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1   INFN Sezione di Bari, Via E. Orabona 4, 70125 Bari, Italy
2   Dipartimento Interateneo di Fisica, Università di Bari, Via Amendola 173, 70125 Bari, Italy
3   Dipartimento di Fisica, Universitá degli Studi di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy
4   INFN Sezione di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy
5   Department of Physics, Helwan University, Helwan Sharkeya, Helwan 4037120, Egypt
6   Dipartimento di Chimica, Universitá degli Studi di Pavia, Corso Strada Nuova 65, 27100 Pavia, Italy
7   Instituto de Física, Universidade do Estado do Rio de Janeiro, R. Sao Francisco Xavier, 524-Maracaná, Rio de Janeiro 20550-013, RJ, Brazil
8   INFN-Laboratori Nazionali di Frascati, Via Enrico Fermi 54, 00044 Frascati, Italy
9   CERN, Espl. des Particules 1, 1211 Meyrin, Switzerland
10 INFN Sezione di Bologna, Viale C. Berti Pichat 4/2, 40127 Bologna, Italy
11 INFN Sezione di Napoli, Complesso Universitario di Monte S. Angelo ed., 6, Via Cintia, 80126 Naples, Italy
12 Laboratoire de Physique Corpusculaire, Clermont Universitè, Universitè Blaise Pascal, CNRS/IN2P3, BP 10448, 63000 Clermont-Ferrand, France
13 Laboratory for Nuclear Science, Massachusetts Institute of Technology, 77 Massachusetts Ave., Cambridge, MA 02139, USA
14 Dipartimento di Fisica, Universitá degli Studi di Torino, Via P. Giuria 1, 10126 Turin, Italy
15 INFN Sezione di Torino, Via P. Giuria 1, 10126 Turin, Italy
16 Dipartimento Interateneo di Fisica, Politecnico di Bari, Via Amendola 173, 70125 Bari, Italy
17 Département de Physique, Université Paris-Saclay, 91190 Paris, France
18 Department of Physics, Korea University, Seoul 02841, Republic of Korea
19 Department de Fisica y Matematicas, Universidad Iberoamericana, Mexico City 01210, Mexico
20 INFN Sezione di Pavia, Via A. Bassi 6, 27100 Pavia, Italy
21 Department of Physics and Astronomy, Ghent University, Proeftuinstraat 86, 9000 Ghent, Belgium
22 Dipartimento di Ingegneria Chimica Materiali Ambiente, Universitá di Roma "Sapienza", Piazzale Aldo Moro 5, 00185 Rome, Italy
23 Inter-University Institute for High Energies (IIHE), Vrije Universiteit Brussel, Pleinlaan 2, 1050 Brussels, Belgium
24 Institut de Physique des 2 Infinis, Universitè Claude Bernard Lyon I, 43 Bd. du 11 Novembre 1918, 69100 Villeurbanne, France
*   Correspondence: liliana.congedo@ba.infn.it
†   This paper is based on the talk at the 13th International Conference on New Frontiers in Physics (ICNFP 2024), Crete, Greece, 26 August–4 September 2024.

Abstract: In high-energy physics, resistive plate chamber (RPC) detectors operating in avalanche mode make use of a high-performance gas mixture. Its main component, Tetrafluoroethane ($C_2H_2F_4$), is classified as a fluorinated greenhouse gas. The RPC EcoGas@GIF++ collaboration is pursuing an intensive R&D on new gas mixtures for RPCs to explore

eco-friendly alternatives complying with recent European regulations. The performance of different RPC detectors has been evaluated at the CERN Gamma Irradiation Facility with Tetrafluoropropene ($C_3H_2F_4$)-CO_2-based gas mixtures. A long-term ageing test campaign was launched in 2022, and since 2023, systematic long-term performance studies have been carried out thanks to dedicated beam tests. The results of these studies are discussed together with their future perspectives.

Keywords: high-energy particle physics; resistive plate chamber; eco-friendly gas mixtures

1. Introduction

Resistive plate chambers (RPCs) [1] are gaseous detectors composed of two parallel planar electrodes made of a resistive material, such as Bakelite with resistivity in the 10^{10}–10^{12} $\Omega \cdot cm$ range, separated by a gap with a typical thickness of 2 mm. Thanks to their low cost per unit area, ease of construction, time resolution at the order of 1 ns and efficiency at the level of 98%, these detectors have been employed in several high-energy physics experiments, such as LHC [2–4], for triggering and particle identification purposes. They have been typically operated in avalanche mode with a "standard gas mixture" providing high detection efficiency based on $C_2H_2F_4$ (>90%), known also as R134a, with the addition of a few percentages of iC_4H_{10} (<10%) and SF_6 (<1%), thus enhancing the quenching properties and electronegativity of the gas mixture.

Two of the standard mixture components, i.e., $C_2H_2F_4$ and SF_6, are fluorinated greenhouse gases (F-gases) with high global warming potential (GWP), measuring the contribution of a gas to the greenhouse effect with respect to an equivalent mass of CO_2. The use of these F-gases has been recently limited by European regulation [5] and consequently also by CERN [6]. This framework has motivated intense research activity with the goal of searching for new eco-friendly gas mixtures for RPCs guaranteeing detector performance comparable with the standard gas mixture. To achieve this aim, RPC communities from different experiments (ALICE, ATLAS, CERN Gas team, CMS, LHCb/SHiP) joined efforts, sharing knowledge and manpower within the RPC ECOgas@GIF++ collaboration.

The first attempts of the researchers were concentrated on the replacement of the main component of the standard gas mixture, i.e., R134a with GWP as high as 1430. This gas has been replaced in industrial applications with gases of the HydroFluoro-Olefins (HFOs) family, such as HFO-1234ze ($C_3H_2F_4$), a nonflammable gas with a chemical formula similar to R134a and GWP~6, which in the following sections will simply be referred to as HFO. New gas mixtures based on HFO diluted with a further gas component reducing the RPC operating voltage up to values compatible with the high voltage (HV) systems currently employed at the LHC experiments have been studied. Among the different alternative gas mixtures investigated (see for instance [7–11]), the first tests suggested to focus efforts on the study of three promising eco-gas candidates based on HFO and CO_2, which will be referred to in the following sections as ECO1 (45% HFO/50% CO_2/4% iC_4H_{10}/1% SF_6), ECO2 (35% HFO/60% CO_2/4% iC_4H_{10}/1% SF_6) and ECO3 (25% HFO/69% CO_2/5% iC_4H_{10}/1% SF_6), with GWP reduced by about 1/3 with respect to the standard gas. In order to validate these mixtures, dedicated tests have been performed at the CERN Gamma Irradiation Facility (GIF++) [12], where a radioactive source allows the testing of long-term RPC operation at the high-luminosity LHC irradiation conditions. The setup and methods used for this study are reported in Section 2, while the main results are presented in Section 3.

2. Materials and Methods

The long-term ageing studies of RPCs operating with eco-friendly gas mixtures are carried out by the collaboration at the CERN GIF++ (Figure 1) equipped with a 12.5 TBq ^{137}Cs source providing a high-radiation environment [13]. A system of adjustable absorption filters (ABS) is installed in front of the source in order to attenuate the radiation field, leading to 27 possible irradiation intensities. Moreover, during dedicated periods, a high-energy (~150 GeV) muon beam from the secondary SPS H4 beam line is available to measure the muon detection efficiency of RPCs.
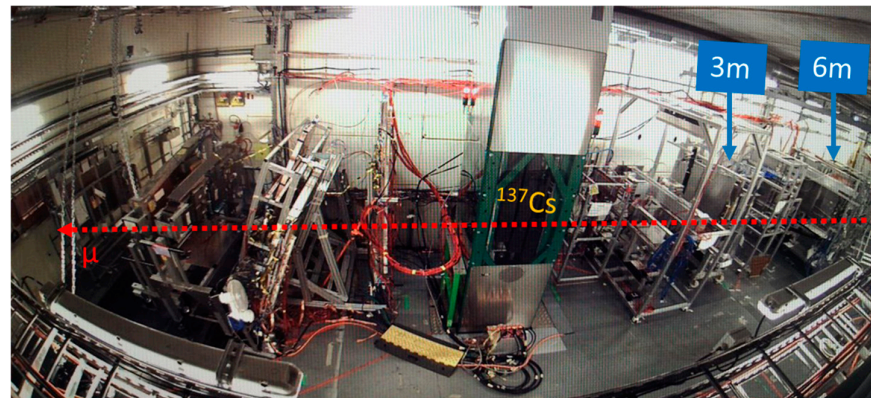

Figure 1. Overview of the GIF++ experimental area.

Two mechanical supports, hosting five RPCs under testing and two scintillators for triggering purposes during beam tests, have been installed at the GIF++ by the collaboration at distances of about 3 m and 6 m from the source, respectively. The RPC chambers have different characteristics, namely size, number of gaps, gap thickness and readout electronics, as summarized in Table 1. All the RPCs are single-gap and rectangular in shape, except for the CMS detector, which is a trapezoidal double-gap. The ALICE and LHCb/SHiP RPCs are readout in (x,y) directions by two planes of perpendicular strips, while the other detectors are equipped with a single strip plane. Signals readout from ALICE, CMS and LHCb/SHiP RPCs, are amplified and discriminated by means of custom front-end (FE) electronics (CMS uses the board mounted at the experiment [14] while ALICE and LHCb/SHiP use FEERIC [15]) transmitting data to a Time to Digital Converter (TDC CAEN mod. V1190A). Instead, digitizers (CAEN mod. V1730 and V1742) are used to acquire the waveforms directly from the strip planes of the ATLAS and CERN EP-DT chambers. Further details about the setup used can be found in [16].

Table 1. Main characteristics of the RPC detectors under test.

RPC	Dimension (cm × cm)	Gas Gap	Readout	Electronics
ALICE	50 × 50	single—2 mm	2D—32 strips	FEERIC + TDC
ATLAS	55 × 10	single—2 mm	1D—1 strip	Digitizer
CMS	(41.5 ÷ 23.9) × 100.5	double—2 mm	1D—128 strips	CMS FE + TDC
EP-DT	100 × 70	single—2 mm	1D—7 strips	Digitizer
LHCb/SHiP	100 × 70	single—1.6 mm	2D—64 strips	FEERIC + TDC

The gas for RPCs is provided by a mixer, allowing for mixing up to four gas components humidified (relative humidity is set to 40%) and distributed to all the detectors. The gas flow is controlled by a dedicated flowmeter and kept stable for each detector. The gas mixtures tested at GIF++ by the collaboration are ECO1, ECO2 and ECO3, and in order to

compare the performance observed with these mixtures, measurements with standard gas (STD) are also performed and used as a reference.

In order to investigate ageing processes, each detector is exposed to a radiation dose of a few mGy/h, corresponding to a background rate of several hundreds of Hz/cm^2 and maintained at a fixed HV, referred to as irradiation voltage, suitably chosen for each chamber in order to limit the working currents. The applied HVs and the absorbed currents are monitored every 30 s and the measured values are stored in a dedicated database. Moreover, once per week the GIF++ source is shielded (in the so-called source off condition), and the currents absorbed without irradiation, namely the dark currents, are measured as a function of the HVs. This operation has two aims: it is used both to estimate the charge integrated by the RPCs during the ageing test and to monitor the dark current density measured at the RPC operating voltage over time.

Figure 2 shows an example of a dark current scan for the EPDT RPC. For low HV values (<5 kV), the current shows a linear increase and therefore it is referred to as the Ohmic current. At these HVs, the multiplication processes in the gas are negligible and therefore the measured Ohmic current is not flowing through the gas, but through other conductive paths in the detector, such as the electrodes, the spacers, etc. Therefore, the current flowing through the gas at the irradiation voltage (I_{gas}) is calculated by subtracting, from the measured current, the Ohmic contribution at this HV, obtained from a linear interpolation of the Ohmic currents as shown in Figure 2. The charge integrated by each RPC is thus calculated from the value of the I_{gas} and reported as a function of the time in Figure 3. After about two years of the irradiation campaign, the charge integrated by the RPCs under testing ranges between 100 mC/cm^2 and 250 mC/cm^2, depending both on the distance of each detector from the GIF++ source and on the specific RPC efficiency value at the irradiation voltage.
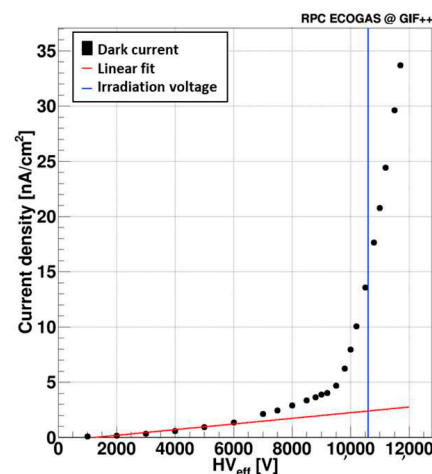

Figure 2. Example of dark current scan performed for the EPDT RPC.

Beyond the measurements of the dark and absorbed currents, in order to investigate the possible ageing effects for RPCs, detector performances should be monitored. This is carried out by the collaboration during the dedicated beam tests. In particular, before the irradiation campaign, which began in August 2022, the performance of the RPCs under testing had been assessed during the 2022 test beam (baseline). The results of measurements performed during 2023 and 2024 are being compared with the baseline.

A role on the RPC ageing effects could be played by the production of impurities (e.g., the HF acid) in the gas volume during detector operation. These pollutants could damage the inner surface of the RPC electrodes. Therefore, in order to check for possible signs of deterioration, the electrode resistivity is also monitored during the ageing test through

dedicated measurements performed with Argon. The gas gaps are thus filled with pure Argon, allowing for the generation of a discharge between electrodes, even applying the HVs of a few kVs. In these conditions, the only contribution to the RPC resistance is due to electrodes, and their resistivity is thus obtained through a linear fit on the plot of the currents as a function of the applied HVs.
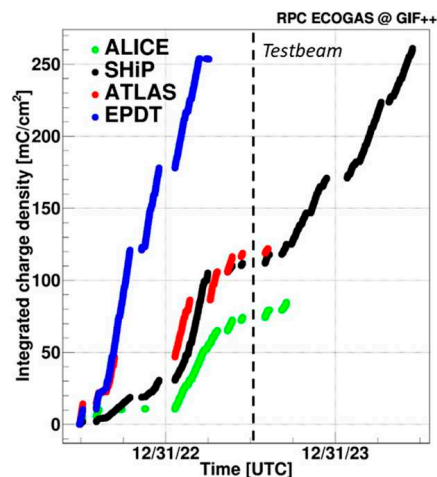

Figure 3. Charge integrated by four RPCs under testing during the ageing campaign as function of time.

3. Results

The first eco-gas mixture tested by the collaboration at GIF++ was ECO1. This mixture showed a large operating voltage with respect to the standard gas, and after a few months of the irradiation campaign with a charge integrated by RPCs of about 10 mC/cm^2, the currents absorbed were continuously increasing with time, and signs of current instabilities were observed [17]. Therefore, the collaboration decided to investigate the RPC performance with the other eco-gas mixture candidates, namely ECO2 and ECO3, with an increased CO_2/HFO fraction. These gas mixtures were tested at different irradiation conditions, and the results were compared with the measurements performed with the standard gas. A detailed description of the results is reported in [16,18–20].

Figure 4 shows, for instance, the efficiency curves and the current densities measured with one RPC (the EPDT chamber) with the three mixtures during a test beam before the irradiation campaign. The applied HV (HV$_{app}$) was corrected for temperature and atmospheric pressure, in order to take into account that at a high temperature (T) and/or lower pressure (p) values, the gas density decreased. Therefore, the effective HV (HV$_{eff}$) applied to the detector was calculated as:

$$HV_{eff} = HV_{app} \frac{p_0 T}{T_0 p} \tag{1}$$

where T_0 and p_0 are the average temperature and pressure measured at the GIF++ bunker, namely 293.15 K and 990 hPa, respectively. The efficiency curves were fitted with a sigmoid function expressed by:

$$\varepsilon = \frac{\varepsilon_{max}}{1 + e^{-\beta(HV_{eff} - HV_{50})}} \tag{2}$$

where ε_{max} is the asymptotic efficiency, HV$_{50}$ is the voltage at 50% of the maximum efficiency and β is the steepness of the curve. The RPC working point (WP), corresponding to the operating HV, is defined as the voltage at the curve knee, namely at the 95% of ε_{max}, increased by 150 V. The WP measured at the fixed irradiation condition with the eco-gas mixtures was increased by a maximum of about 1 kV with respect to the standard gas and

was reduced by enhancing the CO_2/HFO content in the gas mixture, as for example shown for ECO3 if compared with ECO2. The WP values observed with these eco-gases should not constitute an issue for the existing setups at LHC.
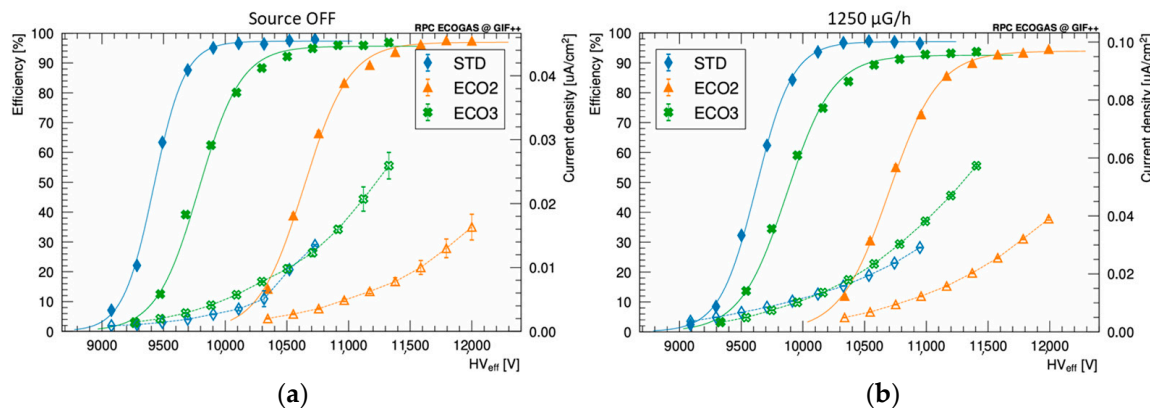

Figure 4. (**a**) Efficiency and current density measured for the EPDT RPC before the irradiation campaign as function of the effective HV without source; (**b**) efficiency and current density measured for the EPDT RPC before the irradiation campaign as function of the effective HV with a background dose of about 1250 μGy/h.

The plateau efficiencies obtained without irradiation were comparable for the three gas mixtures (Figure 4a) and at the level of 97%. With an irradiation background of about 1250 μSv/h (Figure 4b), the maximum efficiency reached by the RPC was slightly decreased with respect to the source-off case, and this reduction was enhanced for the eco-gas mixtures. The plateau efficiency in Figure 4b measured with eco-gases was comparable within a few percentages with the standard gas case, without significant differences between ECO2 and ECO3. Instead, the current density at WP increased when passing from STD, to ECO2 and ECO3, namely increasing the CO_2/HFO fraction, and reached a maximum value being approximately twice the standard with ECO3. This effect was due to a correspondent increase in the fraction of events with large charge content for eco-gases [16]. Since the observed increase in the current and charge could enhance the production of impurities leading to ageing effects, long-term ageing studies with eco-gas mixtures are required. The results obtained during the ageing campaign of RPCs flushed with the ECO2 mixture are discussed in the following section.

Ageing Study

The ageing study of RPCs operated with eco-gas mixtures was based on three different activities: the monitoring of dark currents, the resistivity measurements and the study of RPC performance during beam tests after the irradiation campaign. The dark current measured for one RPC is reported in Figure 5a as a function of the integrated charge. It was approximately stable, up to charge values at the level of about 100 mC/cm^2. For larger integrated charges, the current increased and the appearance of instabilities was spotted.

Also, the Ohmic current increased correspondingly, and this behavior suggests a possible decrease in the electrode resistivity with time, since the two quantities are dependent according to Ohm's law. In order to verify this hypothesis, the electrode resistivity was measured with Argon and the values obtained are reported in Figure 5b as a function of time for the SHiP/LHCb RPC. The resistivity slightly increased with time, and the origin of this phenomenon, observed also for other detectors, is currently under investigation. A possible contribution could be due to the possible damage of the RPC electrode inner surface; therefore, the surface itself will be examined for all the RPCs also by means of eventual chemical analyses.
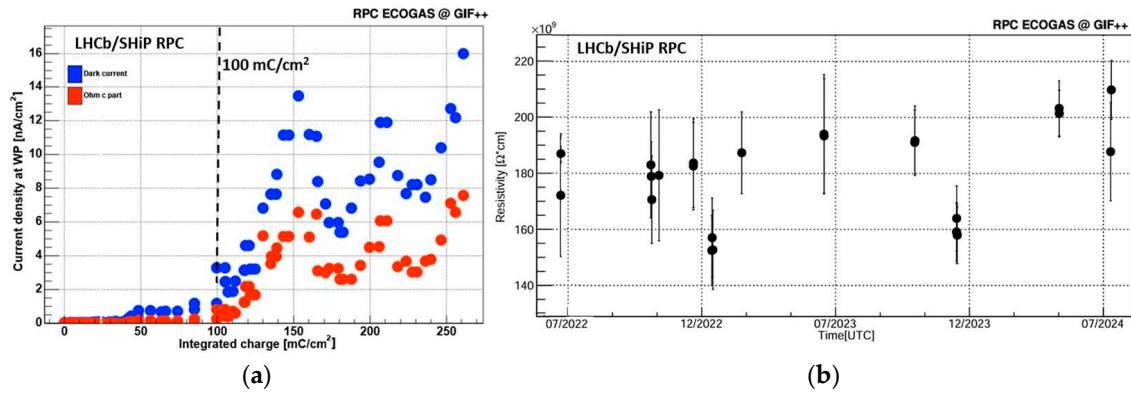
Figure 5. (**a**) Dark and Ohmic currents measured with the LHCb/SHiP RPC as function of the integrated charge; (**b**) electrode resistivity obtained with Argon measurements for the LHCb/SHiP RPC as function of time.

The comparison of the RPC performance measured at the test beams during the irradiation campaign with the baseline performance obtained before this campaign is crucial in order to study the ageing effects. The absorbed currents measured for the LHCb/SHiP RPC at different background conditions in 2022 (baseline) and after one year of irradiation, with an integrated charge of about 110 mC/cm^2, are shown in Figure 6. The currents absorbed at the WPs in 2023 increased with respect to the 2022 values, both for the standard gas and for the eco-gas mixtures. In particular, for the standard and ECO2 gases, the increment of dark currents and absorbed currents at all the background conditions was approximately the same, at the level of a few tens of μA. The current rise observed with ECO3 was instead enhanced at high absorbed doses, for instance, at about 20000 μGy/h, the current increased by a few hundreds of μA. The measured increment of currents as well as the larger electrode resistivities affected the voltage drop across the RPC electrodes (V$_{el}$). Since the voltage applied to the detector (HV$_{app}$) was obtained as the sum of V$_{el}$ with the voltage applied to the gas gap (V$_{gas}$), the increase in V$_{el}$ at the fixed HV$_{app}$ corresponded to a decrease in V$_{gas}$ and thus in the electric field in the gas gap, with a consequent reduction of the chamber efficiency at that HV$_{app}$. Therefore, the efficiency curves after the irradiation campaign shifted towards higher HVs, as shown, for example, in Figure 7 for the LHCb/SHiP RPC. The plateau efficiency measured with this chamber for each gas mixture was comparable between 2022 and 2023 both in the source off condition (Figure 7a) and with a background dose of about 6000 μG/h (Figure 7b). In addition, no signs of efficiency degradation have been observed up to now with the other RPCs under testing.
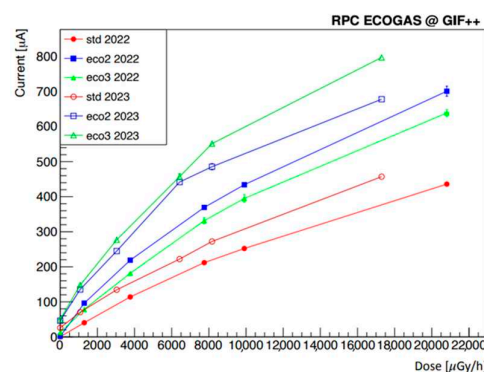

Figure 6. Currents absorbed by the LHCb/SHiP RPC during 2022 and 2023 beam tests as function of the background radiation dose.
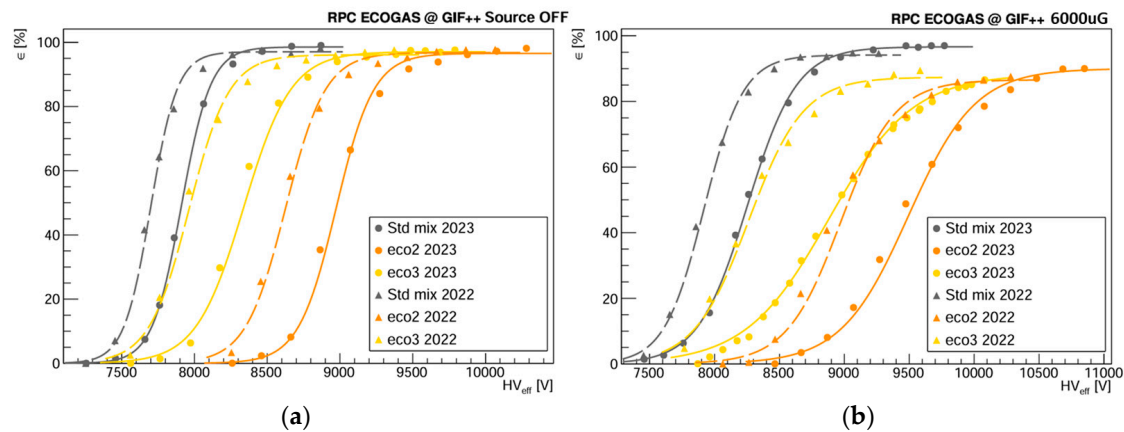
Figure 7. (**a**) The 2D efficiency of the LHCb/SHiP RPC before (2022) and during (2023) the irradiation campaign measured without irradiation as function of the HV_{eff}; (**b**) the 2D efficiency of the LHCb/SHiP RPC before (2022) and during (2023) the irradiation campaign measured with a background of about 6000 $\mu G/h$ as function of the HV_{eff}.

4. Discussion

The RPC ECOgas@GIF++ collaboration is committed to intensive research of new eco-friendly gas mixtures for RPCs complying with recent European regulations limiting the use of greenhouse gases. Alternative eco-gas mixtures based on CO_2/HFO have been tested with different RPCs at various background irradiation conditions at the CERN GIF++, showing promising results. In order to validate these gas mixtures, an ageing campaign was started in 2022 and is currently ongoing. After one year, with about one hundred mC/cm^2 integrated by RPCs, the performance of the detectors operated with the new eco-gases in terms of efficiency was preserved. An increase in currents and in electrode resistivity was instead observed. Possible explanations of these phenomena are currently under investigation and a contribution could be due to a deterioration of the electrode inner surfaces during the irradiation campaign; therefore, the status of these surfaces will be examined also by means of chemical analyses.

The ageing study of RPCs operated with alternative eco-gas mixtures will continue with the aim of testing long-term RPC operation at the high-luminosity LHC. The total charge to be integrated at the end of the ageing campaign by the detectors under testing is defined by the different LHC groups depending on the irradiation conditions of the RPCs for the specific experiment, and it is between 100 mC/cm^2 and 1 C/cm^2.

Conflicts of Interest: The authors declare no conflicts of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript; or in the decision to publish the results.

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