Treatment of the confined air of a spacecraft cabin

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ABSTRACT

A test bench at scale 1/15 has been implemented in order to evaluate the performance of photocatalysis and membrane bioreaction to treat the polluted air of a spacecraft cabin containing 30 mice. Ten pollutants could be generated with low concentration and seven of them could be analysed simultaneously. Formaldehyde was tested alone because of the needed volume for its analysis. First results have shown that photocatalysis process could lead to good efficiency except for methane and formaldehyde. This one is not sufficiently eliminated. A new configuration of photocatalysor allowing higher illumination in order to try to increase formaldehyde elimination will be tested. Membrane bioreactor have to be tested too. Interesting results are expected testing the two processes in series.

1 INTRODUCTION

Indoor Air Quality (IAQ) has been a matter of great interest for several years now. Indeed a very large number of pollutants, among which a lot of organic compounds and especially volatile organic compounds (VOCs), have been detected in residential indoor (Mosqueron and Nedellec, 2004; Edwards *et al.*, 2001). Moreover, as the cabin of a car can be considered to be a part of the living environment (because of the spent time in cars) some authors have also focused on identification of airborne organic compounds in a cabin of a car (Yoshida and Matsunaga, 2006). In the same manner, cabins of spacecrafts are also concerned with indoor air pollution. Improvement of IAQ lies in three kinds of solution:

- removal of, or, at least, reduction in pollution sources,
- ventilation and thus air renewal,
- polluted air treatment.

As far as spacecrafts are concerned, ventilation and air renewal being obviously impossible, indoor air treatment is unavoidable. Since porous materials offer high capacity for VOCs, adsorption is a widely used process. But a non negligible airborne quantity of porous materials can be needed according to the duration of the space flight. This is a major disadvantage since little room is available in spacecraft. Regeneration of porous materials that could be considered would also need significant energy amount that could be prohibitive to space application.

Hence the present study focuses on two other treatment processes: photocatalysis and membrane bioreactor. The objective of the study is to demonstrate the applicability and performance of these processes to the removal of pollutants in a spacecraft containing 30 mice. In the present study there are no outdoor pollutant sources neither sources arisen from human activities. Only pollutants coming from the mice metabolism, from the waste decomposition and from the material emission have then been considered to establish the list of the ten chosen pollutants (Yu and Crump, 1998; Phillips et al., 1999; Ruzsanyi et al., 2005; Sato et al., 2001; Jungbluth et al., 2001; Van Kempen et al., 2001; Mackie et al., 1998; Miller et al., 2001 and 2002; Powers et al., 2005; Schade et al., 1995; Zahn et al., 2001). That's why the list is quite different from lists usually encountered in IAQ studies. For each pollutant, two concentrations have been taken into account : the maximum allowed concentration (MAC) for long term exposure and the short term exposure limit (STEL) which is the maximum acceptable concentration level during 15 minutes. Moreover, daily production rate have been estimated. Table 1 provides the list of the chosen pollutants and the MAC, the STEL and the production rate of each pollutant.

Actually isoprene has been replaced by limonene $(C_{10}H_{15})$ which is easier to handle for performance assessment of the photocatalysor and of the bioreactor.

An adequate airstream in the mice cages is necessary to provide removal of waste towards the filters of the cages. A fraction of this stream is led to the air conditionning system loop which contains the treatment process (as well as HEPA (high efficiency particulate air) filter, charcoal bed, condensing heat exchanger...). Thus the global removal efficiency depends on the proper efficiency of the treatment process and on this fraction of the air stream as well. The higher this fraction is, the lower the proper efficiency of the process can be to maintain the same global removal efficiency. Two values of the air flow have been chosen : $4 \text{ m}^3/\text{h}$ and $10 \text{ m}^3/\text{h}$.

Ta	ble	1.

Compounds	Short Term Exposure Limit (STEL) ppm	Maximum Allowed Concentration (MAC) ppm	Daily production rate (30 mice) ppm	
Ammonia (NH ₃)	35	25	1260	
Methane (CH ₄)	Not relevant	0,5 vol % *	900	
Acetic Acid (CH ₃ COOH)	15	10	10,5	
Propionic Acid (C ₂ H ₅ COOH)	15	10	10,5	
Acetone (CH ₃ COCH ₃)	750	250	10,2	
4-methylphenol (m-cresol) (CH ₃ C ₆ H ₄ OH)	5	2,3	1,7	
Hydrogen Sulfide (H ₂ S)	10	5	1,3	
Isoprene (C ₅ H ₈)	Not available	2	0,8	
Formaldehyde (HCHO)	0,1	0,016	0,3	
Trimethylamine ((CH ₃) ₃ N)	15	2	0,2	

Main gaseous pollutants to be removed and relevant concentrations (P=1atm, T=298K).

* The Lower Explosion Limit of methane is 5 vol %

The global removal efficiency of the two processes will be considered according to two parameters:

- ✓ Capacity to come back from the STEL value to the MAC value within less than 15 minutes
- ✓ Capacity to maintain the pollutant level below the MAC value on the long term taking daily production into account.

2 MATERIALS AND METHODS

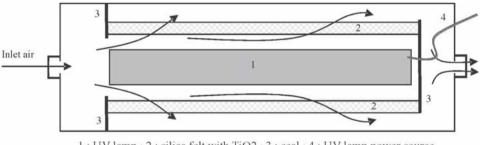
2.1 Photocatalysor

The photocatalysor is cylindrical. The UV lamp stands on the axis with a photocatalyst layer all around. The inside walls of the shell are UV reflective in order to optimise the illumination level. The catalyst layer is a silica felt containing TiO_2 . The polluted air is forced through it (Figure 1).

In order to test different illumination levels the following parameters can be changed :

- UV lamp length : 300 mm, 400 mm, or 900 mm
- UV lamp electrical power : 10, 15, 25 or 30 W
- distance photocatalyst/UV lamp : 1 or 2 cm
- photocatalyst surface.

In the present study, different configurations of the photocatalysor have been tested depending on the chosen values of these parameters.



1 : UV lamp · 2 : silica felt with TiO2 · 3 : seal · 4 : UV lamp power source Figure 1. Shematic representation of the photocatalysor.

2.2 BIOREACTOR

Since we deal with a space application, it is important to ensure the confinement of the biomass and the complete separation of the gaseous phase and the liquid phase containing the biomass. A classical biofilter would not allow this, that's why a membrane bioreactor has been used. It is made up of a shell containing a bundle of 2600 hollow fibres with diameter of 600 μ m. The polluted gaseous stream is forced through the fibres while the liquid phase and the biofilm stand on the outer faces of the fibres, in the shell of the bioreactor. The fibres are microporous hydrophobic membranes. Indeed dense membrane would have been probably too selective regarding the number of pollutants which must diffuse through it. Figure 2 shows the principle

of the microporous membrane bioreactor. The micro-organisms come from liquid sludge of waste water treatment station. During six months the liquid sludge tank has been stirred, aerated and nutrients have been added. The liquid sludge has then been filtered with 100 μ m sieve and introduced in the bioreactor shell.

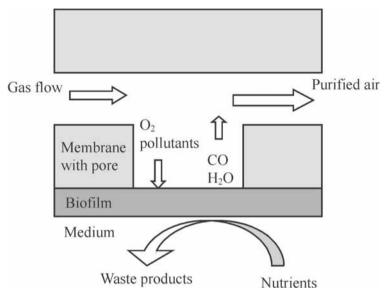


Figure 2. Principle of the microporous membrane bioreactor.

2.3 Analytical methods

 $\rm NH_3$ concentration was determined using a gas sensor (Dräger Sensor $\rm NH_3$ 8101711). H₂S concentration was measured by means of a specific apparatus dedicated to the sulphur compounds analysis, the ChromatoSud airMEDOR which is a gas chromatograph with an electro-chemical detector. Varian 3800 gas chromatograph equipped with a flame ionization detector allows the measurement of CH₄ and acetone. Formaldehyde in the polluted air is sampled by an active-DNPH-silica cartridge (Waters XPosure Sampler WATO47205), extracted with acetonitrile and analysed with a Varian Pro Star HPLC-UV detector. All the other pollutants are analysed by means of a gas chromatograph with spectrometry mass detector (Thermofinnigan Trace GC-MS). A cryogenic preconcentrator is used in order to ensure a good detection. Indeed it removes water and carbon dioxide which would be responsible for interferences in the analyse if not, and concentrates the effluent at the same time. The preconcentration is made using a semi-volatile method. The detection mode of MS is Single Ion Monitoring (SIM) which decreases the detection limit of all compounds.

2.4 EXPERIMENTAL SETUP

The test bench is a closed circuit which total volume is representative of real volume of mice habitats at scale 1/15. So the air flow is $4/15 \text{ m}^3/\text{h}$ or $10/15 \text{ m}^3/\text{h}$. Experiments are performed at atmospheric pressure, at temperature about 20°C and a humidity about 40% (obtained by injection of distilled water by means of a syringe in a heated point). At the start of each experiment, the circuit is first open and a pure air is flushed from a zero air generator (dry air with maximal concentration in carbon compounds about 50 ppby) in the circuit to remove any impurity. Then the gaseous mixture is generated by different means. In order to introduce formaldehyde, trimethylamine and hydrogen sulphur, a calibration gas generator constituted with two permeation ovens is fed by zero air. Methane and ammonia are introduced in the air stream using gas tight syringes. Then a gaseous mixture of acetic and propionic acids, acetone, limonene and m-cresol is generated by introducing these components one by one in a heated air stream using a syringe diver (Harvard Apparatus PHD-2000). These liquids are vaporised in the air stream which is dragged by a pump to a twenty liters balloon flask (in order to represent the mice habitat). Two mass flow meters allow the regulation of the air flow within the two treatment processes (photocatalysor and membrane bioreactor) which are implemented on this loop. Numerous valves allow numerous configurations :

- by-pass of the two processes (this configuration is used when the gas mixture is generated),
- by-pass of only one process,
- both processes in series (photo-bio or bio-photo),
- both processes in parallel.

After the generation phase, the treatment phase begins with the choice of one of the previous configurations. It can be either a STEL to MAC test (Capacity to come back from the STEL value to the MAC value within less than 15 minutes) or a MAC to MAC test (Capacity to maintain the pollutant level below the MAC value on the long term taking daily production into account). Hence in the first case every pollutant has been first generated till the STEL concentration and in the second case till the MAC. At the end of the treatment phase, a sample is made using a tedlar gas sampling bag. Actually formaldehyde must be tested alone since the whole volume of the circuit must be forced through the active-DNPH-silica cartridge to ensure its detection.

It can be noted that the two processes have been tested within a closed circuit to ensure the test bench the more representative as possible of real conditions. This choice led to the following constraints :

- to ensure airtightness, we had to check for leaks regularly and carefully,
- only one analysis could be performed for each test because of the needed sample volume. Moreover, formaldehyde have been tested alone as it has been explained previously.

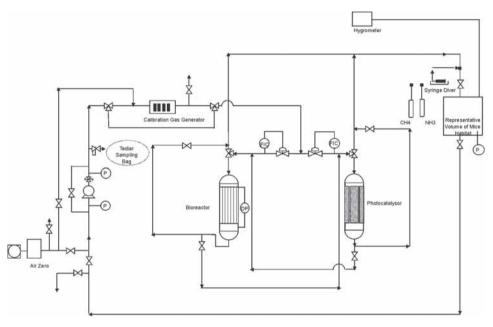


Figure 3. Experimental setup.

3 RESULTS AND DISCUSSION

3.1 CALIBRATION

Numerous problems had to be solved during calibration step.

m-cresol is a quite viscous liquid. So it is introduced in the circuit within acetone. But it seams that during the analysing step, it is difficult to eliminate it from the preconcentrator. Hence the calibration is very difficult, and at present we have eliminated it from our tests in order to ensure that it will not interfere with other analyses. We will reintroduce it at the end of the tests when an optimal process configuration will have been chosen.

The MS detector doesn't ensure a separate detection of acetone and trimethylamine. So tests are performed with a gas mixture containing either acetone or trimethylamine. Thus the number of calibration tests and the number of treatment tests is twice. No calibration is required for H_2S because airMEDOR apparatus is equipped with an internal calibration. No calibration is either required for ammonia.

Calibrations of methane, acetone, limonene, propionic acid and formaldehyde are quite good with linear regression coefficient being 0,9 or more.

Calibrations of acetic acid and trimethylamine are not so good (linear regression coefficient between 0,7 and 0,8). Moreover, the quantification limit of acetic acid is higher that the MAC.

At last, as is has been already said, apparatus can't be properly calibrated for m-cresol.

3.2 Tests

Only the photocatalysor has been tested by now.

At present, three configurations have been used for STEL to MAC tests.

Table 2 provides each pollutant concentration after treatment. STEL (which is initial concentration of each test) and MAC are recalled.

It can be seen that pollutants are eliminated with good efficiency except methane. It is expected that methane will be removed by bioreactor process.

Concentrations of ammonia, propionic acid, acetone, hydrogen sulfide, limonene and trimethylamine are lower than MAC that is an encouraging result. Since the quantification limit of acetic acid is higher than its MAC, no conclusion can be drawn from the previous results for this component. But since it is a smaller molecule than propionic acid which is well eliminated, we can expect that acetic acid is also eliminated.

Formaldehyde is not sufficiently removed : its concentration is still higher than the MAC. A new configuration allowing higher illumination has to be tested.

4 CONCLUSIONS

The following conclusions can be drawn from the results presented in this study.

A test bench at scale 1/15 has been implemented in order to evaluate the performance of photocatalysis and membrane bioreaction to treat the polluted air of the spacecraft cabin containing 30 mice.

Ten pollutants could be generated with low concentration and seven of them could be analysed simultaneously. Only one pollutant could not be analysed (m-cresol). Formaldehyde was tested alone because of the needed volume for its analysis.

First results have shown that photocatalysis process could lead to good efficiency except for methane and formaldehyde. This one is not sufficiently eliminated.

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illumination level (mW)	390	390	394,2		
Air flow	4/15	10/15	4/15		
(m3/h)					
Ammonia	< 2,5	< 2,5	< 2,5	35	25
(NH ₃)					
Methane	8624				
(CH_4)	7062				
	7507	8097	8136	10 000	5000
Acetic Acid	<12,5 (QL)	<12,5 (QL)	<12,5 (QL)	15	10
(CH ₃ COOH)					
Propionic Acid	<6 (QL)	<6 (QL)	<6 (QL)	15	10
(C ₂ H ₅ COOH)					
Acetone	213				
(CH ₃ COCH ₃)	193	210	87	750	250
Hydrogen Sulfide	2,50,6	0,07	0,01	10	5
(H ₂ S)					
Limonene	0,380,38	0,390,44	0,82	4	2
$(C_{10}H_{15})$					
Formaldehyde	0,063	0,031	0,1	0,016	
(HCHO)					
Trimethylamine	0,99	1,74	0,78	15	2
((CH ₃) ₃ N)			0,74		
QL : Quantification Limit		tration after 15 treatment (ppm)		Short Term Exposure Limit (STEL) ppm	Maximum Allowed Concentration (MAC) ppm

Pollutants concentrations after 15 minutes of treatment (P=1atm, T=298K).

Next steps of this study will be :

- test of a new configuration of photocatalysor allowing higher illumination in order to try to increase formaldehyde elimination,
- MAC to MAC tests on the photocatalysor selected configuration after STEL to MAC tests,
- STEL to MAC tests with the membrane bioreactor (methane removal is expected)
- MAC to MAC tests with the membrane bioreactor,
- tests with the two processes in series.

Interesting results are expected testing the two processes in series. Use of photocalatysis as a pre-treatment of bioreaction have already been investigated for wastewater (Mohanty *et al.*, 2005) and more recently for airborne toluene and o-xylene (Moussavi *et al.*, 2006). Indeed, photocatalysis partially oxidized pollutants into more biodegradable intermediates. Moussavi *et al.* have shown that the coupled UV-bioflitration system provided up to 60 % additional contaminant removal compared to the sum of that offered by UV and reference biofilter. Thus such a coupled treatment applied in the present study could lead to reduce the electrical power needed for photocatalysis and to reduce the volume of the bioreactor too.

5 ACKNOWLEDGEMENTS

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