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Recuperation systems for fluorinated gases at the CERN LHC Experiments

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ABSTRACT

Particle detectors at the LHC experiments are very often characterised by large detector volumes and by the need of using very specific gases, some of which are greenhouse gases (GHGs). Given their high Global Warming Potential (GWP) and the increasingly stringent European regulations regarding the use and trade of these gases, CERN is today strongly committed to reduce GHGs emissions from particle detector operation. Different approaches have been adopted for reducing the GHG emissions. To achieve this objective, the CERN Gas Team has developed gas recuperation plants: i.e. systems designed to extract GHGs from the exhaust of gas recirculation systems allowing further re-use and, therefore, reducing drastically GHGs emissions without changing detectors operation conditions. They are industrial-scale systems, each of which relies on different principles for gas separation and purification. Considering the unique gas mixtures used in particle detectors, these recuperation systems have been specifically developed as no industrial apparatus currently exists to address these requirements. Recent developments are concerning plants for recuperation of CF_4, $C_2H_2F_4$ (also called R134a), SF_6 and C_4F_{10}, which are used respectively for Cathode Strip Chambers, (CSCs) Resistive Plate Chambers (RPCs) and Ring-Imaging Cherenkov (RICH) detectors. The separation of fluorinated gases is carried out mainly through membranes, absorbers, or distillation. Two state-of-the-art recuperation systems are briefly described in this paper.

1. Introduction

At the CERN LHC experiments, several gaseous detectors use gas mixtures containing greenhouse gases (GHGs), which guarantee optimal detector performance and long term operation. These gases have a high Global Warming Potential (GWP) and therefore their use has to be limited. This is accomplished by using gas recirculation systems [1]: the gas mixture is collected at the output of the detectors, sent to the surface, cleaned from possible contaminants (usually H_2O or O_2) and afterwards sent back to the detectors. In case of no leaks and no detector constrains, the recirculation can be ~100%. In some cases unfortunately, it is not possible to recycle completely the gas mixture and a fraction of it has to be replenished by exhausting it to the atmosphere. In these cases, a gas recuperation plant can be added to collect the gas at the exhaust and to extract the GHG for its possible re-use [2]. The recuperation plants are industrial-scale systems specifically designed for the separation of the gas mixtures employed for particle detectors as no industrial apparatus currently exists to address these specific needs. Different working principles for gas separation and purification are employed, depending on the gas mixture components:

Membrane separation The separation is due to the difference in thermodynamic activities across the membrane and interacting forces working between membrane material and permeating molecules. The process is driven by several factors: permeability, solubility and diffusivity.

Pressure and thermal swing adsorption The separation of gases is done according to the different interaction of the gas and the absorbent material.

Distillation The separation of two or more compounds is based on the differences in boiling points or volatility.

2. The CF_4 recuperation system

The CMS Cathode Strip Chambers (CSCs) make use of a gas mixture containing 50% CO_2, 40% Ar, 10% CF_4 and, due to detector constrains, around 800 l/h have to be constantly sent to the exhaust line. In this case, it is worth to collect the gas mixture and to recuperate the CF_4 fraction. The recuperation is performed through warm separation in three different phases, making use of gas separation membranes and selective adsorption during pressure swing cycles in different molecular sieves.

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Phase 1 The gas passes through the membrane module and the CO_2 is almost completely removed: the gas mixture at the output of the membrane is ~1% CO_2, 15% Ar, 83% CF_4, 2% N_2.

Phase 2 All the remained CO_2 is absorbed in the Molecular Sieve 4A.

Phase 3a CF_4 is absorbed in the Molecular Sieve 13X, N_2 and Ar are vented and the CF_4 is then recovered through pressure swing adsorption.

Phase 3b CF_4 is extracted and stored in the battery, ready to be re-injected in the mixer.

Currently the recuperation efficiency is ~65% mainly due to the pressure swing adsorption process. No deadtime is present since the recuperation plant has two columns both for Phase 2 and Phase 3. The quality of recuperated CF_4 is very satisfactory (few percentages of Ar and ~10,000 ppm of N_2). During Run 2 and Run 3 the CMS CSC system used 50% CF_4 recuperated and 50% fresh CF_4 in its gas mixture without observing any change in the CSC performance.

3. The $C_2H_2F_4$ recuperation system

The Resistive Plate Chambers (RPCs) make use of a gas mixture containing 95.2% $C_2H_2F_4$, 4.5% iC_4H_{10}, 0.3% SF_6, so it contains a large fraction of GHGs. In this case, they have been validated under ageing test for maximum 90% of recirculation fraction [3] and therefore part of the gas mixture is constantly replenished. In the case of ATLAS and CMS experiments, the fresh gas flow is nowadays between 700 l/h and 1000 l/h. A new custom-made recuperation system has been developed to recuperate the $C_2H_2F_4$ from the gas mixture that otherwise would have been sent to the atmosphere. An intense R&D program was finalised to find a good way to separate the $C_2H_2F_4$ from the iC_4H_{10} as they form an azeotropic gas mixture, i.e. a mixture of liquids whose proportions cannot be altered or changed by simple distillation because intramolecular force of same-species is much higher than the reciprocal attraction. The recuperation plant consists of four independent separation columns, which can work both in parallel or series. Each column is divided into two buffers. Fig. 1 shows a picture of the $C_2H_2F_4$ recuperation plant installed at CMS for the RPC detectors. The recuperation process is divided into several steps.

Phase 1 The first buffer is cooled at −36 °C to remove the N_2 and SF_6 and to liquify the R134a and iC_4H_{10}. The N_2 and SF_6 stay in their vapour phase and they are exhausted.[1]

Phase 2 The second buffer is connected to the first one but it is at 20 °C as it is used to slowly heating the liquified azeotrope to enrich the liquid of R134a and the vapour phase of iC_4H_{10}. The vapour is sent back to the first buffer and it is again liquified. By iterating this process, it is possible to recuperate almost pure $C_2H_2F_4$.

Phase 3 Compression of vapour $C_2H_2F_4$ in liquid storage.

Several tests were performed to find the best working parameters. Indeed the efficiency of the plant and quality of recuperated $C_2H_2F_4$ depend on several factors: the input flow, the chiller temperature, the pressure in the buffers, the velocity of emptying the buffers, etc. For example, by increasing of one degree the chiller temperature, the efficiency is decreasing of about 5% but the iC_4H_{10} contamination is reduced to less than 1000 ppm (instead of 1500 ppm). Therefore a trade off between efficiency and recuperated gas quality has to be made. Nowadays the efficiency of the recuperation plant is around 80%, which is the intrinsic limit due to the azeotropic gas mixture. The quality of the recuperated R134a is excellent as it contains only few hundreds ppm of iC_4H_{10}.

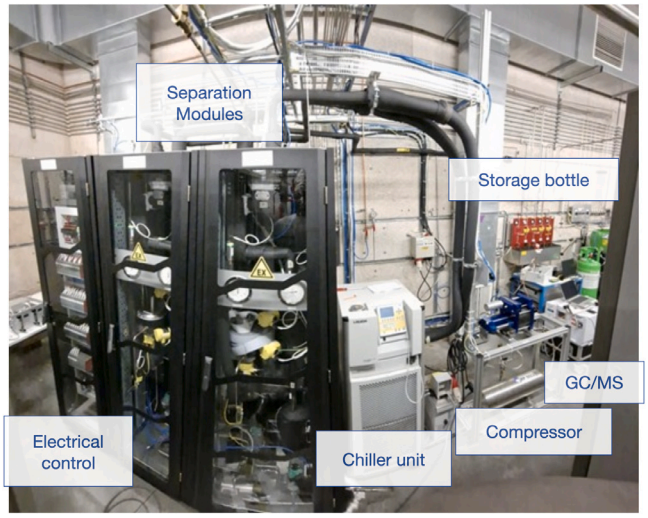

Fig. 1. Picture of the $C_2H_2F_4$ recuperation system installed at CMS for the RPC detectors.

4. Conclusions

The recirculation systems are fundamental to reduce as much as possible GHG emissions in particle detectors when high GWP gases are used. Sometimes it is necessary to add gas recuperation systems, that allow to further reduce the emissions by extracting the GHG from the exhaust gas mixture and to re-use it. At CERN several recuperation systems were developed in the last years using different separation techniques. They provide very good efficiency and they allow to recuperate and re-use greenhouse gases. The emissions and operational costs are lowered and they can mitigate issues in case of gas shortage for the operation of the LHC experiments.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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[1] R&D studies are on-going to try to recuperate the SF_6, that, even if it is a very small fraction of the gas mixture (usually around 0.3%) it has the highest GWP in the world.