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Original research paper

METAL THIN-FILM DOSIMETRY TECHNOLOGY FOR THE ULTRA-HIGH PARTICLE FLUENCE ENVIRONMENT OF THE FUTURE CIRCULAR COLLIDER AT CERN*

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Abstract. *The Future Circular Collider (FCC) design study aims to assess the physics potential and technical feasibility of a new synchrotron accelerator expected to reach an energy level of 100 TeV colliding proton beams circulating in a 100 km tunnel located in the Geneva area in Switzerland. Inside the FCC detectors, over the 10 years of scheduled operation, unprecedented radiation levels will presumably exceed several tens of MGy with more than 10^{17} particles/cm². Current dosimetry technologies, such as silicon pin diodes, are not capable of integrating this particle fluence, thus requiring a new type of sensor to be used as dosimeter in future irradiation facilities and, at a later stage, in the FCC accelerator. As a solution for the Ultra High Fluence monitoring, we have focused our research on metal nanolayers. The technology consists of thin film resistive structures deposited on silicon wafers, where sensitivity to displacement damage, measurable in a variation of their electrical properties, can be trimmed by variating geometrical (thickness, W, L) and physical (material) properties of the nanolayers. The first prototypes of these new dosimeters have been fabricated at EPFL Centre of Micronanotechnology, and specific high-fluence irradiation tests (with gamma, protons, neutrons) have been carried out in several facilities inside and outside CERN. In this paper, after presenting the process flow for the fabrication of these dosimeters, we show the results of annealing tests performed on devices previously irradiated with 23 GeV protons. These measurements suggest the occurrence of an oxidation process that was enhanced by the radiation damage.*

Key words: *Dosimetry, Future Circular Collider, FCC, radiation effects, radiation monitoring*

1. INTRODUCTION

The European Organization for Nuclear Research (CERN) in Geneva, Switzerland, is presently considering the roadmap of future high energy physics (HEP) experiments and assessing the possible upgrade options that its accelerator complex can offer in the upcoming years. Among different projects, the Future Circular Collider (FCC) is by far the most ambitious of the proposed machines. This 100 km-long synchrotron will supersede today's Large Hadron Collider (LHC) and became the new world's largest and most powerful particle accelerator.

The FCC is going to be able to accelerate particles up to energies of 50 TeV per beam allowing 100 TeV collisions in the detectors at the interaction points (IP) where different experiments will be located. The final version of this machine, the FCC-hh, will allow colliding protons at a peak luminosity of $5 \times 10^{34}\,cm^{-2}s^{-1}$, intercepting up to 53000 bunches filled with 2×10^{10} particles every 5 ns [1]. Such great performances come at a cost of generating an extreme radiation environment, several times harsher than in the current LHC. This will pose new challenges for the design and selection of materials, electronic components, and systems to be used in the FCC detectors and in the tunnel. Monte-Carlo simulations were made to estimate the radiation environment cumulated over 10 years of FCC operation. These have shown tens of MGy with more than 10^{17} particles/cm² in the most exposed layers of the FCC detectors, few cm from the IP (Figure 1)[2]. These values are more relaxed in the FCC tunnel (KGy, $>10^{15}$ particles/cm²)[3].

In this context, the FCC Radiation Hardness Assurance working group was formed in order to fully understand what radiation environment to expect and to provide qualification protocols, instrumentation and solutions to these radiation issues. Until now at CERN, state-of-the-art dosimeters have been used to monitor the degradation of the electronics and detector components and to perform dosimetry during irradiation tests of these prior to their installation [4–6].

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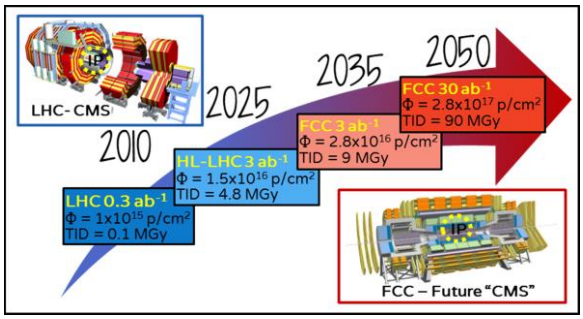

Figure 1. Simulated particle fluence (Φ) and total ionizing dose (TID), integrated over 10 years, in the inner detectors for different upgrade phases of CERN accelerators.

However, the levels expected in the FCC are several orders of magnitude higher than the ones maximally detectable with any of the currently available dosimetry technology, thus calling for new systems for ultra-high dose and fluence monitoring [7]. Specifically, innovative technologies are being studied in the field of nanometric structures which, thanks to their reduced volumes, are capable of withstanding the harsh radiation environment. In particular, metal thin films are of great interest because they are known to be insensitive to low particle fluence [8] (where current silicon-based devices saturate), and because they have been proven to show several changes of their properties (chemical, mechanical, transmutations, etc.) induced by very high particle fluence [9–11]. An example of such recent developments are, for instance, dosimeters for ultra-high dose monitoring based on optic fibers coated with a thin layer of PMMA and gold, where the dosimetric information is related to shifts in the peak of resonance of such structure [12].

The new technology, presented in this paper, is a dosimeter for ultra-high particle fluence monitoring: the Radiation Dependent Resistors (RDR). In this paper, in Section 2, we firstly outline the easy and inexpensive microfabrication process of the Radiation Dependent Resistors, followed by a description of the irradiation tests that were performed and the temperature measurements. Section 3 details the experimental results and discusses them, whereas Section 4 finally gives the conclusions.

2. Experimental Details

All the prototypes of Radiation Dependent Resistors described in this paper were produced in the class-100 cleanroom of the CMi (Center of Micronanotechnology, [13]) at EPFL (École Polytechnique Fédérale de Lausanne, Switzerland), whereas the electrical characterization, the temperature measurements and most of the irradiation tests were performed at CERN.

2.1. Microfabrication

Several wafers were produced with different deposition and/or patterning techniques of the metal layer. At first, we tried different metals such as chromium, aluminium and copper; then, based on the results from the first irradiation tests, we focused our interest on copper only.

As shown in Figure 2, several serpentine shaped devices were designed with different width W, and length L, by varying the number of turns.

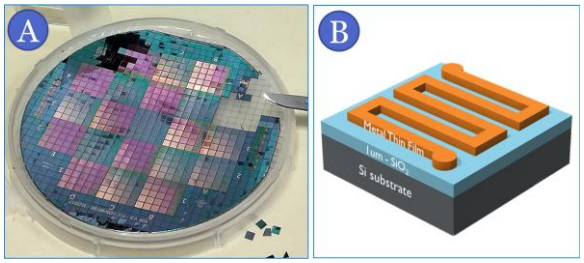

Figure 2. A) Photo of one of the produced wafers with 500 nm thick copper RDRs with in purple areas passivated with SiO_2, B) 3D model of a single 3x3 mm² RDR.

Table 1 lists the main fabrication steps followed to produce the Radiation Dependent Resistors. At first, the 100 mm silicon wafers were exposed to wet oxidation using a Centrotherm Furnace at 1050°C for 185 min, which allowed obtaining a 1 µm-thick insulating SiO_2 layer.

The second step was to deposit a thin film of copper, preceded by a Titanium layer needed to enhance the adhesion of Cu with the underlying SiO_2. Two machines were used to obtain a sputtered and an evaporated Ti/Cu stack. The Alliance-Concept DP650 sputtering chamber was used at room temperature with a power of 400 W reaching an average deposition rate for Ti and Cu of 3.1 Å/s and 12.6 Å/s respectively; the Alliance-Concept EVA 760 evaporator, at a working distance of 450 mm, was used to evaporate Ti and Cu at a rate of 5 Å/s. With both machines, copper layers of 50, 500 and 1000 nm were obtained (with Ti adhesion layers of 5, 10 and 10 nm, respectively).

The metal deposition was followed by the lithography step. The automatic Süss ACS200 Gen3 coater was used to obtain a uniform 2 µm-thick layer of Microchemicals AZ9221 positive photoresist, chosen for its high thermal stability during dry etching, for good adhesion and steep sidewalls. The exposure was performed with a Heidelberg Instruments MLA150 direct laser writer providing a dose of 105 mJ/cm², followed by the resist development with the Microchemicals AZ400K developer available in the ACS200.

Table 1. Process flow of a copper Radiation Dependent Resistor microfabrication, with different thin film deposition and etching techniques (a,b).

Fabrication Step	Machine	Details
1. Wet Oxidation	Centrotherm Furnace 2.2	1000 nm
2.a Cu Sputtering 2.b Cu Evaporation	Alliance-Concept DP650 Alliance-Concept EVA 760	(10 nm Ti + 500 nm Cu)
3. Photolithography	Süss ACS200 Gen3 Heidelberg MLA150	AZ9221 (2 µm) 105 mJ/cm²
4.a Dry Etching 4.b Wet Etching	Veeco Nexus IBE350 Wet bench	34 nm/min 200 nm/min
4. SiO_2 Passivation	Pfeiffer SPIDER 600	300 nm SiO_2
5. Dicing	Disco DAG810 System	3x3 mm² dies

To complete the patterning of the RDRs, an ion beam etching was performed with a Veeco Nexus

IBE350 etcher. The wafers were bombarded with a low power beam at an angle of 10° achieving an etch rate of 34 nm/min. Alternatively, a selection of wafers was etched in a solution of Ammonium Persulphate and Sulphuric Acid (60 g/l and 10 ml/l, respectively) achieving an etch rate of 200 nm/min.

As the last processing step, a selective passivation of some of the devices of the wafers was obtained by sputtering, through a hard mask, of 300 nm of SiO_2 at a deposition rate of 40 nm/min.

Finally, all the produced wafers were diced using a Disco DAG810 dicing system, resulting in 3x3 mm² chips (Figure 2.B) ready to be electrically measured using a Süss PM8 probe station equipped with a Keithley 4200A Semiconductor Parameter Analyzer. The selected chips were then wirebonded with a wedge-wedge 33 µm aluminium wire using a TPT HB10 Bonder on a specifically rad-resistant PCB called FCC-RADMON shown in Figure 3.

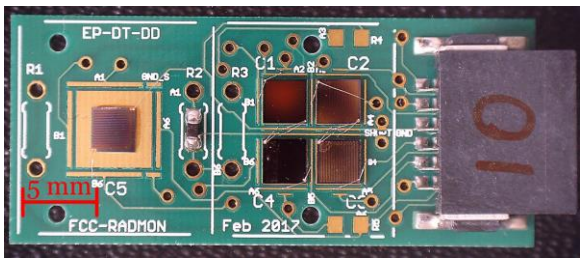

Figure 3. An FCC-RADMON printed circuit board (1.5 x 3.5 cm²) produced on a radiation resistant Doosan DS-7402 Hal-Free 0.2 mm substrate.

2.2. Irradiation Tests

As shown in Table 2, several irradiation experiments were performed (or are still ongoing), to test the RDR sensor performance under conditions similar to the ones in which they will be used. For this purpose, experimental studies were carried out at the TRIGA Nuclear Reactor of the Jožef Stefan Institute (JSI) [14] in Ljubljana, Slovenia, at the Proton Irradiation Facility (IRRAD) [15] at CERN, next to the Linac target in the CERN GBAR experiment [16], as well as next to the luminosity monitors (BRAN) in the Neutral Beam Absorbers (LHC-TAN) of IP1 at LHC [17].

Table 2. List of irradiation experiments already performed and/or ongoing on the RDRs. *(* tests still ongoing)*

Test Location	Radiation Type	Max Φ/ TID
TRIGA Nuclear Reactor at JSI	Neutrons	$1x10^{18}$ n_{tot}/cm^2
IRRAD Proton Facility at CERN	Protons	$1x10^{17}$ p^+/cm^2
GBAR at CERN	Gamma rays	~10 kGy*
LHC-TAN at CERN	Mixed field	~7 MGy*

The results from the irradiation campaigns at JSI and IRRAD of 2017, reported in [18], have confirmed our proposed idea of using metal thin films for performing dosimetry of non-ionizing radiation. The test in GBAR at CERN, in a pure ionizing radiation field, is currently ongoing, but is already confirming the ability of the RDR in discriminating the Φ from TID, being the RDRs unaffected by ionizing radiation (test being performed in CERN-GBAR). Further experiments are currently continuing, as a second run with only the copper samples in JSI and IRRAD, as well as the ongoing test in the LHC-TAN.

2.3. Temperature Measurements

Metals are known to be positive temperature coefficient (PTC) materials, meaning that their electrical resistance increases with increasing temperature. While such temperature coefficient α is constant and well-defined for bulk metals, for thin films, the coefficient α is not constant over a wide range of temperatures, and is found to be strongly thickness dependent [19]. For this reason, the prepared FCC-RADMON boards were characterized in different ovens, shown in Figure 4, and their temperature characteristics, discussed in Section 3, were extracted before and after irradiation.

The pre-irradiation characterization was performed in a commercial Climats SPIRALE 3 climatic-chamber available at CERN. Such chamber allowed testing samples at different temperatures with a controlled humidity.

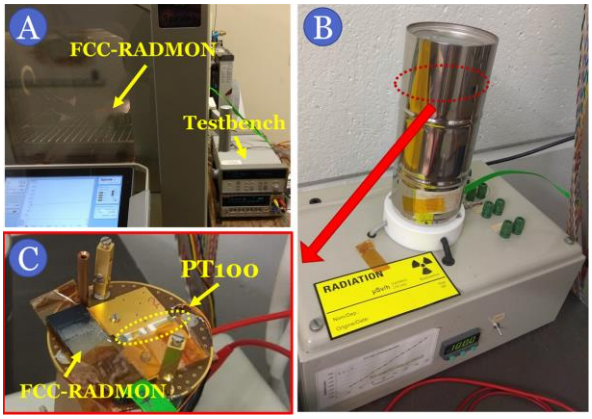

Figure 4. A) Climatic chamber to test samples before irradiation and B) Self-made oven to test after irradiation; C) Detail of the inner part of the cylinder highlighting the PT100 sensors and the FCC-RADMON under test.

The post-irradiation characterization was instead performed in a custom-made oven, shown in Figure 4, which can also be used for radioactive samples. This oven uses a 50 Ω, 100 W resistor to uniformly heat a 4 cm Ø support. The power through the resistor is controlled with a power width modulated (PWM) 1 A current controlled by a proportional–integral–derivative (PID) controller. The temperature was monitored using the two PT100 mounted on the sample support, as well as the NTC soldered on the FCC-RADMON PCB.

Measurements were performed over a long period of time (several hours) to reach a stable temperature of ±0.2% around the setpoint.

3. Experimental Results and Discussion

Figure 5 shows an extension of the results from the irradiation tests performed between June and December 2017 in IRRAD with 23 GeV protons, on RDRs of copper, aluminium, previously presented in [18].

The experimental data have clearly shown a dependence of RDR resistivity against particle fluence. Such effect can be justified by inspecting the effect of cumulative particle damage: with increasing particle fluence grows the probability of the release of non-ionizing energy in the metal lattice, which induces a displacement of an atom from its original position. Such displacement rate, often expressed in literature as displacements per atoms (DPA), introduces defects in the lattice, increasing the electron scattering, thus increasing the resistivity of the metal. This damage resulted to be permanent, without any annealing processes when stored at room temperature. Nevertheless, in order to study the stability of this resistivity increase, we have performed an annealing test which is reported here.

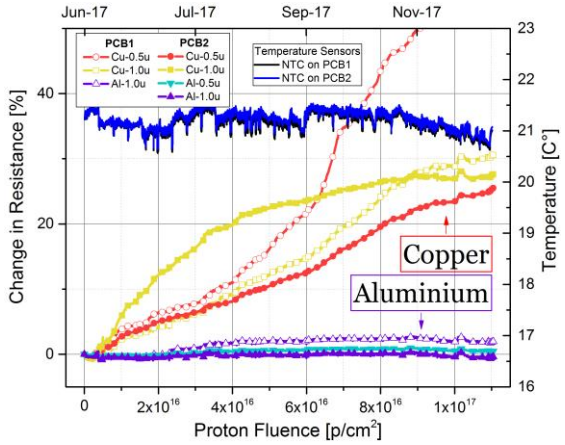

Figure 5. Resistance variation from the initial value in % (left y-axis) vs the integrated particle fluence. On the right y-axis: the measured temperature during the irradiation test, considerable constant around 21˚C. Experimental data from the RDR irradiation test performed in 2017 with 23 GeV protons in IRRAD, CERN.

3.1. Temperature Test Results

Using the experimental equipment described in Section 2.3 and shown in Figure 4, samples were annealed before and after irradiation. The measurements before irradiation (not presented in this paper) were performed with 10 hours annealing in air, ramping at constant rate from 20˚C to 60˚C and from 20˚C to 80˚C. After these measurements, temperature coefficients were extracted for each of the sample. Moreover, none of the 30 different RDRs tested have shown any measurable permanent increase or decrease of resistivity, thus indicating an overall good stability and uniformity of the sputtered metal films.

Concerning the annealing measurements on RDRs irradiated in IRRAD, only the PCB2, shown in Figure 5, has been tested, since the *Cu-0.5u* sample of PCB1 has failed after a fluence of ~7x10^{16} p/cm^2. The sample has been annealed for about 150 hours, as shown in Figure 6. Such test was performed 5 months after the end of irradiation, once the activation of the sample was reasonably low. During this time, the PCB was stored at CERN in a radioactive storage at ambient temperature.

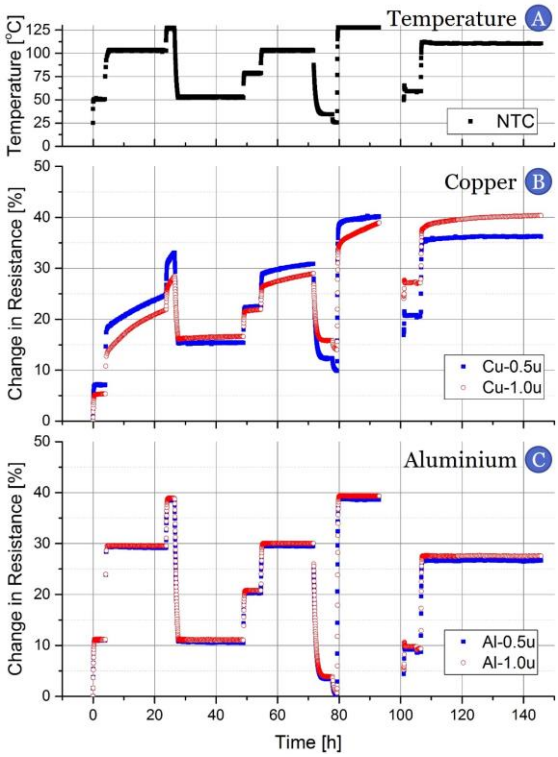

Figure 6. Change in resistance of copper (B) and aluminium (C) vs time at varying temperature (A). Annealing performed on the PCB2 previously irradiated in IRRAD during 2017 run.

As shown in Figure 6, after observing no apparent variation of resistivity for 4 hours at 50˚C, the temperature in the oven has been increased to 100˚C for 20 hours. During this time, while the aluminium samples (Figure 6.C) have not shown any change in resistance at constant temperature, the copper samples have shown a positive increase (Figure 6.B). This behavior was even more visible when increasing the temperature to 125˚C for the next 4 hours, at which the slope of change of resistance for copper almost doubled. By further decreasing and increasing the temperature, following the temperature curve in Figure 6.A, the behavior of the aluminium samples remained the same, whereas the copper ones, after more than 100 cumulative hours at high temperature, have shown a saturation of such increasing effect.

An explanation of what has been recorded during this post-irradiation annealing test is an oxidation process of the copper film together with a diffusion of the oxygen-rich copper oxide layer inside the bulk. Normally, metal films create a native oxide layer after exposure to air. In both aluminium and copper, such a layer measures few nm and, after its formation, it drastically reduces further oxidation at room temperature [20]. While, in case of aluminium, this native oxide layer is shown to be effectively protecting the metal at a wide range of temperatures, whereas in case of copper, at temperatures (200˚C - 300˚C) the

oxygen-rich native CuO oxide layer is shown to penetrate the Cu layer, creating an enlarging Cu_2O [21, 22]. This oxidation rate is demonstrated to be dependent on temperature and Cu_2O grain size, thus on the amount of free diffusional paths available for the oxygen to further oxidize the underlying copper. Such documented effect may have been enhanced by the radiation damage of the film, where displacement damage in the native oxide resulted in an increase in the number of these diffusional paths, resulting in a visible and measurable increase of resistivity of the film even at low temperatures as around 100°C. Another possible explanation for the resistivity increase is the presence of agglomeration processes induced by temperature and radiation, causing discontinuity in the film. This will be checked upon morphological analysis with a scanning electron microscope (SEM) of the samples before and after irradiation.

4. Conclusions

In this paper, we presented the microfabrication steps, the irradiation tests, and temperature characterization of a novel dosimetry technology for monitoring very high particle fluence. Such device, called the Radiation Dependent Resistor (RDR), has shown an increase of resistance with increasing particle fluence and, differently from silicon based p-i-n diodes commonly used for fluence dosimetry, the RDR has shown no saturation or breakdown until a tested extreme fluence of 10^{18} neutrons/cm^2 and 10^{17} protons/cm^2. Such behavior confirms the possibility to relate particle fluence to an increase of resistivity of metal thin films.

Annealing tests were performed before and after irradiation. While, before irradiation, no effect was recorded on metals resistivity under prolonged annealing at 60°C and 80°C, after irradiation, copper has shown a continuous and permanent increase of its resistance which started saturating only after 100 hours at 100°C. Such behavior could be explained by an increase of diffusional paths available to the oxygen for oxidizing the copper bulk samples after irradiation, thus increasing the sites where oxygen can oxidize copper. Such behavior can also be explained by temperature-induced film agglomeration.

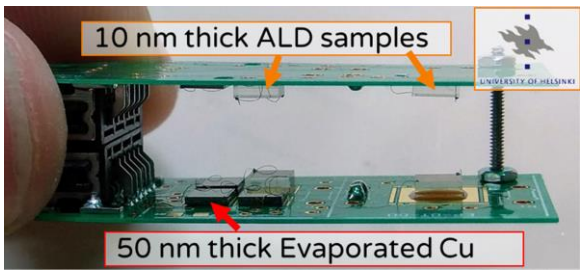

Figure 7. New batch of PCB currently undergoing irradiation tests. New samples of copper RDR were produced, along with ultra-thin ALD samples produced by the University of Helsinki.

Further studies on the effects of particle fluence on copper thin films is ongoing by producing a larger set of different devices, as shown in Figure 7, fabricated with different techniques (e.g. evaporated Cu instead of sputtered), with different thicknesses (10 nm to 1000 nm), and by passivating some of the devices to exclude oxidation processes during irradiation. Special attention will be given to samples prepared with the atomic layer deposition (ALD) technique, where extremely thin continuous copper layers can be obtained starting from 8 nm [23].

Finally, an annealing test will be performed in different atmospheres (e.g. helium) to assess the impact of the oxygen concentration on the increase of the oxide layer on the copper thin film, followed by an extensive morphological analysis such as scanning electron microscope (SEM) and atomic force microscopy (AFM) imaging as well as X-ray diffraction (XRD) and X-ray photoelectron spectrum (XPS) techniques, of copper samples before and after irradiation.

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