# Attempt to Correlate the Ionic Model with Observations in BaBar RPC Chambers and R&D Tests

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Abstract—The paper presents a model of ionic conduction in the Bakelite-based RPC detectors. This model explains why these detectors need to add water while they operate. The electrode current, according to the presented model, is formed mainly by the ionic sequence involving the phenol impurities left in the Bakelite and water, both left from the initial production. Similar ionic process is present in the Linseed oil polymer, where the current is carried by the fatty acid molecular impurities and by water. The Bakelite RPC operation requires the entire ionic sequence to operate smoothly to keep the electrode resistance constant.

### I. INTRODUCTION

In this presentation, I will discuss the BaBar design only, i.e., the Bakelite-based chambers with the Linseed oil, and operating in the streamer mode. I will not talk about obvious errors, such as over-oiling, over-voltaging, over-heating, or running a voltage while oil was soft, etc. After a short introduction to the BaBar RPC efficiency decay, I will discuss the problems from a point of view of the ionic model [1]. This model was first discussed at DESY Aging Workshop [2].

I will not talk about the electrical behavior of RPCs. This aspect is much better understood, and nicely described, for example, by Riegler [3].

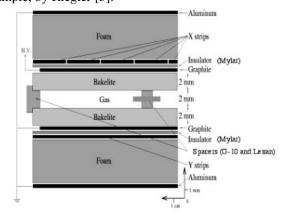


Fig. 1. BaBar design of the single gap RPC.

BaBar has 774 RPC chambers covering a total area of ~2300m<sup>2</sup>. The design has 18 gaps in iron, each equipped with a single-gap RPC. Fig. 1 shows the chamber design.

Various electrodes in the BaBar RPC have these typical resistivity values:

- a) G-10 side spacers, Lexan button spacer:  $\rho_V \sim 10^{13} \ \Omega \text{cm}$ .
- b) Bakelite electrodes:  $\rho_V \sim 10^{11} 10^{12} \Omega \text{cm}$ .
- c) Graphite surface resistivity:  $\rho_S \sim 100 k\Omega/square$ .

The BaBar RPC chambers when constructed, were filled 3-times with Linseed oil/n-pentane (70:30 mix), and flushed with air for 60 hours after each filling. The chambers work with a gas of 60.6% Ar + 34.7%  $C_2H_2F_4$  + 4.7%  $C_4H_{10}$  in a streamer mode operation, typically with ~1000pC/track. The electronics has 53000 channels; threshold ~40mV.

Fig. 2 shows a simple-minded static resistive equivalent model. We want to use it only for illustration purposes. The effective resistances for a " $10x10cm^2$  one-button RPC segment" are:  $R_{Bakelite} = \rho_V (t_{gap}/Area) \sim 5x10^8 \, \Omega$ , where  $t_{gap}$  is Bakelite thickness and Area is  $10x10cm^2$ , and  $R_{Lexan\ button} = \rho_V (t_{gap}/Area) \sim 3.4 \, x \, 10^{11} \, \Omega$ , where  $t_{gap}$  is button thickness and Area is its footprint area. For these values, the RPC gap voltage is a full power supply voltage  $V_{GAP} \sim V_{PS} / (1 + 2R_{Bakelite}/R_{Lexan\ spacer}) \sim V_{PS}$ . For this condition, the RPC works well. To illustrate possible problems related to the resistivity changes, assume Bakelite volume resistance to increase by a factor of 60. For that condition one gets  $V_{GAP} \sim 0.85 \, x \, V_{PS}$ , which would already mean that the chamber would start losing efficiency [1].

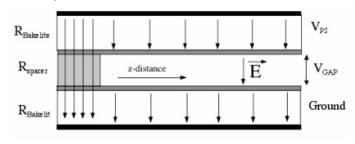


Fig. 2. Equivalent model of the electric field in "well behaving" RPC.

As we will discuss there are several ways to change the Bakelite resistivity. For example, one can influence it by a dry gas, heating or running a large charge through its volume. Typically, as we will see in the next chapter, a charge density as small as ~0.1-0.2 C/cm² can already affect the volume resistivity substantially [1]. Fig. 3 shows that many BaBar RPC chambers can reach much larger charge density just from the particle background alone. In addition, a local sparking, for example around buttons and edges, can increase these densities even more, as we will see later.

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Consequence of the Bakelite resistivity increase is a loss of the rate handling capability. As was shown by R. Arnaldi et al. [4], a Bakelite-based RPC operating in the streamer mode can start losing the efficiency above ~20 Hz/cm². For example, Fig. 4 shows a projection of the BaBar RPC rates based on the present background projections. One can see that the rates are not very large for most of the BaBar chambers, however, any persistent local sparking or a large Bakelite volume resistivity increase, say by a factor of ~10, can start affecting the efficiency.

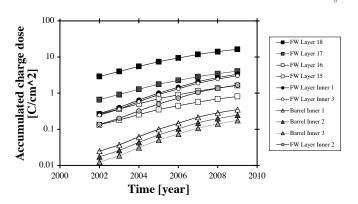


Fig. 3. Projection of the accumulated charge density in the BaBar RPC detectors based on running experience of past 3-4 years. The estimated charge is due to the accelerator background only, i.e., it does not include anothing effects.

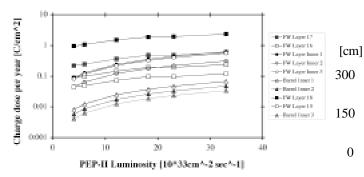


Fig. 4. Projection of the rates in BaBar RPC detectors based on experience of past 3-4 years.

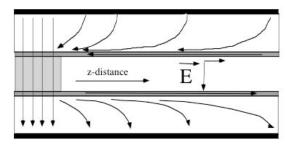


Fig. 5. Electric field has a tangential component in a damaged RPC, where the Linseed oil resistivity is lowered due to a chemistry reaction with the Freon molecule in a presence of UV light.

So far we have not mentioned the Linseed oil. Its resistivity can also change due either to dry gas environment or a large charge density. However, since the Linseed oil is exposed to a Freon-based chemistry under an influence of the UV light from the avalanches, one can also lower its resistivity substantially [1]. Especially bad consequence is lowering resisitivity of the Linseed oil-covered buttons. Such effect can divide a voltage across the RPC gap near the button, as described pictorially on Fig. 5, the efficiency near a button will be lowered.

We will be dealing mostly with the RPC chambers installed to BaBar in November 2000. These chambers already had many improvements in the Linseed oil handling and have also much better Bakelite surface. Several of these chambers were removed from BaBar after 2 years of operation, so one can perform various tests.

### II. BABAR EXPERIENCE

We will spend only a small amount of time on the BaBar experience, as there is a dedicated talk by H. Bend. A fate of the original BaBar chambers is well known. Fig. 6 shows the 2-D efficiency scan of FW Layer 2 chamber, which is the very original construction and had low rates in BaBar. This is the only example of the old-style chamber in this paper.

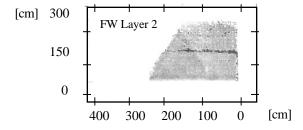


Fig. 6. Efficiency decay in FW Layer 2, the original BaBar RPC chamber. It shows a periodic pattern of lower efficiency (black spots) near buttons, as well as along the edge [5].

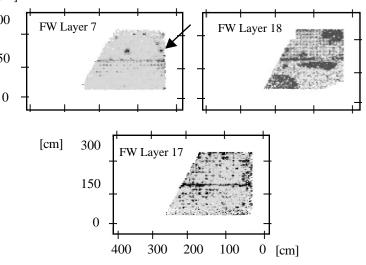


Fig. 7. Efficiency decay in Nov. 2000 BaBar RPC chambers [5]. FW
Layer 7 had a small rate, FW Layer 18 and 17 had a high rate in
BaBar during their 2-year long exposure. One can see that there
is a large number of low efficiency spots near the buttons. The
arrow indicates a spot discussed in Fig. 8.

However, even the new RPC chambers from so-called November 2000 production suffered large efficiency losses. For example, Fig. 7 shows 2-D efficiency scans of some of these chambers. Some were running with very low particle rates, for example FW Layer 7, and some at high rates, for example FW Layer 18. One can see that there are many low efficiency spots located near the buttons. FW Layer 18 has the largest number, FW Layer 7 much smaller, but still visible. However, even well shielded FW Layer 7 had a significant loss of efficiency of ~ 7.2% (module 1) and 12.7% (module 2) in one year of BaBar operation [5]. After the FW layer 7 was removed from BaBar in 2002, we have decided to look at a spot indicated by the arrow on Fig. 7. Fig. 8a,b,c show the spot appearance inside the chamber. There was a large star-like pattern near a button on the anode surface. On the corresponding spot on other side of the Bakelite one could see a gas bubble between the Graphite and the Mylar, and the Graphite was discolored. In many cases, when Mylar is peeled off, its glue takes away the disturbed Graphite around the button with it, again indicating that the Graphite is attacked. Further investigation revealed a large number of star-like patterns in this chamber, all on anode side only - see Fig. 8d. We believe that they correspond to breakdowns near the buttons. There was perhaps 50-60% of buttons affected. All these spots correspond to local sparking and a large local charge density.

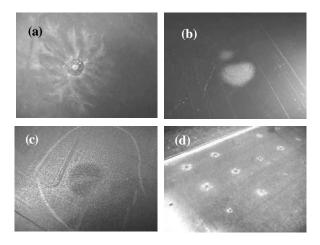


Fig. 8. A spot, indicated by an arrow on Fig. 7, appears inside the FW Layer 7 chamber as (a) a star on the anode surface, (b) a bubble between the graphite and the Mylar, and as (c) a discolored spot on the graphite. There are many star-like patterns on the anode as is seen in (d).

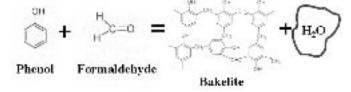


Fig. 9. The Bakelite is produced by a chemical reaction of Phenol and Formaldehyde. A byproduct of this reaction is water. The Bakelite is randomly cross-linked polymer of Phenol molecules. It has low conductivity on its own.

### III. IONIC MODEL

To understand the presented model one has to first understand how the Bakelite is produced. Fig. 9 shows a basic chemistry involved in its production. The important point is that water is an important byproduct of the chemical reaction, and therefore it is present right from the beginning in the Bakelite volume. Furthermore, it is reasonable to assume that there are also Phenol impurities left in the Bakelite, uniformly distributed throughout the volume. There are other impurities, no doubt. For example, our elemental analysis of the electrodes confirmed presence of sodium [1].

Fig. 10 shows our proposed ionic model of the Bakelite conductivity [1,2]. A possible ionic sequence is as follows: (a) Phenol + potential  $\rightarrow$  H<sup>+</sup> + Benzene-O<sup>-</sup>; (b) either Benzene-O<sup>-</sup> ion delivers the charge to anode and Benzene-O returns to the fluid, or Benzene-O<sup>-</sup> ion transfers a charge to OH<sup>-</sup> ion via a reaction Benzene-O<sup>-</sup> + H<sub>2</sub>O  $\rightarrow$  Phenol + OH<sup>-</sup>; Phenol returns into the cycle and OH<sup>-</sup> transfers the charge to anode; (c) H<sup>+</sup> ion delivers the charge to the cathode, where it forms an H<sub>2</sub> molecule and escapes; (d) 2OH  $\rightarrow$  H<sub>2</sub>O + 2O, and 2O  $\rightarrow$  O<sub>2</sub>, which delivers oxygen near the anode. Oxygen at the anode could react with a graphite layer.

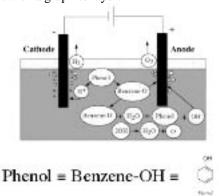


Fig. 10. Ionic model of the Bakelite conductivity [1,2].

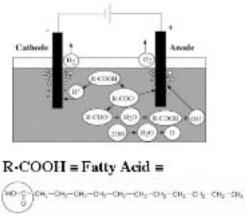


Fig. 11. Ionic model of the Linseed oil conductivity [1,2].

Fig. 11 shows our proposed ionic model of the Linseed oil conductivity [1,2]. The Linseed oil is usually polymerized. However, it is hydroscopic and it has many impurities, for example, Fatty Acid molecules (R-COOH). The proposed sequence is as follows: (a) R-COOH + potential  $\rightarrow$  H<sup>+</sup> + R-COO<sup>-</sup>; (b) either R-COO<sup>-</sup> ion delivers the charge to anode and R-COO returns to the fluid, or R-COO<sup>-</sup> ion transfers a charge to OH ion via a reaction R-COO<sup>-</sup> + H<sub>2</sub>O  $\rightarrow$  R-COOH + OH;

R-COOH returns the fatty acid back into the cycle and OH transfers the charge to anode; (c) H<sup>+</sup> ion delivers the charge to the cathode, where it forms an H<sub>2</sub> molecule and escapes; (d)  $2OH \rightarrow H_2O + 2O$ , and  $2O \rightarrow O_2$ , which delivers oxygen near the anode. Oxygen at the anode could react with a graphite layer. The relevant point is that water modulates conductivity. If we remove water, R-COO- or OH- will only deliver the charge, R-COO will just plate on anode, but it will not return R-COOH back into the current forming cycle, i.e., the current will slowly stop.

The most important conclusion of this model is that water has to be added right from the beginning of the experiment, otherwise unwanted molecules will plate at the electrode boundaries and the resistance will increase, and probably it will not return to its original value once water is added.

One inevitable consequence of adding water is a possible increase in the rate of the Freon-based chemistry. This has to be thoroughly tested. Perhaps, one should think about Oxygen-based electronegative additives.

# IV. RESULTS OF SMALL TESTS

In the following we want to show examples of author's small test results, which support the proposed model [1].

Fig. 12 shows author's measurement showing that the Bakelite volume resistance increased by a factor of ~3 after one week of heating at 40°C. It does not depend on whether the Bakelite is coated by the Linseed oil or not. The accumulated charge density due to the current monitoring is negligible in this test. The explanation for this result is that water is evaporated from the outer Bakelite layer, causing its resistivity to increase. The Bakelite samples were test samples never used in BaBar.

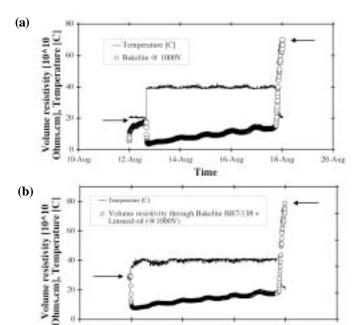


Fig. 12. The Bakelite volume resistance can be increased by a factor of ~3 by a simple exposure to 40°C for a week [1]. It does not matter if the Bakelite is covered by the Linseed oil or not.

Time

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Fig. 13 shows that the Bakelite volume resistance can increase by a factor of ~60 by accumulating a charge density of  $\sim 0.2 \text{C/cm}^2$  while the sample is in dry N<sub>2</sub> atmosphere of less than 200ppm of water. On the other hand, a sample in a wet atmosphere of 60-70% rel. humidity increased its resistivity by a factor of ~5 only. The Bakelite samples were the new type of Bakelite from the Nov., 2000 production.

Fig. 14 shows that the Bakelite volume resistance can be modulated by the presence of water. It appears that a very large humidity of more than ~50% of rel. humidity is needed to keep the Bakelite resistance within its original value if one reaches the accumulated charge density of 0.2-0.3 C/cm<sup>2</sup> [1]. Keeping the Bakelite-based chamber at a humidity level of less than a few hundred ppm of water is not enough to keep the resistance constant. The process may not be 100% reversible and some residual resistivity offset may develop.

One should mention that other people independently reached the same conclusion as the author, i.e., that the Bakelite volume resistance could increase [6,7]. However, this paper and earlier Ref. 2 show a possible modulation by water for the first time, and also provide an explanation of this effect.

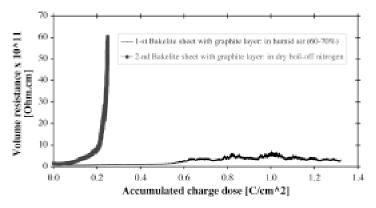


Fig. 13. The Bakelite volume resistance can increase by a factor of ~60 by accumulating a charge density of ~0.2C/cm<sup>2</sup> while the sample is in a dry N<sub>2</sub> atmosphere of less than 200ppm of water. On the other hand, a sample in wet atmosphere of 60-70% rel. humidity increased its resistivity by a factor of only ~5 [1]. The test uses the new Bakelite with the new Linseed oil treatment.

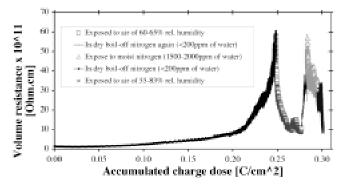


Fig. 14. The Bakelite volume resistance can be modulated by the presence of water. It appears that a very large humidity is needed to keep the Bakelite resistance within its original value if one reaches the accumulated charge density of 0.2-0.3 C/cm<sup>2</sup> [1]. The test uses the new bakelite with the new oil treatment.

The Linseed oil resistivity is not a fixed constant either. This can be see in Fig. 15 and 16, which show that the Linseed oil resistance is increasing as one accumulates the charge dose. This is certainly true for liquid oil, and very likely for oil, which did not have a chance to polymerize (it is sticky). However, we argue that even polymerized oil has water and Fatty Acid impurities, which are responsible for the ionic exchange. If water is not present, the current would stop.

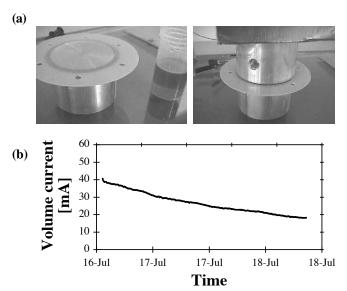


Fig. 15. (a) A setup to measure the volume resistivity of liquid Linseed oil. (b) One can see a resistivity increase just in a few days (data taken at 1kV and 24°C).

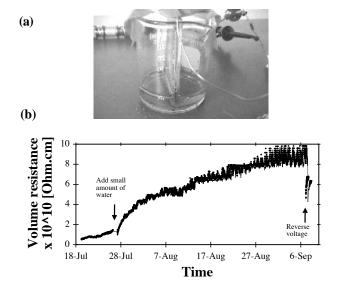


Fig. 16. (a) A different setup to measure the volume resistivity of liquid Linseed oil. (b) Resistivity increase as a function of charge. The electrode area submerged in the oil is ~1.5cm<sup>2</sup>.

However, we found another disturbing effect with the Linseed oil. We scraped some oil present in the "over-oiled" first generation RPC chambers, which operated in BaBar for several years. We found that the volume resistivity of this "brown sticky" oil is substantially lower by a factor of ~30-40

 $(\rho_{V-Brown\ sticky\ oil} \sim 2.1 x 10^8\ \Omega cm$  compared to a nominal value of  $\rho_{V-Fresh\ Uncured\ Italian\ Linseed\ oil} \sim 7.6 x 10^9\ \Omega\,cm)$  [1]. This effect might be explained by a Freon-based chemistry inside the operating RPC. Lowering the resistivity of the Linseed oil covered buttons and at the same time increasing the resistivity of the Bakelite near the buttons showing the sparking (see Fig.7c) could create the local inefficiency spots near the buttons observed in Fig. 6 and 7. These spots are observed in both the original and the new "Nov. 2000" RPC chambers in BaBar.

## V. CONCLUSION

The paper presents a model of ionic conduction in the Bakelite-based RPC detectors. This model explains why these detectors need to add water while they operate. The current, according to the presented model, is formed mainly by the ionic sequence involving the phenol impurities left in the Bakelite and water, both present at the Bakelite initial production. As long as these two ingredients are present in the original amount, the Bakelite resistance is constant. If either water or phenol impurity are depleted from various reasons, either due to a large current or operating in dry gas, the Bakelite resistance increases. According to the presented model, water should be added right from the beginning and not half way through the experiment. Similar ionic process is present in the Linseed oil polymer, where the current is carried by the fatty acid molecular impurities and water. The Bakelite RPC operation requires the entire ionic sequence to operate smoothly to keep the electrode resistance constant.

An increase of the Bakelite electrode resistance by a factor of ~20 has already a significant effect on the RPC efficiency.

The graphite on anode can react with released oxygen, and this may cause a loss of the electrical contact if the graphite layer is too thin.

The Linseed oil can dramatically reduce its resistance in the presence of the Freon-based chemistry, modulated by a UV light from the avalanches. This may be a significant effect near buttons where sparking may occur. The local Bakelite resistivity in those areas may increase. Both effects can work to lower the local efficiency near the buttons, which is observed.

Therefore, the applications planning to use this technology should perform appropriate R&D to show that they are not sensitive to these phenomena.

In any case, it is now already clear to almost all that the Bakelite electrodes are not simple fixed resistors as was originally assumed.

## VI. ACKNOWLEDGMENT

I would like to thank my friend J. Maly, a good chemist, for constructive critique of several ideas presented in this paper.

# VII. REFERENCES

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