

Studies of purification of the Resistive Plate Chamber gas mixture for the Large Hadron Collider experiments

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Abstract—The Resistive Plate Chambers (RPCs) installed as part of the large muon detectors at the Large Hadron Collider (LHC) experiments use a gas mixture of 94.7% $C_2H_2F_4$, 5% iC_4H_{10} and 0.3% SF_6 . Based on economical grounds, the design philosophy of the gas systems for the ATLAS and CMS RPC's foresees to re-circulate the gas mixture in 90-95% closed loop circulation. At the LHC, RPC chambers are operated in a high radiation environment, conditions for which large amount of impurities in the return gas have been observed in earlier studies. They are potentially dangerous for the stable operation of the detectors, the materials in the detector and the gas system. While several purification stages have been foreseen in the present gas systems, chemical reactions between the absorber and the impurities are yet not well understood. Furthermore, the effects on the gas mixture of the foreseen factor 10 increase of luminosity for the LHC upgraded phase should be studied. We present the results of systematic studies of the impurities produced in the RPC chambers irradiated in the high intense photon flux of the CERN Gamma Irradiation Facility (GIF), in order to understand the properties of possible absorbers in a series of orderly tests. The final objective is a proposed optimization of the filters for the LHC closed-loop gas systems and their operation to ensure that RPC chambers can sustain stable, long-term operation under radiation in a cost-effective manner.

I. INTRODUCTION

Resistive Plate Chambers (RPCs) are gaseous parallel-plate detectors that combine high time resolution (~ 1 ns) with good spatial resolution (~ 1 cm). RPCs are extensively employed at the Large Hadron Collider (LHC) experiments as part of their muon trigger systems. They will identify unambiguously the relevant bunch crossing at which the muon tracks are associated, even at the high rate and background expected (up to 1000 Hz/cm²).

RPCs are suitably operated with a three component non-flammable mixture made of 94.7% R134a ($C_2H_2F_4$), 5% iC_4H_{10} and 0.3% SF_6 [1]. Water vapour should be added to the gas mixture in order to maintain a relative humidity of about 45% (i.e. 8000-12000 ppmV) and avoid any changes in the resistivity of the Bakelite electrodes. The large detector volume (about 16 m³ per each experiment) and the use of a relatively expensive gas mixture make a closed-loop circulation system unavoidable (currently the cost with a 95% closed-loop circulation is about 500 €/day).

In the past years, several aging tests have been performed [2] in order to ensure the detector functionalities over a long term period and in conditions similar to the ones foreseen during LHC running. In addition, recently, an increasing attention has been addressed to the study of the gas quality and to the possible formation of *corrosive* compounds during operation. The RPC collaboration went through an extensive measurement campaign at the CERN Gamma Irradiation Facility (GIF) [3] which allowed a simultaneous operation of RPCs in intensive gamma radiation and a high-energy muon beam. More recently a new test in which several RPCs are operated in closed loop mode with cosmic rays is also under way. The aim of these studies is the optimization of the gas purification in the re-circulation system and the set-up of appropriate gas monitoring [4].

II. EXPERIMENTAL SET-UP

The present study was performed using six CMS (Compact Muon Solenoid) double gap RPCs of final design. Each detector has a surface of about 120×205 cm² and 2 mm gas gap. These detectors were part of the standard CMS-RPC production and were certified following the usual quality assurance procedure [5]. Fig. 1 shows a schematic view of the detectors layout.

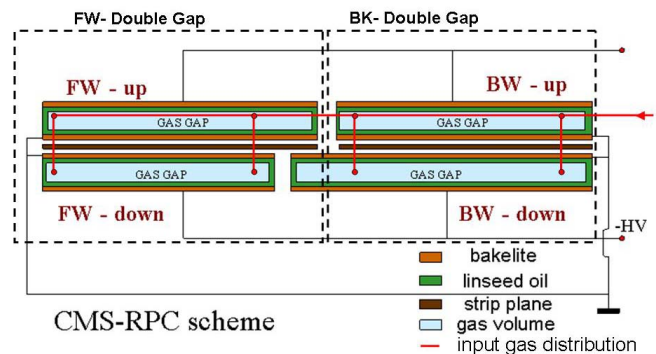


Fig. 1. Schematic view of the RPC detector used during the test: gas volume, double gap position and layout of the gas distribution circuit are indicated.

The intense gamma source located at the CERN Gamma Irradiation Facility has been used in order to simulate the radiation background expected during LHC operation. The GIF is equipped with a 650 GBq ^{137}Cs source. A set of movable lead filters located in front of the source allows to

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reduce the gamma rate to the desiderate value. During the present test, the RPCs were located at about 1.5 m from the ^{137}Cs source and an attenuation factor between 5-10 was used in order to have a maximum counting rate of 300 Hz/cm² and a dose rate of 1 cGy/h.

Previous tests [6] have shown that several extra-components appear in the return gas from RPCs. Chemical analysis performed using gas chromatography, liquid chromatography and Fluoride Ion Selective Electrode (ISE) allowed to monitor the impurities and to correlate the production of hydrofluoric acid with the detector performance. Currently the gas systems of the RPC detectors at the LHC experiments are equipped with a set of purifier cartridges tested and validated during long-term ageing tests carried out at the GIF set-up. The purifiers contain a 24 l cartridge filled with two molecular sieve (5% 5 Å and 95% 3 Å) and a second 24 l cartridge filled with 25% Cu-Zn filter type R12 from BASF, 25% Cu filter type R3-11G from BASF and 50% Ni-Al₂O₃ filter type 6525 from Leuna. If necessary, for the high luminosity LHC run, the volume of the second cartridge can be doubled. Each purifier is equipped with an automatic regeneration system.

The aim of the present study is to identify all the impurities, to evaluate the possible impact on the long term detector performance and, finally, to optimize the mixture purification.

A schematic view of the gas system used during the test is shown in Fig. 2. The heavily irradiated detectors are mainly used to generate a large concentration of the typical RPC impurities. After being recompressed, the exhausted mixture is sent to the filters rack containing several cartridges in order to systematically study the filtering capacity and the long-term purifying efficiency of a set of cleaning agents under test.

The filters rack can host up to six different cartridges. The return gas mixture from the RPCs is divided into six channels by mean of flow regulators also used to measure the gas volume flowing through each cleaning agent. Several connections before and after each cartridge allow to both sample the gas mixture and set-up sequence of different filters.

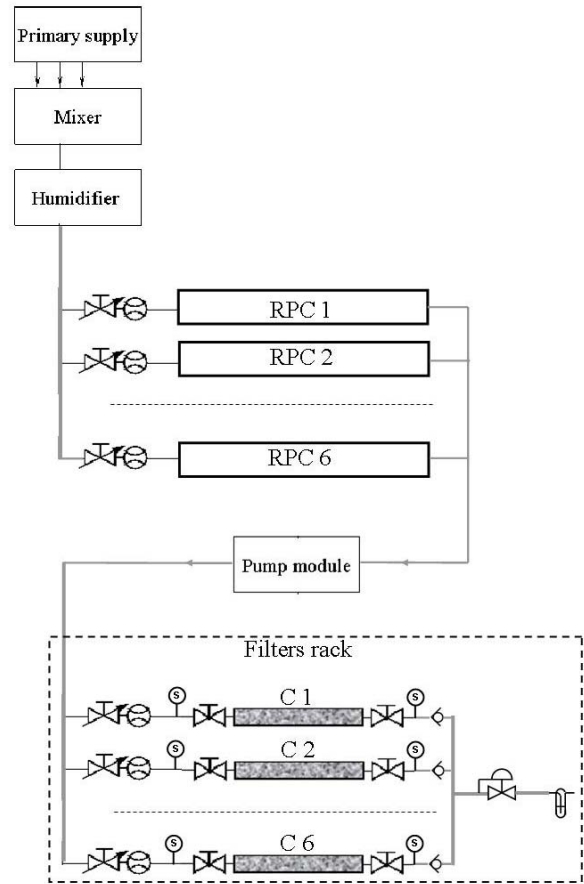


Fig. 2. Schematic view of the RPC gas system used during the test. The RPCs are used to generate the typical impurities in the exhausted gas. After the detectors, the mixture is collected and sent to a filters rack containing several independent cartridges (C1,..C6) allowing a parallel and independent study of different cleaning agents. Several sampling point ("S") are also available.

A network of several sensors (temperature, relative humidity and pressure) monitors environmental conditions and the gas mixture. The gas mixture is prepared in the so-called Mixer module by means of electronic mass-flow controllers (MFC). The gas composition is continuously monitored by recording the reading of each MFC. All these information, together with the current drawn by each chamber gap, are recorded by the control system in order to analyze data and react promptly in case of anomalies.

III. FLUORIDE PRODUCTION STUDY

Fluoride ions, largely produced inside the gas volume during RPC operation (even in presence of cosmic rays only), are very reactive impurities. A detailed study and a description of the different analytical techniques can be found in [6]. During the present test the use of a double channel ISE station (Fig. 3) allows the simultaneous measurement of fluoride ions concentration in different detectors or different gas sampling points (i.e. before and after a selected cleaning agent).

The measurement of the F^- production rate was performed recording the increase of concentration in a well defined volume of water sample contained in a bubbler connected to the gas sampling point. Fig. 4 shows a typical result for two different detectors. For each RPC, the change of slope between the operation at 9.4 kV and 9.6 kV is clearly noticeable, as well as the very low accumulation even with the detectors turned off (most probably due to the gas replacement in the gap and also to the cleaning effect produced by the gas flow).



Fig. 3. Picture of the Hanna double channel analysis station used for the measurement of the fluoride production. The two bubblers containing the water used to trap the F^- ions present in the RPCs exhaust mixture are visible.

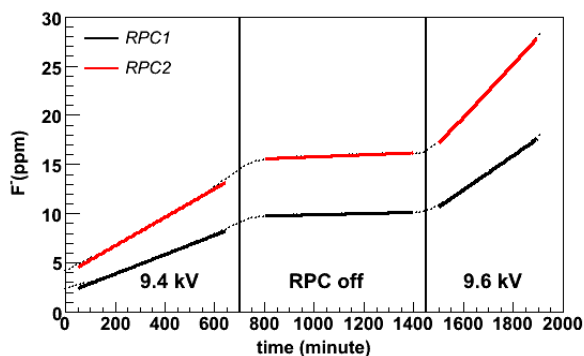


Fig. 4. F^- ions concentration as a function of time in the two bubblers connected to different RPC detectors. The change in the accumulation rate between the period in which detectors are operated at 9.4 kV and 9.6 kV is clearly visible. A very low accumulation is also measured when the RPCs are off; this can be attributed to both a gas replacement inside the gas volume and a cleaning effect of the internal electrode surface produced by the gas flow.

In order to find a possible way to minimize the F^- production at a given chamber efficiency without changing significantly the ratios of gases in the gas mixture, the detectors were operated varying the concentration of iC_4H_{10} between 3.5 and 7.0% and the SF_6 from 0.15 to 0.60 %. Preliminary results indicate that no clear benefit is achieved (Fig. 5), especially for detectors operated in double gap mode (i.e. lower high voltage applied for equivalent efficiency with respect to the single gap mode).

The F^- absorption capacity in the Molecules Sieves 5 Å and 3 Å has been studied. Fig. 6 shows how F^- are effectively filtered with the combination of the two Molecular Sieve

agents currently in use. Similar tests are being performed on all the other cleaning agents.

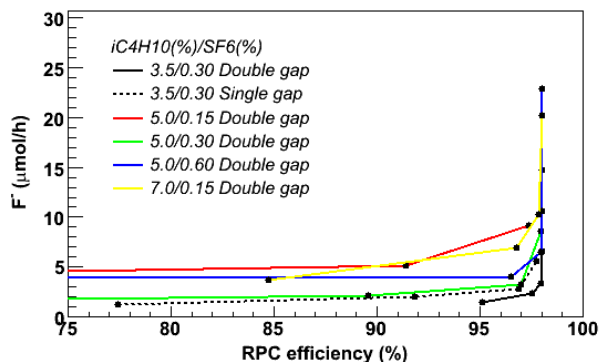


Fig. 5. Production of F^- ions as a function of the RPC efficiency for several gas mixtures containing slightly different concentrations of iC_4H_{10} and SF_6 . Further studies are underway in order to eventually identify the mixture producing the lower amount of F^- at full detector efficiency.

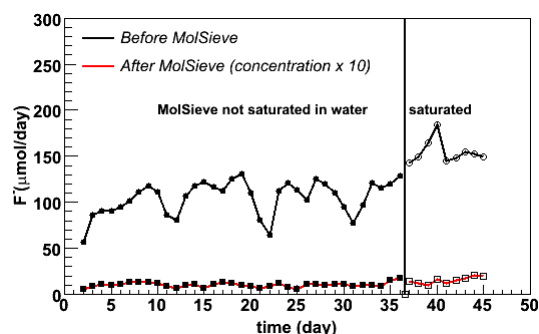


Fig. 6. Absorption of F^- ions in the current configuration of molecular sieve filters. The F^- concentration is decreased by more than a factor 100 after the filters.

IV. OTHER GASEOUS PRODUCTS

In addition to the important amount of fluoride ions measured at the exhaust of RPCs, many other molecules are produced as a result of the breaking of the main mixture components by means of ionization, UV photons, charge multiplication and the subsequent recombination/re-arrangement of the fragments. The following study was performed using a micro Gas Chromatograph (microGC Agilent 3000) coupled with a mass spectrometer detector (Agilent MDS 5975). The microGC is equipped with three modules each containing a column with precise separation characteristics. The first module uses an OV1 polar column mainly indicated for separation of relatively heavy hydrocarbons (from C_4 to C_{12}). The second module contains a PoraPlotU column suitable for separation of hydrocarbons in the range C_1 to C_4 , CO_2 , CH_4 , H_2O , H_2S , SO_2 , N_2O . The last module uses a MolSieve 5 Å column. It separates light components and noble gases (i.e. Ne, H_2 , O_2 , Ar, N_2 , CH_4 , CO). Each microGC module is a complete chromatograph with micro injector, own analytical column and Thermal Conductivity Detector (TCD). One of the microGC outputs can be selected and coupled to the mass spectrometer, making possible the identification of each separated compound. Most

compounds are detected by the analysis station in a concentration scale below 10 ppm with a very good linearity of the answer.

Fig. 7 shows the chromatogram of the fresh RPCs gas mixture as it enters the chambers. The main four components are identified. Fig. 8 shows a detail of a chromatogram of the gas exhausted by the RPCs. In the retention time interval between 20 and 65 s impurities are detected and clearly visible as extra peaks. The extra-components are mainly other Freon and hydrocarbons molecules; each signal is associated with the corresponding molecular formula; only signal (2) has not been identified possibly due to a very low concentration.

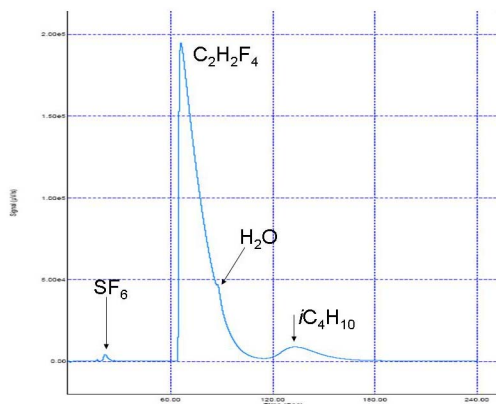


Fig. 7. Gas chromatogram of the gas mixture as it enters the RPCs. The chromatogram has been obtained using a PoraPlotU column and a TDC detector. The main components ($C_2H_2F_4$, iC_4H_{10} , SF_6 and H_2O) are visible.

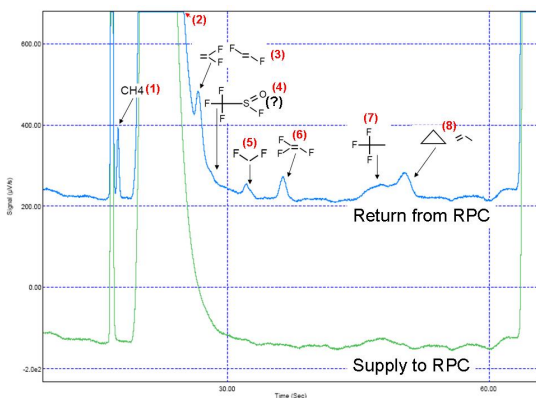


Fig. 8. Extra-components found in the return mixture from RPC detectors. Only the detail of retention time between 20 and 65 s is shown since all the impurities are concentrated in this time interval. The chromatogram obtained for the supply gas mixture is also shown as reference.

During the first operation of some purifier agents (measured to be up to 10-100 volume changes) the main components of the RPC gas mixture can be trapped producing a temporary alteration of the gas mixture composition. This is very relevant for the operation of closed-loop gas systems, where the purified gas is sent back to the loop. In order to solve the problem a so-called *conditioning phase* has been introduced in which the purifier is flushed with a defined equivalent volume of process gas before it is sent into the closed-loop. Fig. 9 shows as an example the effect observed on the iC_4H_{10} for two

of the tested molecular sieves. The molecular sieve 5 Å strongly absorbs $C_2H_2F_4$ during the first 30 volume changes; therefore, during that period of time the concentration of iC_4H_{10} in the gas mixture becomes higher with respect to the nominal value.

For some other purifiers (Fig. 10 shows as example the result for the $Ni-Al_2O_3$ catalyst) we have observed extra-signals appearing in the gas chromatograms, signals that are strongly enhanced after a first phase of operation and then disappear again. Therefore the conditioning phase of some purifiers need to also include also this undesirable effect.

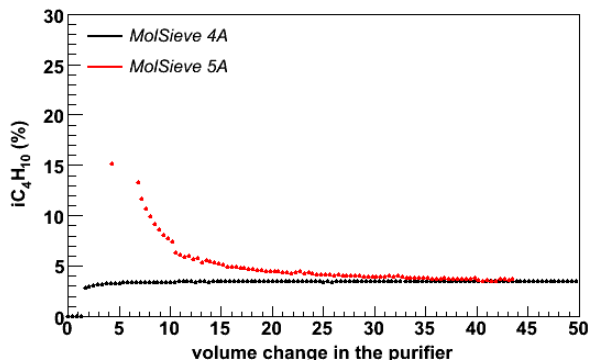


Fig. 9. iC_4H_{10} concentration in the mixture when it is filtered by two different molecular sieve (4 Å and 5 Å) as a function of the gas volume change in the purifiers. The amount of iC_4H_{10} in the gas mixture filtered by the 5 Å sieve is increased as a result of the strong absorption of $C_2H_2F_4$ in the first operation of that filter, till saturation is reached.

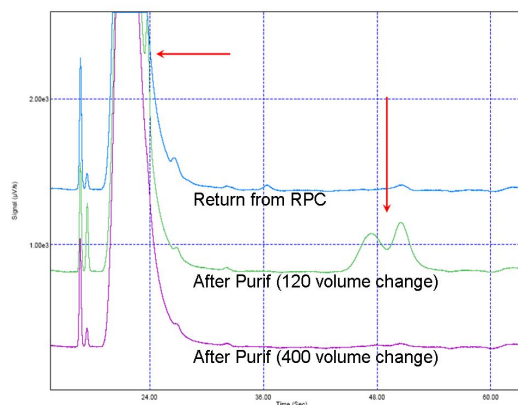


Fig. 10. Two different extra-components (indicated with arrows) appear in the mixture analyzed after the $Ni-Al_2O_3$ catalyst. Their concentration reaches the maximum after about 120 volume changes and then it decreases again.

Table 1 summarizes the results obtained for 7 cleaning agents. A preliminary estimation of the conditioning phase (column 1), the nominal main component filtered by each cleaning agents (column 2) and the effect on the extra-components detected in the exhausted gas of powered RPCs are reported. The impurity labelled with 2 since it is produced by the interaction of the exhausted gas with only some purifiers, is indicated as “Not present” for the other cases. The best combination of purifiers will be chosen on the basis of this table, eventually completed with new filtering elements under test. The next phase involves the long-term validation of a selected combination of purifiers with a set of RPCs

irradiated at the GIF facility. The goal is to reach stable and efficient operation of the LHC RPCs and closed-loop gas system for an amount of time equivalent to 10 LHC years in terms of accumulated charge by the detectors.

TABLE I
SUMMARY OF THE CHARACTERISTICS OF THE CLEANING AGENTS TESTED AND THEIR EFFECTS ON EACH EXTRA-COMPONENT FOUND IN THE GAS CHROMATOGRAPH ANALYSIS.

	Conditioning (Purifier volume change)	Main component filtered	1-CH ₄	2-(?)	3-C ₂ H ₂ F ₂	4-CF ₄ SO(?)	5-CH ₂ F ₂	6-C ₂ HF ₃	7-C ₂ H ₃ F ₃	8-C ₃ H ₆
MS3A	3	H ₂ O	Unchanged	Not present	Unchanged	Removed	Unchanged	Unchanged	Unchanged	Unchanged
MS4A	10	H ₂ O	Unchanged	Not present	Unchanged	Removed	Removed	Removed	Removed	Removed
MS5A	50	H ₂ O	Unchanged	Not present	Back after 1000 vol change	Removed	Removed	Removed	Removed	Removed
Cu (Basf R11)	20	O ₂	Unchanged	Not present	Unchanged	Removed	Unchanged	Removed	Removed	~Removed
Cu/Zn (Basf R12)	20	O ₂	Unchanged	Present up to 400 vol change	Enhanced after 900 vol change	Removed	Unchanged	Removed	Removed	Present up to 450 vol change
Ni Al ₂ O ₃ (Leuna)	15	O ₂	Unchanged	Not present	Unchanged	Removed	Unchanged	Removed	Present up to 150 vol change	Present up to 150 vol change
Ni SiO ₂ (Leuna)	15	O ₂	Unchanged	Present up to 400 vol change	Unchanged	Removed	Unchanged	Removed	Unchanged	Unchanged

V. CONCLUSIONS

Due to the large detector volume and the use of a relatively expensive gas mixture, the LHC-RPC gas systems have been designed to operate in closed-loop mode. In order to ensure a safe operation together with a re-circulation factor as close as possible to 95% the systems have been equipped with a purifier module containing four cleaning agents (molecular sieves 5 Å and 3 Å, Cu-Zn filter type R12 from BASF, Cu filter type R3-11G from BASF and Ni-Al₂O₃ catalyst type 6525 from Leuna).

The present study focuses on the identification of the extra-components produced in the mixture during RPC operation and the optimization of the filtering stages for closed-loop gas systems. Preliminary results are summarized in Table 1. The impurities detected in the RPC chambers and identified by mean of gas chromatography are mainly other Freon and hydrocarbons. The findings of this study will permit the optimization of the gas mixture purification for the future operation of RPC detectors during the LHC high luminosity phase.

Several gas mixtures, with slightly different concentration of *i*C₄H₁₀ and SF₆, have been tested in order to evaluate possible important effects on the production of hydrofluoric acid during RPC operation. Preliminary results have been presented, however further investigation are on going.

The F⁻ absorption has been tested for the combination of molecular sieves currently in use. Test results show a clear absorption of the F⁻ with a concentration reduced by more than a factor 100 after the purifier. Similar tests will be performed on the other cleaning agents.

ACKNOWLEDGMENT

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