GF-40 active carbon as adsorbent. For this concentration, heat recovery efficiency can be as high as 54%, which corresponds to an equivalent saving of 36.5 Nm^3/h of natural gas.

Adsorption + Combustion in a gas turbine. In this case, the ventilation air is concentrated in a fixed bed equipped with Basolite C300 adsorbent. This adsorbent is capable of concentrating methane from 0.57% to 1.2%. The resulting concentrated stream is harnessed in a gas turbine with a generation of 490 kW of net power as electricity.

Next we describe case 2, where the concentration step is carried out in a fixed bed temperature-swing adsorption (TSA) operation. This unit is inherently discontinuous, with adsorption happening in the first step and, once the adsorbent is saturated, methane is recovered by desorption at a higher temperature. The concentration step must increase methane concentration to a minimum of 1% in order to use a lean-burn gas turbine for the combustion.

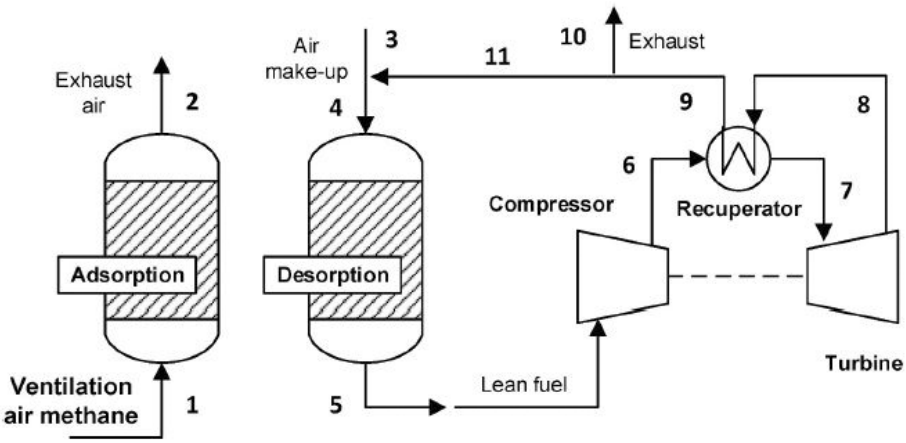

Figure 2: *Flowsheet of the TSA-turbine integrated process.*

The lean-burn gas turbine is made of three elements: compressor, recuperator and turbine. The turbine is capable of generating net work and, hence, produce electricity. The recuperator is used to pre-heat the feed before combustion using part of the energy of the combustion gases. In addition, part of these combustion gases can be used as the drag stream of the desorption process as shown in upper Figure [5]. This level of mass and heat integration is critical in order to save energy and improve the economy of the process.

4 ECONOMIC EVALUATION

The next economic evaluation shown is for adsorption and combustion in a gas turbine case, as described in the previous section.

The process is mass and energy integrated, since the heat required by the adsorption/desorption operation is fully supplied by the exhaust from the gas turbine.

The entire design includes the material and devices needed to carry out the whole project. The economic evaluation has been done using the guidelines provided by the US Environmental Protection Agency (EPA). This organisation has published a document entitled “Air Pollution Control Cost Manual” [6], which provides guidance for the design and costing of the equipment used for pollution control.

Table 1: *The capital investments–Adsorption and combustion in a gas turbine*

CAPITAL INVESTMENT		
Direct Costs		
Purchase equipment costs		
Equipment cost	A	2 313 031 €
Instrumentation	0.1A	231 303 €
Sales taxes	0.03A	69 391 €
Freight	0.05A	115 652 €
Total	B=1.18A	2 729 377 €
Direct installation costs		
Foundations & supports	0.08B	218 350 €
Handling & erection	0.14B	382 113 €
Electrical	0.04B	109 175 €
Piping	0.02B	54 588 €
Insulation for ductwork	0.01B	27 294 €
Painting	0.01B	27 294 €
Total	0.3B	818 813 €
Total Direct Costs	DC=1.3B	3 548 190 €
Indirect Costs		
Engineering	0.10B	272 938 €
Construction	0.05B	136 469 €
Contractor fees	0.10B	272 938 €
Start-up	0.02B	54 588 €
Performance test	0.01B	27 294 €
Total Indirect Costs	IC=0.28B	764 226 €
Contingency costs	CC=0.10(DC+IC)	431 242 €
Total Capital Investment	**TCI=DC+IC+CC**	**4 743 657 €**

The Main Equipment Costs exclude the piping, instrumentation and auxiliary equipment. These costs are accounted for as direct costs of the Capital Investment, which are estimated as a function of the total Cost of Main Equipment (Cost reports and guidance, 2018). Final calculated Total Capital Investment (TCI) is 4.74M €.

The Annual Operating Costs have also been estimated.

Table 2: *Annual operating costs of the integrated adsorption and gas turbine*
Annual operating costs of the integrated adsorption and gas turbine.

ANNUAL OPERATING COSTS		
Direct Annual Cost	8000 h/year	
Operating Labour		
Operator	0.5 h/shift	12 925 €
Supervisor	15% operator	1 939 €
Operating Materials	-	
Maintenance		
Labour	0.5 h/shift	13 191 €
Materials	100% labour	13 191 €
Adsorbent replacement		6 972 687 €
Utilities		
Electricity consumed	0.07€/kWh	16 324 €
Electricity generated	0.07€/kWh	-274 288 €
Total Direct Annual Costs		6 755 968 €
Indirect Annual Cost		
Overhead	60% op. maint.	24 747 €
Administrative charges	2% TCI	94 873 €
Property taxes	1% TCI	47 437 €
Insurance	1% TCI	47 437 €
Capital recovery		-167 623 €
Total Indirect Annual Cost		46 870 €
Total Annual Cost		**6 802 838 €**

These costs include labour, materials, maintenance, utilities and administrative charges. A yearly operation of the plant of 8,000 hours has been considered in the calculations. Additionally, the necessary replacement of the adsorbent used should also be considered as Annual Operating Cost, since these materials have a lifetime that is lower than that of the main equipment, such as vessels or fans. It has to be replaced periodically due to degradation and loss of capacity. For this reason, its cost must be divided and accounted for as annual, following the recommendations of the EPA, considering a lifetime of n = 4 years and an interest rate of i = 4.25%, which results in a factor FWF = 0.2346. An additional 8% is added to account for freight and taxes. The cost of the adsorbent is a key parameter in the economic study, so a sensitivity analysis was performed in order to study the viability of the process depending on it. From now on, an arbitrary X value is considered for the cost of 1 kg of adsorbent material.

The utilities consist electricity consumed by the fan, which provides the required gas flow rate through the different equipment. Considering an electricity price of 0.07 €/kWh and the power consumption of the fan (147 kW), the annual electricity consumption cost is estimated at 82,320 €/year. However, the gas turbine is able to generate 490 kW of net work, directly as electricity. This power should be discounted from that consumed by the process. Considering the same electricity price, the gas turbine generates earnings of 274,288 €/year, which are included as negative costs in Table 2. As shown, the total annual cost depends heavily on the adsorbent material cost

(X). In addition to the costs reflected in previous tables, the positive environmental impact of the process should be taken into account. This is accounted for by the carbon emission allowances.

The economic evaluation points out the importance of the adsorbent cost. In fact, for the process to become profitable, a lower adsorbent cost of 0.6 €/kg is desirable.

Results of the economic evaluation for different adsorbent costs (X). Annual cash flow (blue line), NPV (orange line). Black arrows point out the necessary adsorbent cost to make the NPV equal to zero.

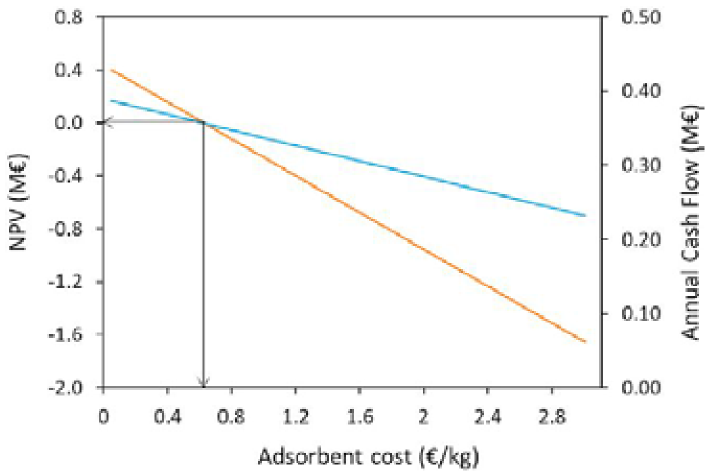

Figure 3: *Economic evaluation at different adsorbent costs*

Therefore, the final conclusion is that the technical feasibility of both combinations has been demonstrated, whereas the economic viability will depend on the development of materials that meet a minimum of cost and performance.

5 CONCLUSIONS

As there were several mining companies involved, different technical solutions were offered depending on the characteristics of each mine site. Among the studied approaches, the combination of an adsorption process and a gas turbine seems to be the most attractive approach for future implementation. This project proposes the combination of three strategies for solving this significant problem:

1) Optimise the ventilation procedures in order to get the highest CH_4 concentrations extractable under safe conditions. This activity has been extended to abandoned and flooded mines, for which higher methane concentration can be achieved.

2) The development of procedures for concentrating the methane in these emissions. The most promising technologies are those based on adsorption and membrane technologies.

3) Study of combustion and chemical transformation technologies in the presence of oxygen. In previous research, it has been observed that flow reversal combustors provide the best performance for the combustion of these emissions, but there remain several underexplored aspects,

such as the environmental effects, the control strategies, and integration with the overall energy management system of the plant.

This work has studied the feasibility of harnessing low-concentrated methane streams by integrating two independent processes: temperature swing adsorption for methane concentration, followed by combustion in a lean-fuel burn turbine for obtaining a surplus of electricity and calorific energy. The design has been made for a VAM inlet stream, which has a flow rate of 4.4 m^3/s, with an average methane concentration of 0.57% CH_4 in air.

Despite the good operational results, after in-depth economic analysis of the integration the process shows a high dependence on the cost of adsorbent.

A lot of activities were carried out in order to widen and exploit the results. It should be noted that an important part of the results was published. Regarding the industrial exploitation of the results, the deep study about methane release during mine operation and flooding will be very helpful for designing ventilation systems. For example, the next fan stations and ventilations systems planned for underground mines in the future could be done in reference to the findings of this project.

Project METHENERGY+ has been proposed as an example of global warming related initiatives performed at the University of Oviedo within the activities heldin Oviedo. Project was presented at several conferences in Spain, Germany and Poland.

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