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BIOMETHANE ACCEPTANCE IN UNDERGROUND GAS STORAGE FACILITIES

February 2022

Contact

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ABOUT MARCOGAZ

Founded in 1968, MARCOGAZ represents 28 member organisations from 20 countries. Its mission encompasses monitoring and policy advisory activities related to European technical regulation, standardisation and certification with respect to safety and integrity of gas systems and equipment, rational use of energy as well as environment, health and safety issues. It is registered in Brussels under number BE0877 785 464.

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1. Introduction

Underground Gas Storage facilities (UGS) are a vital part of the gas infrastructure within the European Union (EU) and in the world. The storage of renewable gases in UGS is essential to support the energy transition and to decarbonize the gas sector. Currently, in the EU, the gas composition in the gas grid is changing because of the injection of fuel gases produced from renewable energy sources. The storage of renewable gases in UGS facilities can be carried out efficiently and is already realized when required gas quality specifications are met.

In this document, the following definitions from EN ISO 14532 are used:

- **Biogas**: gas, comprising mostly methane and carbon dioxide, but also minor or trace components (e.g. H_2S) obtained from the anaerobic digestion (fermentation) of biomass.

- **Biomethane**: gas comprising mainly methane, obtained from either upgrading of biogas or methanation of biosyngas.

- **Anaerobic digestion biomethane**: gas comprising predominantly methane, obtained from upgrading of biogas

As far as biomethane acceptance in UGS facilities is concerned, two main topics must be addressed. The first one is about **trace components** and the second is about **oxygen (O_2) content**. This last topic is still under investigation and this paper draws a panorama of what is known and proposes a position.

According to available studies, the level and species of micro biological organisms that are roughly the same both in biomethane as in natural gas [19] is not considered by now as a concern.

The present paper is dealing with the implications of biomethane injection into the gas transport grid and then in all types of underground gas storage facilities, even if some concerns are specific to certain types of storages.

2. Trace components of biomethane and their impact on UGS facilities

Many UGS facilities in France are aquifer storages and their operators are very cautious about the impact of the injected gas quality on the subsurface geo-chemistry and geo-microbiology. That is the reason why they launched a dedicated project ("Gazelle") on trace components in biomethane in 2013. Since 2016, a campaign of biomethane sampling and analysis has been conducted throughout France on behalf of the four main French gas infrastructure operators (GRDF, GRTgaz, Terréga and Storengy). The project targets the trace compounds that could potentially be harmful for UGS in aquifer. Results of the study were presented at the World Gas Conference 2018 [7] and at the International Gas Research Conference 2020 [8].

The most frequent compounds are:

- **Ketones**: 2-butanone and 2-pentanone
- **Terpenes**: Limonene, pinene and camphene
- **BTEX**: benzene and toluene
- **Hydrocarbons**: methylhexane, methylcyclopentane, cyclohexane, methylcyclohexane, n-heptane, n-octane and n-decane
- **Siloxanes**: Octamethylcyclotetrasiloxane (D4)

Thiophenes were also detected, especially Thiophene and THT (Tetra-Hydro-Thiophen).

Some siloxanes were detected in more than 50% of the analysis, but at very low concentrations.

Heavy metals were present with very low concentrations, below 1 $\mu g/m^3_s$.

Polycyclic-aromatic compounds (PAHs) and halocarbon compounds have not been detected in biomethane samples.

Data science study on the available results also showed that:

- Biomethane coming from landfills contains about 40 compounds, which represent less than 10% of the total 400 analysed compounds.
- The cryogenic purification and PSA[1] processes generates biomethane containing less trace compounds than the membrane-based purification process.
- Many trace compounds are only observed in the biomethane produced from agricultural waste and/or derived from a membrane-based purification process.

The results of this study show that very few trace elements are detected in biomethane. Most trace elements are already present in natural gas at same or lower concentration, are non-toxic, or are present at a very low concentration to impact the quality of water in aquifer.

[1] PSA – Pressure Swing Absorption

Moreover, the trace components concentrations are far below the limit values for contaminants in biomethane based on health assessment criteria, as calculated in the CEN Technical Report - CEN/TR 17238:2018 "*Proposed limit values for contaminants in biomethane based on health assessment criteria*" - by Technical Committee CEN/TC 408 "*Natural gas and biomethane for use in transport and biomethane for injection in the natural gas grid*".

Further analysis on other production plants with different upgrading processes would be useful to confirm these results. As of December 2021, GERG is conducting pre-normative research on biomethane (refer to chapter 5.1 - Annex) and expects to produce a database on the composition of biogas and biomethane in Europe.

As a conclusion, if the composition of biomethane complies with standards and regulations, its storage in deep aquifer shouldn't create any significant impact on the water quality of the aquifers.

3. Oxygen impact on UGS facilities

On one hand, it has to be pointed out that natural gas contains no oxygen at production stage. An EASEE-gas measurement study proved that oxygen contents in the transported natural gas is lower than 1 ppm.

On the other hand, biomethane usually contains around 3000 ppm of oxygen, and sometimes higher concentration (4000-4500 ppm) [14]. This oxygen may come from the feedstock but it is in overall mainly a consequence of the desulfurization process. Indeed, to remove H_2S produced by fermentation, air or oxygen is injected in the fermenter. Presence of oxygen allows sulphur precipitation on nets in the upper part of the fermenter and in active coal vessels in the gas treatment plant downstream the fermenter. As the injection of air is coarsely dosed, an excess of oxygen remains in the biomethane that leaves the biogas plant.

As other components defining the gas quality according to local standards, the oxygen content should be monitored before the injection into the grid and into UGS. A detailed analysis, with special attention to the interaction of critical components such as oxygen, carbon dioxide and hydrogen sulphide, is mandatory in order to ensure integrity and safety of the facilities. Fluctuation of component concentrations, changing flow directions and the relevant positive mixing effect in the gas grid have to be considered.

3.1 Possible negative effects on UGS facilities (surface facilities, wells and reservoir)

Concerning the impact of oxygen on storage facilities, the specific conditions of each UGS, which are different from each other, have to be considered individually. For instance: high pressures, high pressure drops, higher temperature, water salinity, wet gas regime and related dehydration requirements, interaction with reservoir minerals, high surface area of pore space and chemical reaction of gas components.

The consequences of oxygen on the gas injection facilities of UGS studied so far ([3], [10] and [11]) may be the following. Some of these effects have been observed in the laboratory, others in situ in reservoir and UGS facilities (refer to annex).

- Corrosion in wet gas equipment (i.e. upstream dehydration towers used during withdrawing operations) and resulting damage of subsurface and surface installations;
- Subsurface and surface sulphur precipitations and resulting corrosion, malfunction of valves and safety devices, plugging of pores and capacity /availability impairments;
- Generation of “Black Powder” / pyrophoric iron;
- Blockage by oxidation of minerals in formation water;
- Impact on geo-biological equilibrium in depleted oil and gas reservoirs as well as in aquifer UGS;
- Accelerated degradation of glycol in dehydration units [15].

The consequences are strongly depending on O_2 concentration.

The main issues related to O_2 in UGS are: integrity, capacity impairment, related costs, safety and security of supply.

Integrity of storage facilities can be jeopardized due to corrosion by O_2 or/and in interaction with other critical gas components or by precipitated sulphur in combination with water. Repair and work over jobs may be required which result in downtime, additional costs or are technically/economically not possible due to severe damages (e.g. of casings in a well completion).

The well-known formation of elemental sulphur deposits, experienced in several UGS, impacts integrity, capacity, costs and safety [16].

H_2S is present in differing concentrations nearly in every natural gas stream. A reaction of oxygen with H_2S will result, aside from other mechanisms, in elemental **sulphur precipitations** and **acidification of the medium**.

These sulphur precipitations may result in severe disruptions of storage operations, i.e. by blocking of valves with elemental sulphur, as well as of corrosion under such sulphur deposits (Figure 1). Due to the sulphur precipitations in armatures and resulting malfunction, safety may be jeopardized.

Figure 1: Sulphur deposits in control valve of UGS

Source: ref [3]

Another very important aspect for porous storages may be the filling and blockage of pores of the reservoir by the precipitation of elemental sulphur leading to decrease of the permeability and the production performances (injection/withdraw flow rate). Another effect may be considered: the **impact on the reservoir from aerobic bacteria**, that are able to produce biomaterial that can possibly clock the pores. These phenomena are under investigation.

In an actual case, sulphur precipitations occurred in an UGS at an oxygen content of 40 ppm. After limiting the oxygen content, no more precipitations were observed, which demonstrates the need for low oxygen concentrations in the injected gas [16].

A severe safety issue is the **generation of pyrophoric iron**, as well known for pipelines (specific concern during pigging). Actual cases of pyrophoric iron generation in UGS are known and are raising safety concerns due to the self-ignition potential.

Finally, another negative effect of oxygen injection in aquifer storages is the **impact on the geo-biological equilibria existing at the water/gas contact**. In deep aquifers, several bacteria communities have been identified, that help degrading organic compounds such as BTEX and prevent aquifer contamination [1]. These bacteria are anaerobic organisms; the presence of oxygen could kill them and therefore disrupt this equilibrium.

Nevertheless, there is a growing interest in O_2 addition when H_2 is present to mitigate impacts of H_2, especially in the case of blending with natural gas (from 1 to 20%). Indeed, some authors report from laboratory experiment that O_2 (in the order of 100 to 1000 ppmv) mitigates steel embrittlement caused by H_2 gas molecules. This could be due either (or conjointly) to the formation of H_2 permeation blocking deposit formation (iron oxides for instance) or to competitive adsorption processes between O_2 and H_2 at the steel surface. In any case, the potential protective effect of oxygen has to be effective prior to the addition of hydrogen. This is still to be demonstrated for in-field equipment as all internals are already covered by various deposits (mill, corrosion products, etc.). This observation could be a laboratory artefact as the test coupons are mirror polished for the experiments.

3.2 European gas property standard vs specific situation in the different European countries

Existing European gas quality standard EN 16726 [2] as well as national ones limit the allowable concentrations. The EN 16726 specifies that, at network entry points and interconnection points, the mole fraction of oxygen shall not exceed 0,001 mol% (10 ppm), expressed as a moving 24-hour average. However, where it can be demonstrated that the gas does not flow to installations sensitive to higher levels of oxygen, e.g. underground storage systems, a higher limit of up to 1 % mol (10 000 ppm) may be applied.

Based on recent national information and published data [17], the situation in Europe is presented in Figure 2. The situation in the different countries is diverse, not always clear or published and evolving.

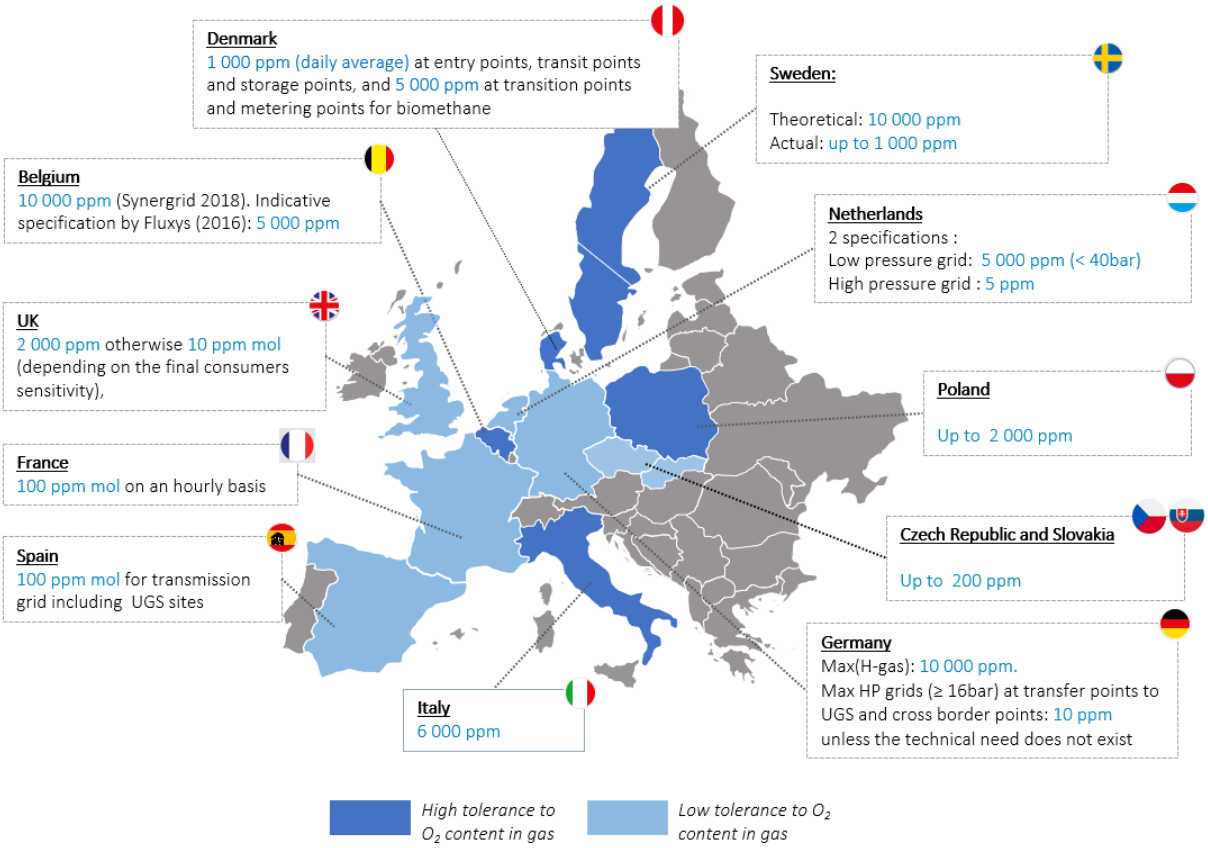

Figure 2: Maximum admissible oxygen content in European gas networks

3.3 Different research programs and studies would provide further knowledge

To increase the knowledge on the topic, several European, national or companies initiatives are undergoing. The Annex (see chapter 5) to this paper presents a quick overview of available results to date (published or communicated by members of MARCOGAZ). Some past experiences are also mentioned.

4. Preliminary conclusions on oxygen acceptance

Biomethane can be injected in the gas grid and thereby **in UGS facilities as exchange gas, equivalent to natural gas, if the usual required natural gas specifications are met.** Storage of biomethane can offer new business opportunities for UGS.

Biomethane may contain a significant oxygen fraction of about 3000 ppm, due to the gas treatment process of biogas, if not sufficiently upgraded (due to cost effectiveness reasons).

Up to now, the oxygen concentration should not create restrictions for the injection of biomethane in the gas grid as the oxygen content could normally be diluted due to mixing with the prevailing base gas in the gas grid. However, as biomethane injection volumes in the networks increase with time thanks to energy transition, dilution effects will not be possible anytime/anywhere. This should not be a major concern to DSO and TSO, but the remaining oxygen fractions may be still too high for the oxygen sensitive gas end-users, such specific O_2 sensitive industries and UGS facilities. Therefore, those O_2 ingresses have to be limited. **The permissible oxygen content is dependent on the specific facility and has to be assessed case by case.**

For some facilities, an oxygen limit of 100 ppm, or even more, is acceptable. In more sensitive storage facilities or oxygen sensitive industries, a permissible oxygen concentration of 10 ppm is required at most.

In general, it is recommended to keep the amount of oxygen in the gas grid and in storage facilities as low as possible, especially in the presence of H_2S and CO_2.

The removal of the oxygen in biomethane may be implemented, if technically and economically feasible:

- **on the biogas plant** - It would need optimization of the gas treatment process and in some instances implementation of specific upgrading processes. On a limited scale, oxygen removal plants are installed in Germany on the grid next to two biogas plants in relation to 210 biogas plants injecting biomethane into the gas grid.
- **on the gas grid** (distribution, transmission and backflow points).
- **at the entrance of the UGS facilities**

The technical and economical feasibility of removal plants at the entrance of UGS facilities is not proved by now, considering the important gas flow to treat and the available processes.

On-going research projects should provide an update of the assessment of the impact of oxygen on UGS facilities, and some may propose treatment processes to lower the oxygen concentration at biogas plants or processes for removal of oxygen. All initiatives and research projects would contribute to the promising greening of the gas infrastructure.

5. ANNEX - Relevant Undergoing Studies And Past Experience

5.1 GERG CEN PNR project on biomethane acceptance.

This is a pre-normative research (PNR) project under CEN/TC 408 supervision which ultimately aims at revising EN 16723 parts 1 & 2. By now, the oxygen level is defined in EN 16726 (H-gas).

This project, funded by the European Union and started in 2018, targets to “remove technical barriers to biomethane injection into the natural gas grids”. It is a four phases project (1, 2a, 2b and 2c). As of December 2021, phase 2b is being executed and phase 2c is to be started mid-2022 and completed by mid-2024. Phase 2b is composed of 4 work packages (WP):

- WP1 is dedicated to siloxane. Its conclusion is that boilers and engines are not significantly impacted by biomethane siloxane.
- WP2 is dedicated to sulphur. H_2S is to be oxidized into SO_2 by combustion. Catalytic converters of vehicles may be impacted and further investigations or tests are needed. Laboratory tests are in progress.
- WP3 deals with H_2 as some biomethane digestion processes may bring some. The focus is on CNG tanks. The literature review has been completed and further laboratory tests are proposed.
- WP4 aims at collecting biomethane gas compositions to build a database and increase the knowledge about the influence of the feedstock and processes, especially cleaning ones, on the compositions. WP4 includes a dedicated task on bio-LNG.

In phase 2c, among other tasks, WP3 would focus on the effect of oxygen both on underground reservoirs and facilities (formation damage, microbial population, sulphur generation, surface process equipment and corrosion tests). Conclusions based on literature review from previous phases (i.e. 1, 2a and 2b) show that storing biomethane in underground storages may increase the risk of bacterial H_2S production by providing nutrients for existing bacterial communities and/or chemical reactions with sulphur species. Conclusions also confirmed that no corrosion risk is to be feared as long as the gas is dry. This is the case in both distribution and transmission networks, but not for UGS where the gas is wet. In the presence of wet gas, as far as O_2 content remains below 10 ppmv, although corrosion risk is confirmed above this threshold, no evidence/indications of expected corrosion rates when above this threshold were reported. This may be obvious as no oxygen was present in the natural gas grid. It is important to point out that these studies reminded that O_2 corrosion risk is proportional to O_2 partial pressure (partial pressure is roughly the concentration multiplied by pressure). And hence pressures in UGS range from 100 to 250 bar, to be compared to distribution and transmission network conditions: the consequence is that partial pressure, in such conditions, is high even if concentration is low.

Core flooding experiments using rock cores from representative reservoirs are to be performed in order to evaluate the impact of biomethane contaminants on the risk of inorganic and biologically formation damages. But case-to-case studies are to be necessary as all geological UGS parameters differ one from the other. Similarly, corrosion experiments with representative concentrations of O_2 / CO_2 / H_2S are needed to better evaluate corrosion rate versus O_2 content. Both experimental approaches are to be tackled during this last phase.

5.2 CEN Sector Forum /TF3 pre-standard work and GERG project

As of December 2021, CEN Sector Forum /TF3 – oxygen - is assessing the effect and implications of oxygen fractions in fuel gases on gas infrastructure and gas applications. The conclusions are expected to support the planned update of the EN 16726. Input from above mentioned GERG project is helpful to complete the task. Preliminary results from TF3 would be available to update EN 16726 (standard on gas quality) even if the GERG project would not be completed.

A literature survey carried out by GL Industrial Services [18] finished in 2019 considered oxygen impurities in UGS reservoirs. The literature review leads to the following conclusions:

- Biomethane does not cause any formation damage mechanisms that are not already present in conventional gas.
- For depleted reservoirs, biomethane could increase the risk of inorganic precipitates (particularly iron compounds) due to the oxygen content. The reports of issues are not well documented. DGMK reports [3] [10] recommend limiting oxygen to 10 ppm based on the effect of sulphur precipitations and on corrosion prevention. It is possible that higher concentrations could be accommodated from a reservoir perspective.
- Bacterial contamination from biomethane could cause formation damage for aquifers and depleted reservoirs due to biofouling. However research has found bacterial contamination in conventional gas storage so the issue is not unique to biomethane.
- Storage of biomethane in caverns should not increase the risk on geological formations.

5.3 RINGS project

The RINGS Project [12] (Research on the Injection of New Gases in Storages) is a pre-normative research program to assess the impact of so-called "new gases", such as biomethane/O_2 and hydrogen in UGS reservoirs. The goal is to rebuild in a HP-HT dedicated triphasic reactor in situ conditions of existing natural gas aquifer storages in laboratory, and identify the main impacts of the injection of new gases on the equilibrium of the storage. Possible biogeochemical processes between reservoir rock, microorganisms, aquifer water and stored gas need to be understood and quantified.

This project, founded by Terέga and Storengy (France), with the participation of SNAM (Italy) and Enagas (Spain) as financial partners, started in 2018. Several research teams from the University of Pau are involved in this project, which includes specialists of microbiology, analytical chemistry, instrumentation and imaging, high pressure process, and geologists.

As of December 2021, two experiments reproducing the storage conditions of a facility located in the South West of France (36 °C, 60 bar, methane with 1% CO_2) were performed at two O_2 concentrations: 10 000 and 100 ppm. The interpretation of the 100 ppm experiment is still in progress.

The experience at 10 000 ppm O_2 is presented in Haddad et al. (2022)[13]. The main results are given below:

- Before O_2 injection:
    - Sulphate was depleted from the liquid phase during the first 130 days of incubation. Biological diversity analysis shown a growth of sulphate-reducers organisms such as Desulfovibrionaceae.
    - Benzene and toluene, hydrocarbons found in traces and known to be biodegradable in storages, were monitored and a decrease of toluene was revealed and associated to the Peptococcaceae family.
- O_2 injection induced:
    - A re-oxidation of sulphates into sulphides
    - The end of the sulphate reducing activity and toluene biodegradation
    - A drastic disappearance of most of the community
- The solid phase analysis, mainly composed of quartz (+ 1% of calcite and kaolinite) did not show any significant morphology/porosity changes.

5.4 CORR-O2 French project

Storengy and Terέga, the two French UGS operators, have launched a research project subcontracted to the UPPA (Université de Pau et des Pays de l'Adour) which aims at defining a relationship between gas O_2 content and corrosion risk (in terms of mechanisms and kinetics) of UGS well tubing's. Using actual aquifer UGS water (two aquifers are to be considered) and steel samples from actual tubing's the experimental set-up would reproduce temperature and pressure conditions of the UGS. Steel coupons are to be exposed to various concentration of O_2 (from 10 to 10 000 ppmv) between 1 month and 6 months. Coupons are to be either in the gas phase, in the liquid or at the interface. When removed the coupons are to be analysed for deposits nature and corrosion rates. The project started in 2021 and should be completed by the beginning of 2023.

In addition, Storengy is to launch corrosion tests specific to salt cavern storages. Indeed, as the inside of the well tubing are partially coated by salt deposits, and considering that due to gas humidity condensation of water is possible on those same surfaces; the presence of O_2 in the gas phase is to generate very corrosive conditions, especially when considering the O_2 partial pressure. Two studies are considered: a first one at low pressures considering many parameters, and the second one reproducing the storages pressure, but with fewer parameters.

5.5 Past experience

Some publications presenting past French in situ experiments give information on oxygen consumption in the subsurface and the impact on the reservoirs and their chemical equilibrium.

Irina Sin, Laurent De Windt, David Dequidt [4] (2021, to be published) reinterprets a 1990' injection of a mixture of nitrogen and oxygen (up to 8%) in the cushion gas of a French aquifer storage. Iron sulphide was oxidized with a fast kinetics, releasing sulphates and lowering pH in water. As a consequence, carbonates dissolve, CO_2 degasses and gypsum precipitates while Oxygen is completely consumed. Batch model is not sufficient to interpret the observation: a coupled model with flow, reaction and transport is necessary.

Colonna *et al.* [5] and Densham et al. [6] demonstrated respectively in 1966 and 1975 that oxygen is consumed in the subsurface (either in situ or in the laboratory in reservoir samples of French aquifer storage reservoirs) according to rapid kinetics (some weeks). The proper chemical mechanism of oxidation was not clear and should be probably reinterpreted.

This rapid consumption of oxygen is to lead to the following situation: during a certain period of time, the withdrawn gas would not contain oxygen, but after this period of time the chemical reactions would no longer happen in the reservoir, the redox level would be changed and the oxygen injected would be withdrawn. The length of this period depends of the amount of certain minerals of the reservoir and the quantity of oxygen injected (rates and concentration).

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