

# CO<sub>2</sub> gas production understanding above a partly flooded coal post-mining area

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## Abstract

INERIS worked for several years on the characterization of O<sub>2</sub>-defficient and CO<sub>2</sub>-enriched gas production and migration processes above a partly flooded former coal mining area.

To understand the origin of this production, a borehole of 90 meters deep was drilled next to the phenomena. Several investigations were conducted to improve the understanding of mechanisms of gas production in relation to the presence of water.

This paper deals with the results of experiments conducted in situ and tests done in the laboratory. One hypothesis is proposed due to the results obtained: marcasite, which has been observed on rock samples, is oxidized by contact with the air that migrates through mining fractures in the rock. CO<sub>2</sub> is produced through carbonate dissolution because of the production of acidic fluids in the water. Noxious gas emissions can occur during atmospheric pressure drops and can enter houses, flowing along cracks.

Keywords: *Noxious gas production ; Water ; Pyrite ; Marcasite ; Oxygen ; Carbon dioxide*

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## 1. Introduction

Variations of atmospheric composition are observed in some houses located above a former coal mine associated with a rising water table. These variations are characterized by a high content of CO<sub>2</sub> gas (up to 8%vol.) and low content of O<sub>2</sub> (down to 7%vol.). In France, legal exposition limits values are respectively of 1%vol. and 19%vol. These noxious gas emissions occur generally during atmospheric pressure drops. Various hypotheses were first proposed due to the gas emissions location and the hydrogeological context.

Lafortune [1] carried out isotopic investigations on carbon isotopes, in order to identify the carbon source. A nearby bog was rejected as the source because the carbon isotopic signatures do not match.

Deep water (the groundwater table is about 90 m deep) analysis permits us to exclude a dissolved gas origin, as well. In order to improve the understanding of the mechanisms involved, a borehole was drilled in March 2012. The borehole location was near the houses impacted by noxious gas emissions and in the

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same geological and hydrogeological context if possible. Hence, different experiments were conducted: during and after drilling (in situ and at the laboratory). This paper deals with these different experiments, the results obtained, and the hypothesis which could be emitted and verified.

### *1.1. Geological and hydrogeological contexts*

The noxious gas emission phenomenon occurs in a town in France located in the Lorraine coal basin. The geological and hydrogeological context can be summarized by (down to up):

- Carboniferous
  - The Westphalian deposit is constituted by numerous exploited coal seams of different thicknesses. These seams are located 600 m depth below the surface. Dewatering was stopped in 2006.
  - Stephanian stratigraphic units: they are constituted by conglomerates, shales and sandstones whose thickness reaches 200 m.
- Permian is constituted by dolomitic conglomerates and sandstones that are not very permeable and represent a stratigraphic level of 25 to 50 m thick.
- Trias: sandstones from Lower Trias are red or yellow, very porous, around 200 meters thick and they contain an important aquifer used for human activities and as drinking water. In July 2012, groundwater table was around +135 m NGF (i.e. 90 m deep).

Obviously, coal mining has induced discontinuous deformations between Carboniferous and Trias stratigraphic levels in several areas, with a maximum collapse of 15 m (see DREAL Lorraine, [2]).

### *1.2. Expected geochemical reaction in link with gas noxious emission*

The chemical reaction proposed by INERIS induces several successive geochemical reactions, depending on the following chronology which was confirmed by Agrinier [3]. In a first step, ferrous sulphur (pyrite or marcasite) is oxidised, according to:

(1)

Then, iron is oxidised, according to:

(2)

And then, iron hydroxides precipitates:

(3)

The water is acidified by these reactions by the cumulated production of  $16 \text{ H}^+$ . If carbonates are present, this can interact and generate  $\text{O}_2$  consumption and  $\text{CO}_2$  production, according to:

(4)

## **2. The drilling**

The chemical reactions previously described need the concomitant presence of water and atmospheric oxygen which can be observed in the capillary fringe. In order to confirm this, a borehole was drilled during March 2012 down to this zone. The method of continuous core drilling was employed and the core equipment was outfitted with a tube that had fringes to permit the flowing of gas from the strata encountered.

### 3. Experimentations after the drilling

#### 3.1. In situ experiments

Gas analyses were conducted from the top to the bottom of borehole to obtain gas concentration logs. The gas analyser permitted us to measure simultaneously CO<sub>2</sub>, O<sub>2</sub>, CH<sub>4</sub> and CO concentrations. These tests were done during a barometric pressure drop, which is a good condition for gas emission to the surface. Water samples were taken and unstable parameters such as temperature and pH (which was around 7.8) were measured with a multi-parameter probe.

#### 3.2. Laboratory experiments

Several analyses were conducted. First, rock samples were chosen to be analysed with a SEM (scanning electron microscope) in order to find sulphur crystals, thin sections were cut to search carbonates. Secondly, rock samples were put in batches with demineralised water to observe if possible the in situ supposed reactions. The gases variations were observed with the same analyser used in situ.

### 4. Results and interpretation

#### 4.1. In situ experiments

Figure 1 shows the results of one of the concentration gas logs carried out in the drilling.

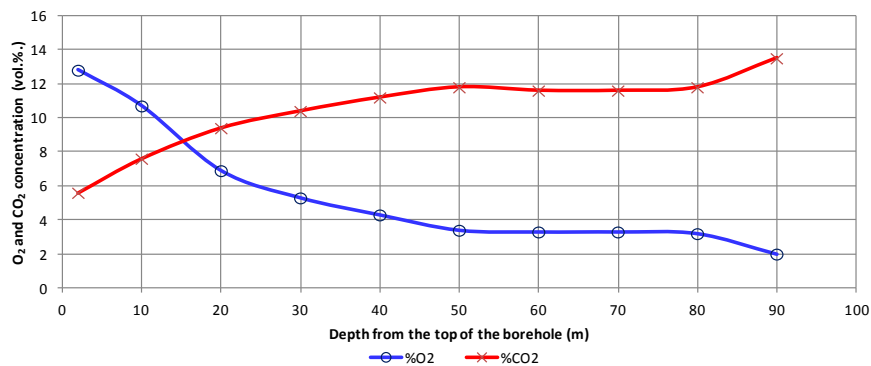


Figure 1: O<sub>2</sub> and CO<sub>2</sub> concentration variations with depth

CO<sub>2</sub> gas content varies from 5.6% at the top of the borehole to 11.8% at 50 m depth whereas O<sub>2</sub> varies from 12.8% to 3.4%. Below 50 m, these concentrations stay mostly constant down to the bottom of the borehole.

#### 4.2. Laboratory experiments

Marcasite (or white iron pyrite) was found on several samples, in an oxidized state or not. It's a ferrous sulphur which is more oxidizable than pyrite. Carbonates were found on rock samples, after thin sections analysis. Laboratory tests were conducted in batches. After having sterilized the samples in order to eliminate all bacteria, it was observed that O<sub>2</sub> was consumed and CO<sub>2</sub> was produced in the proportion of the theoretic line plots on Figure 2. This has been interpreted as the result of marcasite oxidation followed

by carbonate dissolution.

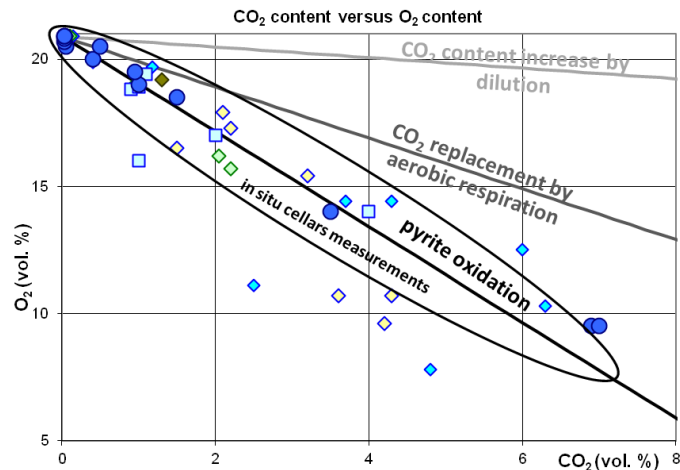


Figure 2: CO<sub>2</sub> versus O<sub>2</sub> content and main theoretical lines (molecular balance line of: pyrite oxidation and carbonate dissolution in black, CO<sub>2</sub> replacement by aerobic respiration in dark grey and CO<sub>2</sub> concentration increase by dilution in pale grey)

## Conclusion and outlook

The presence of high content of CO<sub>2</sub> and the lack of O<sub>2</sub> were confirmed by measurements in the borehole. With all the experiments conducted, it was confirmed that marcasite, water, and carbonates are present on the site in the rocks at different levels. Other rock samples still have to be analysed. The hypothesis of marcasite or pyrite oxidation is proposed to explain noxious gas production. Another gas concentration log along the borehole and after an air lift (with nitrogen) will be made to try to locate the gas production zone. Research will be continued on pyrite oxidation reaction because some other assumptions could be made as Schieber [4] has done in a different context.

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