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Aging and conductivity of electrodes for high rate tRPCs from an ion conductivity approach

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ABSTRACT:

Resistive Plate Chambers (RPCs) are multipurpose ionizing gas detectors broadly used in Particle Physics research because of the outstanding time resolution they offer at a very affordable price, their robustness and their easy manufacture. However, to extend their use to a broader range of applications, some properties, specially the counting rate capability, need to be improved. The straightforward way to increase the rate capability is to develop new electrodes with resistivities in the range of 10^7-$10^8\Omega$m offering, besides a "resistive" character, conductivity high enough for allowing faster charge depletion in the chamber. In this work, we have carried out electrical measurements on several materials that are currently used or, according to their conductivity values, are good candidates to be used as resistive plates in high rate RPCs. We have analysed their behaviour in order to identify and to better understand their conduction mechanisms and to determine the nature of the degradation processes.

XI workshop on Resistive Plate Chambers and Related Detectors (RPC2012)
INFN-Laboratori Nazionali di Frascati, Italy
February 5-10, 2012

[1] Speaker

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1.Introduction

RPCs are multipurpose detectors with demonstrated value in Nuclear and Particle Physics experiments and in Astrophysics. In the near future, their use in medical devices, as in Positron Emission Tomography (PETs), has been also foreseen [1].

One of the main restrictions of the RPCs is their limited rate capability (below 2 kHz/cm^2 for minimum ionizing particles, MIPs, with float glass RPC's at room temperature)[2]. As the resistive plate itself are the main barrier to the gas ionization charges evacuation, it seems that one of the most straightforward ways of improving that feature is the upgrade of the resistive plates electrical properties. According the DC model [2], resistivity at least one or two orders of magnitude below the present $10^{9\text{-}10}\Omega$m would be required to achieve an improvement of the same order in the rate capability. Unfortunately, such kind of materials, also known as static-dissipative or lossy materials, are quite unusual and most of the times they have to be specially designed to fulfil the requirements.

Besides resistivity, other properties, as the permittivity, the dielectric strength (breakdown field) or the current stability have to be considered. Also, as resistive plates of high rate RPC based detectors need to drift more charge than of the low rate ones all these features should show a good behaviour, along the whole detector life time. For instance, the charge that will pass through the RPC TOF at the CBM experiment [4] at FAIR, along the expected 5 years lifetime is around 1C/cm^2; this value is usually taken as a benchmark reference for ageing tests.

In this document, we present the results of the analysis we have done about resistivity and charge depletion properties of several materials that have been used either in low or in high rate RPCs. We also analyse the same properties for some new ceramics that we are being developed for their use in high rate timing RPCs. We have developed a model showing the ionic conductivity component and the current dependence with electric field and time. All the materials we have analysed are shown at Table 1.

Material Type	Material	Provider
Glass	Soda Lime Silicate Glass (SLS Glass)	Hades experiment, GSI(Darmstadt)
	Low Resistive Silicate Glass (LR S Glass)	Tsinghua University
Polymer	Bakelite (High Pressure Laminate)	CMS experiment
Ceramics	Mullite/Mollybdenum (Mullite/Mo)	ICMM/CSIC, Madrid
	Ferrite Ceramic	ICMM/CSIC, Madrid

Table 1: Set of materials that have been tested in this work. They have been chosen among three kinds of materials in order to get a broader scope.

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2.Ion conductivity and ageing model

2.1 Ionic conductivity electric field dependence

Due to the nature of the analysed materials, we may suppose that most of the charge is carried out by ions. Then, the current density J is given by:

$$J = nq\mu \tag{1}$$

where n is the number of carriers per unit of volume, q is the ion charge and μ is the mobility, or the velocity attained by these ions under an electric field. The average drift velocity of an ion affected by a high electric field is given by [5]:

$$\mu = 2\, l\, \nu_0 e^{-\frac{W}{kT}} \sinh\left(\frac{qEl}{2kT}\right) \tag{2}$$

where l is the so called jump length, ν_0 is the oscillation frequency of the ion in the potential well (~$10^{13}s^{-1}$), W is the potential barrier and E is the "microscopic" electric field seen by the ions. Finally, the current density, J, can be written as a function of the applied electric field as:

$$J = 2\, l\, q\, \nu_0 n\, e^{-\frac{W}{kT}} \sinh\left(\frac{qEl}{2kT}\right) \tag{3}$$

According to this law, the current density has a linear dependence on E at low electric fields and follows an E^3 tendency with increasing values of E.

Fig.1 shows the results of the measurements done of the current density J as a function of the applied electric field for some of the analysed materials together with the results of the fits to function:

$$J = C_1 \sinh\left(C_2\, E\right) \tag{4}$$

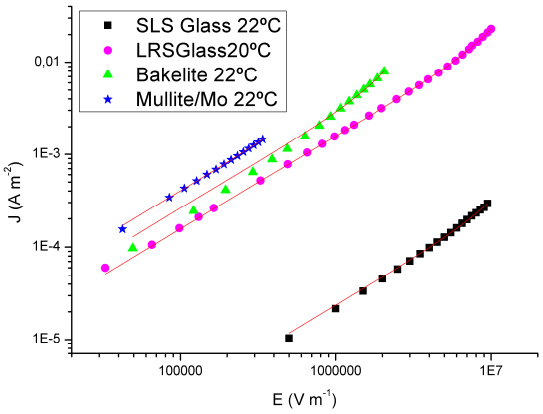

Figure 1: Current density as a function of the applied electrical field measured for some of the materials we are testing. The dashed lines show the results of the fits done to the function explained in the text.

According to Eq. (3) the fitted coefficients C_1 and C_2 are:

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$$C_1 = 2\,l\,q\,\nu_0 n\,e^{-\frac{W}{kT}} \qquad C_2 = \frac{ql}{2kT} \tag{5}$$

And consequently resistivity, ρ, and mean free path, l, are:

$$\rho \propto \frac{1}{C_1 C_2} \qquad l = \frac{2kT}{q} C_2 \tag{6}$$

Table 2 show the results we have obtained for all the analysed materials assuming the ionic conductivity model. It is interesting to observe that the mean free length of the mullite/Mo plate is significantly bigger (10^{-7}m) than the ones of the other materials. This is a well known effect caused by the amplification of the electric field that appears in metal/insulator composites being close to the percolation threshold.

	C_1(A m^{-2})	C_2(m V^{-1})	ρ(Ω m)	l (m)
SLS Glass	$1.7 \pm 0.2 \cdot 10^{-4}$	$1.4 \pm 0.1 \cdot 10^{-7}$	$4.2 \pm 0.7 \cdot 10^{10}$	$7.0 \pm 0.6 \cdot 10^{-9}$
LR S Glass	$1.00 \pm 0.01 \cdot 10^{-2}$	$1.57 \pm 0.01 \cdot 10^{-7}$	$6.4 \pm 0.1 \cdot 10^{8}$	$7.9 \pm 0.2 \cdot 10^{-9}$
Bakelite	$3.3 \pm 0.4 \cdot 10^{-3}$	$7.8 \pm 0.7 \cdot 10^{-7}$	$3.8 \pm 0.8 \cdot 10^{8}$	$4.0 \pm 0.4\text{x} \cdot 10^{-9}$
Mullite/Mo	$2.0 \pm 0.1 \cdot 10^{-3}$	$2.0 \pm 0.1 \cdot 10^{-6}$	$2.5 \pm 0.3 \cdot 10^{8}$	$1.01 \pm 0.08 \cdot 10^{-7}$

Table 2: Results of the fit of the ionic current model explained in the text to different materials; both resistivity (ρ) and mean free length (l) have been derived from the function coefficients, C_1 and C_2.

2.2 Ionic aging model

In order to check the RPC's stability under an applied electric field, measurements of the relevant magnitudes were done for extended time periods. Also, for achieving a meaningful amount of charge transferred keeping the experiments within a reasonable time span, grinding the materials and rising the temperature was done when needed. All the measurements were done inside a Faraday cage for minimizing external interferences. Gas environment was under control, keeping N_2 steadily flowing inside the box to avoid external charge carriers additions, as O_2 or humidity. Temperature was acquired regularly by a pt100 sensor placed close to the sample and regulated by a Peltier cell controlled by computer. The electric field was applied to the resistive plates by the electrometer itself (a Keithley 6517A) using painted silver electrodes.

Fig. 2 shows the evolution of the resistivity with the transferred charge for several materials at different temperatures. They show quite different behaviours that can be analysed using the following model: First, we assume that RPC plates are poor ion conductors with blocking electrodes, so that ions move to the corresponding electrode when a DC current is applied. When a carrier reaches the electrode it gets blocked there. In fact, some electrochemical reactions take place at the material/electrode interface. In any case, all carriers that arrive to the interface do not participate anymore in the ion conduction process. Then, the carrier density loss is related to the faradaic current through equation:

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$$\frac{dn}{dt} = -\frac{J}{qd} \tag{7}$$

Where d is the sample thickness. In case of low to moderate electric field, J can be approximated as Eq. (3).

$$J(t) = l^2(t)\, n(t)\, \frac{E\nu_0 q^2}{kT} e^{-\frac{W}{kT}} \quad (8) \qquad n(t) = \frac{J(t)}{l^2(t)} \cdot \frac{kT}{E\nu_0 q^2} e^{\frac{W}{kT}} \quad (9)$$

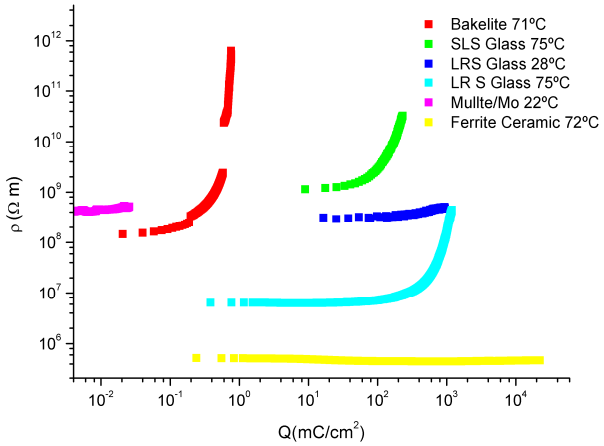

Figure 2: Resistivity vs. transferred charge measured for several materials and different temperatures. Both, Bakelite at 71ºC (which can not stand even 1 mC/cm^2 when there is no a gas moisture able to provide H+ carriers) and Ferrite Ceramic at 72ºC (which keeps a flat response even after having delivered 22000 mC/cm^2) show the extreme behaviours.

According to the results of Fig. 2, we have explicitly introduced the dependence of the mean free path with the time, while the remaining terms are assumed to be constant. As the variation of l(t) is expected to be not very large (~25% for 1 month, with SLS glass) we have introduced a quadratic time dependence of the form:

$$l(t) = l_0\left(1 - \frac{t}{\tau_1} + \frac{t^2}{\tau_2^2}\right) \tag{10}$$

Integrating n(t) from Eq. (7) we get:

$$J(t) \simeq J_0\left[1 - 2\frac{t}{\tau_1} + t^2\left(\frac{1}{\tau_1^2} + \frac{2}{\tau_2^2}\right)\right] e^{-\frac{t}{\tau_n}\left[1 - \frac{t}{\tau_1} + \frac{t^2}{3}\left(\frac{1}{\tau_1^2} + \frac{2}{\tau_2^2}\right)\right]} \tag{11}$$

Where parameter τ_n is related to the carrier leakage rate and τ_l to the matrix deformation rate.

$$\tau_n = \frac{d\, kT\, e^{\frac{W}{kT}}}{l_0^2 \nu_0 qE} \tag{12}$$

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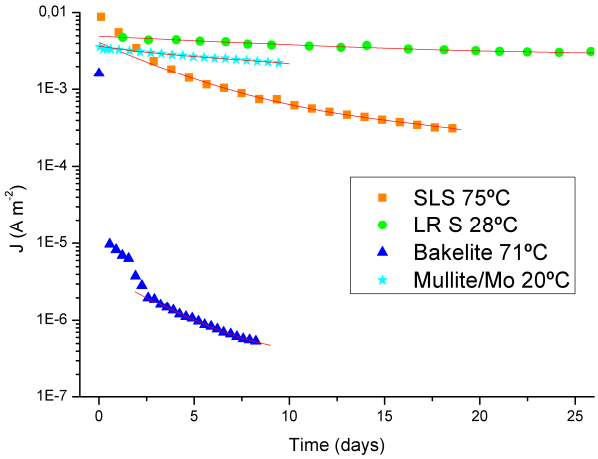

Figure 3: Density current vs. time behaviour for some of the analysed materials. The dashed lines show the results of the fits to the ionic conductivity model. As the measurements of the first days are affected by polarizations and other transitory effects they have been excluded from the fit.

Fig. 3 shows the results of the fit to several materials and Table 3 show the values of the fitted parameters; in general, there is a good agreement between data and model. Bigger values of τ_n stand for greater times to lose conductivity due to the charge drift. For instance, SLS Glass looks to be a very poor candidate for long life high rate RPCs. On the other hand, LRS Glass has a poor agreement to the model (uncertainties are very big), being the main reason that the loss of conductivity is mainly not ionic.

	J_0 (A m^{-2})	τ_1 (days)	τ_2 (days)	τ_n (days)
SLS Glass	$4.2\pm0.6 \cdot 10^{-3}$	26.3±4.2	32±8	5.6±0.9
LR S Glass	$4.9\pm0.3 \cdot 10^{-3}$	100±100	91±25	200±1200
Bakelite	$4.45\pm0.06 \cdot 10^{-6}$	14.7±0.6	31.3±0.5	48±2
Mullite/Mo	$3.7\pm0.2 \cdot 10^{-3}$	333±667	20±4	12±2

Table 3: Fitted parameter of the ionic conductivity ageing model for several materials. Larger times and a worse fit to the model stand for a poorer ionic conductor and a better high rate RPC candidate. Ferrite ceramic is not included in the table because it did not suffer any relevant charge depletion.

3. Conclusions

In order to get a better insight into the electrical properties of the resistive materials used in high rate RPCs we propose two models allowing to identify an ionic conductivity charge transport mechanism. The models are based on the conductivity response to an external electric

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field and the time needed to deliver the charge; they have been applied to several materials that have been developed for their use in high rate RPCs. Under the light of these models, materials were classified by its ionic ageing rates. As the ionic conductors always lead to charge depletion phenomena, unless an extra charge is supplied by the plate environment, the working lifetime will be finite and strongly related with the delivered charge.

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