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# Triboelectric charging of a glass bead impacting against polymers: antistatic effects in glass/PU electrification in a humidity-controlled environment

Ugo Lesprit<sup>1,2</sup>, Thierry Paillat<sup>1</sup>, Nouredine Zouzou<sup>1</sup>, Anna Paquier<sup>2</sup>, Marc Yonger<sup>2</sup>

<sup>1</sup>Institut Pprime, CNRS UPR 3346, Université de Poitiers, ISAE – ENSMA, 86962 FUTUROSCOPE France

<sup>2</sup>Saint-Gobain Research Paris, 39 quai Lucien Lefranc, 93300 AUBERVILLIERS France

ugo.lesprit@univ-poitiers.fr

## Abstract.

A single impact triboelectric charging experimental setup has been used to investigate the antistatic effect of two additives coated at the surface of beads on impact charges resulting from impact between a glass bead and a polyurethane (PU) target, at various additive concentrations and two different RH conditions. Three configurations of additives have been studied exhibiting different behaviours with RH, underlining the importance of humidity conditions in antistatic properties. Results for additive A, an organic oil, showed almost no impact charge decrease at RH 1% while results for additive B, a quaternary ammonium salt, presented no difference between RH 1% and 45%. This behaviour could be explained by a difference in surface water desorption dynamic when placed in a low humidity environment. In a second part, additional measurements analysing the effect of RH and temperature in the same glass/PU impact configuration in absence of additives are discussed. No significant variation of impact charge was observed on glass/PU triboelectric charging, confirming that previous results were indeed related to the presence of additives at the surface of the beads. A comparison with another triboelectric pairing, glass/PTFE, also shows the likeliness of different charge transfer mechanisms being involved in glass/PU and glass/PTFE as well as a general effect of temperature on impact charge which could be connected to relaxation mechanisms.

## 1. Introduction

The phenomenon of contact electrification, also commonly referred as triboelectric charging, describes any situation where a contact between two solids causes electrostatic charging of their surface around the point of contact. It is a big concern for many industrial purposes in particular when small particles are involved, because they are often subjected to a large amount of collisions, either between particles or with walls of the system [1, 2]. In some cases, these collisions can lead to the generation and accumulation of a large amount of electrostatic charges that can bring about undesirable consequences, such as electric discharges, wall adherence and/or material alteration [1, 3].

Despite having been studied for centuries and some important progress being made regarding its understanding, triboelectric charging is still proving to be a challenging subject nowadays with active debates on some of its most fundamental aspects [1-6]. The charge transfer mechanisms responsible for contact electrification is a recurring question, main theories behind divided between electronic transfer, ionic transfer and mass transfer [2, 6]. One obstacle that has been difficult to overcome in studying triboelectric charging is that it is heavily dependent on local physicochemical contact surface conditions, which can be hard to determine, control or reproduce in the frame of experiments and explains the lack of a general theoretical framework to describe triboelectric charging. It is indeed a complex phenomenon depending on many parameters and experimental works on the subject commonly encounter reproducibility challenges, with notably contradicting results being observed in some cases [5-7].

To address this problem, an important focus has been placed on developing more robust experimental setups with emphasis on controlling as thoroughly as possible the contact conditions between the triboelectrically charged surfaces [1]. A first approach to experimental research on triboelectric charging has been focused on multiple impacts experiments such as pneumatic conveyers or fluidized

beds because they emulate more closely industrial processes [8-11]. However, this type of design can be less appropriate to investigate fundamental mechanisms of triboelectric charging. A second approach which offers ideal conditions to achieve that consists in generating a single contact between surfaces under controlled conditions. Various technics have been used to initiate contact or friction between the solids [12-14], culminating in recent use of Atomic Force Microscope (AFM) in order to access extremely fine resolution [15, 16]. Amongst these experiments, an approach in particular is to propel a spherical sample towards a target providing relatively simple contact geometry for physical modelling and a good control over impact conditions for high resolution measurements [17-21].

It is well established that humidity has an important interaction with contact electrification, however the fundamental relationship between these two phenomena is not properly understood [1, 2, 6]. It has been shown that relative humidity (RH) in an environment is connected to the thickness of water layers adsorbed on solid surfaces, which can significantly modify the electric conditions around the point of contact during triboelectric charging [22]. On one hand, there are reports concluding that RH mostly affects charge relaxation mechanisms through surface water layers inducing a contact charge decrease, which has been observed in experiments when RH is increased over a threshold [23-25]. On the other hand, some articles have hinted at RH also affecting charge transfer mechanisms directly [6]. Notably, reports have shown in experiments that triboelectrically generated charge can increase with RH between 0 and 40-50% [26-28]. These results have been associated with ionic transfer as ion exchange models often rely on water to mediate the charge transfer [6].

As material properties hold an important role in triboelectric charging, the local contact conditions can also largely influence results of contact electrification during an experiment. This observation has been used widely as a mean to tune the quantity of charges generated during contact by altering the surface of a sample or coating it with other materials [5, 26, 29]. In some applications, it can indeed be very useful to either increase the triboelectric charge (electrophotography, electrostatic painting, electrostatic separation...) or decrease it for security or quality purposes for instance (food industry, pharmaceutical industry...) [1]. A wide array of coating materials has been investigated for these purposes, such additives including ionic dyes, polymer resins or quaternary ammonium salts [5, 26, 29]. It is not fundamentally understood how antistatic additives interact with triboelectric charging but a frequently referred mechanism for some additives associate the triboelectric charge decrease with an increase of surface conductivity due to water adsorption promotion from the atmosphere [2, 30-32]. Baytekin et al. have recently reported a different mechanism where radical scavengers also work to reduce the triboelectric charge on some polymers because radicals seem to be important to stabilize the charge after contact, showing that antistatic effects are not necessarily linked with water [33].

We have recently reported the development of a novel single impact triboelectric charging experimental setup capable of measuring the impact charge generated during the impact of a dielectric bead and a polymer target under carefully controlled experimental conditions [21]. During the experiment, the bead is pneumatically propelled toward the target using an air gun and various measurement electrodes provide the results. In this work, we use the experimental setup in order to investigate the antistatic mechanisms of two additives over single impact triboelectric charging of glass against polyurethane (PU), under various relative humidity conditions (RH). The effect of relative humidity and temperature over impact charges generated are also explored for this pair of material and compared with their respective role in glass/polytetrafluoroethylene (PTFE) charging to enable more thorough discussion.

## **2. Material and methods**

The operating diagram of the experimental setup is represented in Fig. 1. Inside of a metallic structure acting as a Faraday cage (1), a glass bead (5) is propelled towards a target (2) using an air jet in order to measure the triboelectric charge generated by the resulting single impact between the bead and the target. The trajectory of the bead first goes through a tube-shaped Faraday cage (5) at the bottom of the

structure in order to measure its initial charge  $Q_i$  ahead of impact. The electrostatic charge left after impact on the target  $-\Delta Q$  is then measured using an electrode placed right behind the target while the final charge of the bead  $Q_f$  is measured simultaneously with a metallic container (3) inside of which the bead falls down after impact. The three results are used during a test to verify charge conservation  $\Delta Q = Q_f - Q_i$  as a validation process. Impact parameters such as impact velocity and impact angle relative to the target plane are obtained from optical measurements made with a high-speed camera recording the movement of the bead inside of the Faraday cage at 3000 frames/s associated with the commercial LaVision software DaVis. The resulting data is then systematically exploited using a code developed in the laboratory in Python to calculate the parameters of interest. The pressure of the air jet provides control over the impact velocity of the bead and the target can be oriented to change the impact angle. The results presented in this article have been obtained for normal impact velocities ranging from 4 to 11 m/s, at an angle  $\theta_s$  of  $10^\circ$ . The whole setup is placed inside of a confined environment as a mean to regulate RH.

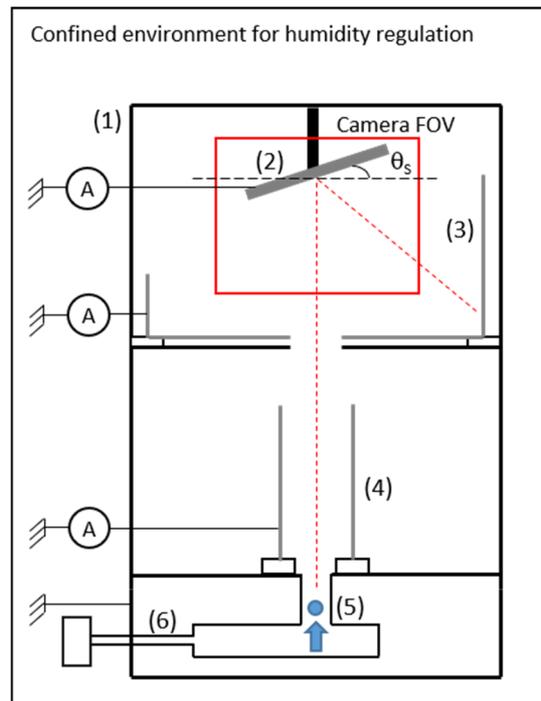


Figure 1 : Schematic representation of the experimental setup. The camera Field of View (FOV) is presented in red. (1) Main Faraday cage (2) Target and its measurement electrode (3) Reception container / Final charge electrode (4) Initial charge electrode (5) Glass bead (6) Pneumatic launcher.

The samples are spherical soda lime glass beads of diameter  $4 \text{ mm} \pm 0.02 \text{ mm}$  used with PU plates of thickness 5 mm for the target. The beads are prepared before each test by placing them into pure ethanol for a duration of 10 minutes and then applying a  $1400^\circ\text{C}$  flame on their surface for less than a second. Targets are washed before each set of tests with soapy water and rinsed abundantly with pure water afterwards, so as to avoid more aggressive detergents due to the sensitive nature of polyurethane (PU). The technical aspects of the experimental setup such as measurements and the different protocols used were reported in a previous article which should be consulted for more extensive details [21].

In order to obtain reproducible results, test conditions are investigated throughout statistical series of tests performed on 20 beads minimum and discussion is made over the average impact charges measured under specific conditions as a function of normal velocity, based on previous results obtained with the experimental setup [21]. These results have shown that the impact charge measured varies linearly with the normal velocity at impact, which is used as a reference in order to eliminate the

impact charge variation due to normal velocity dispersion over different tests. Thus, for each test condition, average values of tests will be presented as a function of normal velocity for tests performed at identical pressure setting. In addition, tests have shown that the impact generated during glass/PU triboelectric charging is limited by a maximum charge value dependent on normal velocity, which indicates that relaxation mechanisms can be essential to study this configuration [21].

Two additives have been studied in the span of this work. They will be labelled additive A and additive B in this article for confidentiality purposes. They are respectively an organic oil and a quaternary ammonium salt, both being generally used for their antistatic abilities. In order to better understand the role of each of these additives in reducing the electrostatic charges generated by triboelectric charging between glass and PU, multiple statistical studies were performed focusing on three different configurations of additives: the additive A only, the additive B only and both additives used simultaneously. For each configuration, three concentrations of additives were studied corresponding to a relative mass of 0.01, 0.02 and 0.05% of the bead mass in addition to a control test without any additive. The bead was coated with the additives using the following protocol:

- The beads are first sprayed with pure ethanol in order to clean them, and left under ventilation until the ethanol completely evaporates.
- A solution prepared by mixing the additive with pure water is then placed inside a mixer with multiple beads.
- The system is then rotated until no liquid droplet is visible in the mixer.
- The beads are finally placed inside of an oven at 60°C for 1h to remove all residual water.

During studies, because antistatic effects were expected to be influenced by humidity conditions, the tests were carried out at two different RH conditions, 1% and 42%. Each bead coated with the additive was only used once for a single test inside of the setup. Two methods were employed to regulate humidity conditions inside of the experimental setup:

- Injection of compressed air circulated inside of a dryer filter filled with desiccant salts ( $\text{CaSO}_4$ ) in order to reach low RH values below 5%.
- Placement of a saturated salt solution inside of the confined environment, stabilizing RH values around different values depending on the nature of the salt between 22 and 55% (potassium acetate, magnesium chloride, potassium carbonate and magnesium nitrate were used).

To ensure homogeneity of the RH value inside of the experiment, a fan was activated in combination with both regulation methods, using a minimum stabilization time of 8 hours for each set of conditions before performing experiments. The measurements are taken with a probe placed right outside the Faraday cage in the confined environment. Preliminary verifications have confirmed homogeneity of RH. Due to the relaxation time of 5 minutes between each test and the short opening time of the electromechanically operated valve, the amount of air injected inside of the system for each test only accounts for a small RH variability causing a standard deviation of +/- 1% in a single set of tests. No test could be performed at RH values over 55% because the electric insulation of the different electrodes falls apart at those values. Additional precautions would be necessary to allow such measurements.

Temperature was not regulated inside of the experiment with the present setup. For each set of tests made at specific conditions, tests were performed within temperature fluctuations of +/- 0.6°C at most to allow relevant comparison between results. The maximum temperature difference investigated was due to temperature variations over a year in a seasonal temperate climate, leading to measurements between roughly 21 and 28°C.

### 3. Results and discussions

#### 3.1. Effects of antistatic additives on contact electrification

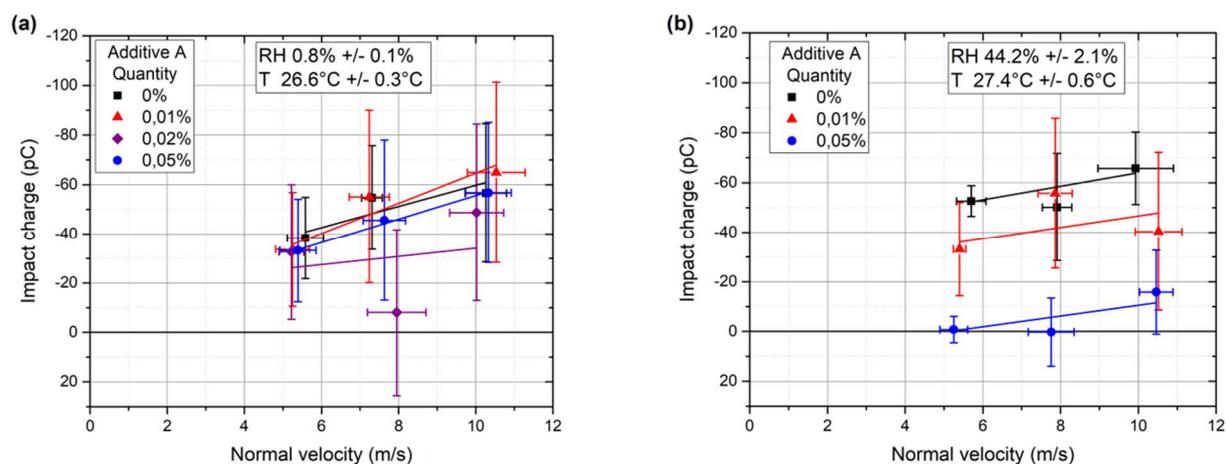


Figure 2: Impact charge as a function of normal velocity and relative mass of additive A at (a) 0.8% average RH and (b) 44.2% average RH.

Results obtained for the first configuration with beads coated with additive A only are shown on Fig. 2 (a) and (b) at 0.8 and 44.2% RH respectively. The average impact charge measured for a set of tests at various pressure settings is represented as a function of normal impact velocity relative to the target. The glass beads predominantly show a negative charge when impacted on a PU target, with or without additives on the surface of the bead. Only the tests performed with beads coated with 0.05% concentration of additive A at RH 44.2% show a large impact charge reduction in this configuration. The beads coated with 0.01% concentration at same RH also show an intermediate behaviour with a smaller charge reduction when compared to the control test results (unfortunately, the 0.02% test condition could not be obtained for this set of experiments due to experimental problems). In the case of low RH however, no significant reduction of impact charge due to additive A is visible. For both RH conditions investigated, impact charge measurements also tend to show a larger standard deviation relative to average impact charge values when only additive A is coated on the beads. This is believed to be a consequence of inhomogeneity of the additive layer over the surface of the bead, potentially caused by dewetting of the additive after coating.

Similar tests were performed with additive B and are presented in Fig. 3. The impact charge is seen to decrease importantly for the 0.02% and 0.05% relative masses of additive B, confirming antistatic effects with the ammonium quaternary salt. In this case however, the antistatic behaviour seems to be unaffected by humidity conditions. In addition, the average impact charge value measured is lower for additive B than for additive A at equivalent additive concentration, indicating that additive B is more efficient in reducing the impact charge. Results also show a significantly smaller standard deviation of measured values relative to the standard deviation of results observed with additive A in Fig 2.

For both additives, the diminution of average impact charges with the addition of more additive coated on the beads displays an intermediary result for the 0.01% data points. The corresponding gradual increase of antistatic effect suggests that surface additive coverage or additive layer thickness may be governing factors in their ability to reduce the measured impact charge.

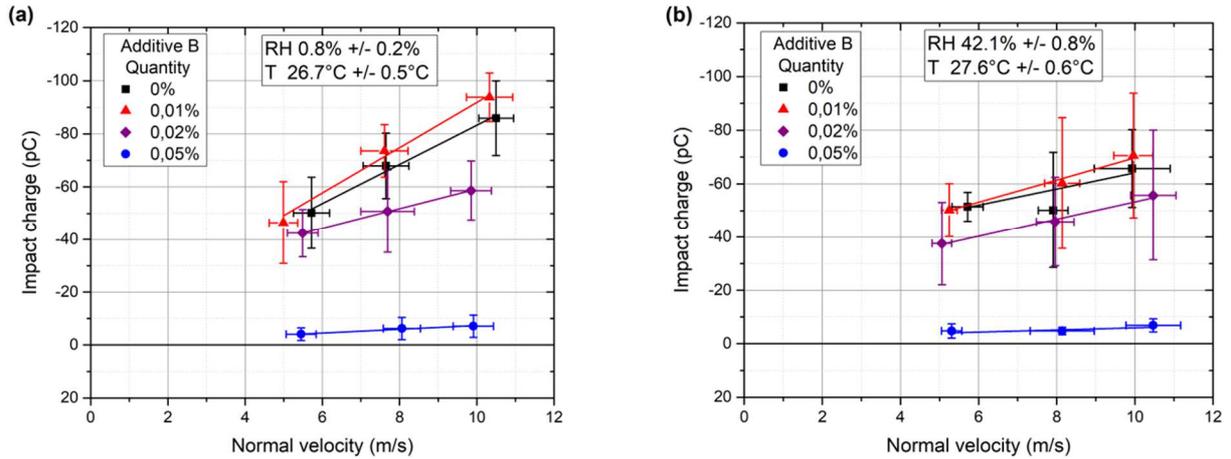


Figure 3: Impact charge as a function of normal velocity and relative mass of additive B at (a) 0.8% average RH and (b) 42.1% average RH.

In the last configuration, a mixture of the two additives was coated on the surface of the beads. Impact charges measured are visible in Fig. 4 (a) and (b). The observed effect of the mixture on impact charge is more similar to the effect of additive B, with an important decrease of impact charge at both humidity conditions. The level of charge measured during these experiments is the lowest of the three configurations on average, showing that this configuration is the more efficient for antistatic purposes.

The polarity of the charge is also inverted on the 0.02 and 0.05% series at low humidity, a phenomenon that could be seen for a few tests with additive A results at low humidity and seems to be occurring more steadily with the mixture of additives. A potential hint to interpret this phenomenon can be found in relatively recent research from Baytekin et al. [34]. They have shown that in the case of some polymers, triboelectric charges were not distributed homogeneously over the contact surface, but rather forming a configuration of smaller regions of alternating polarity. In light of these findings, the presence of additives could interact locally with other parameters such as roughness, residual contaminations or residual charges to modify the balance of negatively and positively charged regions and induce polarity reversal. The fact that positive impact charges are more consistently measured for RH 0.9% than RH 41.7% suggests that adsorbed humidity on the surface could also be important in the competing physical mechanisms responsible for polarity reversal.”

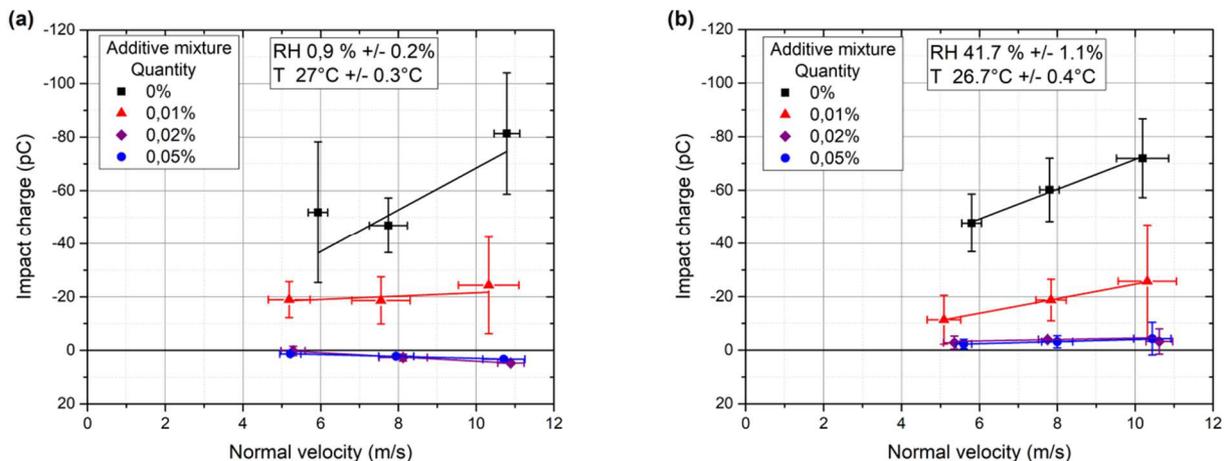


Figure 4: Impact charge as a function of normal velocity measured and additive mixture relative mass at (a) 0.9% average RH and (b) 41.7% average RH.

In addition to reducing the average impact charge measured to lower values, the mixture of additives achieves this result with a lesser additive mass compared to the additives used separately, with the impact charge for 0.01% samples already significantly smaller than the control test and the maximum antistatic effect being reached with the 0.02% set of beads. The 0.02 and 0.05% sets for the additive mixture also show the smallest dispersion with almost no deviation from the linear regression, even at low humidity where additive A alone cannot explain an impact charge decrease. Hence, the use of both additives simultaneously enhances the antistatic effect of additive B at low additive mass. One hypothesis is that additive A could increase the coverage of additive B on the beads making it more homogeneous and explaining the appearance of the antistatic effects with less additives.

The most important aspect of these results is that they are observed in a configuration where a very short single impact of roughly 50  $\mu$ s takes place between the two surfaces. Most experiments studying the effects of additives over triboelectric charging have done so over longer contact times or numerous impacts between many samples. The antistatic effects displayed during tests performed with this experimental setup thus confirm that the reduction of impact charge is occurring in the span of a single contact/separation process. The fact that no significant impact charge decrease is observed for additive A only at low RH suggests that the presence of water layers at the surface of the bead - which disappears in large proportion when the relative humidity is reduced to 0.8% - are significant in the antistatic mechanisms. On the other hand, the two other configurations both involving additive B show a strong antistatic effect independent of humidity conditions in the experimental setup. This result could indicate that the presence of water is irrelevant to the antistatic effect of additive B. Another hypothesis would be that additive B is capable of maintaining water adsorption at the surface of the bead at low RH for a longer time than the stabilization time of experiments. Reports have associated antistatic effects of ammonium quaternary salts with increased water adsorption on surfaces, which would support this second hypothesis [32]. Following this, coatings with additive B may have still some adsorbed water at the end of the stabilization time (of at least 8 hours) before each set of experiment conditions, while coatings with only additive A may not, leading to different antistatic effects.

### *3.2. Investigation on humidity and temperature interaction with triboelectric charging*

The studied additives have exhibited some antistatic effects over impact charging of the glass beads, but these effects are dependent on humidity conditions. To delve deeper into the physical mechanisms responsible for the antistatic behaviour, it is thus essential to better understand the influence of RH conditions over glass-PU charging and triboelectric charging in general. In addition, RH conditions are also dependent on temperature, hence it is essential to examine its role as a parameter in impact charging.

To achieve that, tests were first performed at various RH conditions in absence of any additive. These results were compared to measurements made against a PTFE target to provide a reference. Fig. 5 shows the impact charge values measured as a function of normal velocity for a PU target and a PTFE target at various RH values between 1.5% and 54%. No variation of impact charge is observed with RH for the glass/PU interaction between 5 and 45% RH, with no apparent correlation of impact charge with RH. This confirms that the impact charge decrease measured in presence of additives was entirely due to their antistatic effects. On the contrary, in the case of PTFE, the impact charge increases between RH of 1.5% and 30%, with a saturation of impact charge value over 30%.

The increase of impact charge against the PTFE target at low RH values must be interpreted either as a charge transfer mechanism effect or a charge relaxation effect. Charge relaxation effects are not expected to be responsible for a charge increase because increased surface conductivity at higher RH generally leads to a charge decrease [23-25]. The impact charge increase is thus more likely connected with a charge transfer mechanism instead. Most ionic transfer theories have water adsorption as a fundamental component of charge transfer and this could be responsible for the behaviour of the

impact charge measured on glass/PTFE at low RH, for example through hydroxide ions transfer [6] or a transfer of the mobile ions generally present at the surface of soda lime glass materials. Indeed, the surface water layer is importantly reduced at low RH because of the low surrounding air water vapor content. The fact that the impact charge does not converge towards a lower value at RH 1.5% is either due to the extreme difficulty to achieve complete desorption of water on the surface of a solid [35], or that there are other competing charge transfer mechanisms acting simultaneously which are not impacted by water.

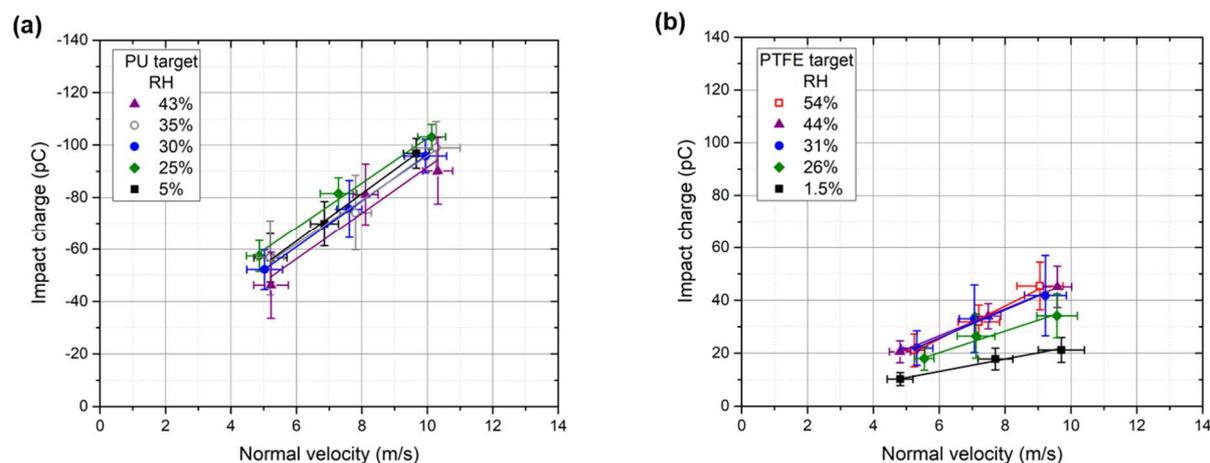


Figure 5: Impact charge measured as a function of normal velocity at different RH conditions for (a) a PU target and (b) a PTFE target. The temperature for these experiments is  $22^{\circ}\text{C} \pm 2^{\circ}\text{C}$ .

The absence of impact charge variation in the case of the PU target on the other hand suggests that the active charge transfer mechanisms in glass/PU charging are partly different than those involved in glass/PTFE charging. As mentioned before, a maximum impact charge limit dependent on normal velocity has been observed and previously reported for glass/PU charging, hinting at the importance of relaxation mechanisms in the measurements performed in that case [21]. It is possible that the aforementioned limit could hide any increase of impact charge with RH if the charge has already reached the limit at low humidity. Nonetheless, there seem to be no significant effect of RH at all on the statistical series of measurement, with the slope of linear regressions being highly similar between the different set of tests and average values changing within the error bars of the different experimental conditions tested. In the previous study on glass/PU, impact charge variations were seen at low tangential velocity values indicating that the maximum limit was not entirely reached at a setting angle of  $10^{\circ}$ , thus the decrease of impact charge at very low RH should be observable if it had any physical relevance in spite of the maximum limit. These points support the idea that the absence of effect of RH over triboelectric charging between glass and PU is due to different charge transfer mechanisms compared to the results on glass/PTFE.

This conclusion is strengthened by water affinity measurement on both target materials which show a very similar contact angle for PTFE and PU of  $110^{\circ}$  indicating that the two materials are strongly hydrophobic. Experimental conditions between the two configurations are rigorously the same otherwise, so it suggests that the impact charge increase with humidity is not due to adsorbed water itself and indeed a consequence of charge transfer mechanisms. It is important to highlight that surface roughness properties are likely to influence of RH interacts with triboelectric charging and could partly explain the difference between glass/PU and glass/PTFE [36]. This issue should be examined in future experiments.

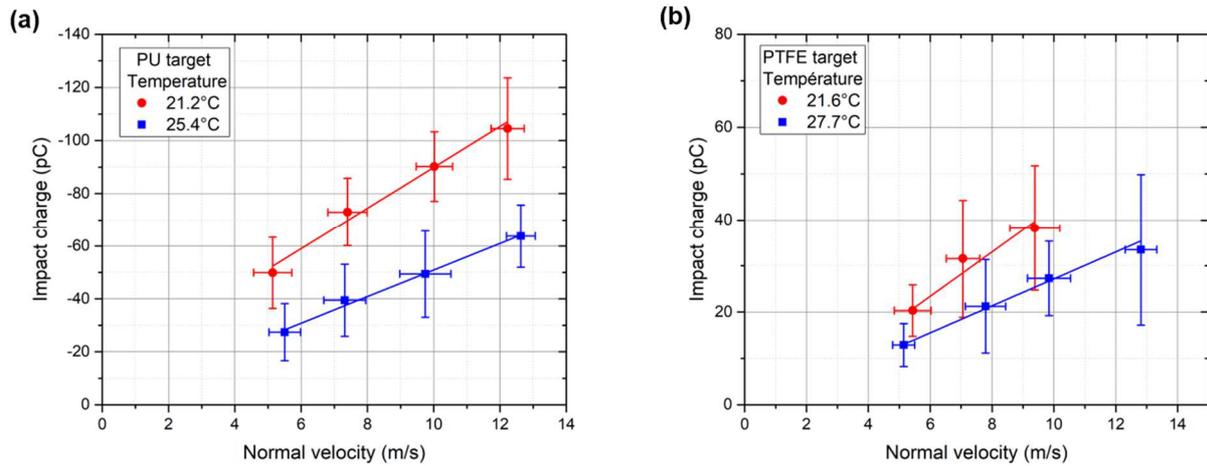


Figure 6: Impact charge as a function of normal velocity at two different temperatures for (a) a PU target at RH  $37\% \pm 0.5\%$ . and (b) a PTFE target at  $32\% \pm 0.5\%$ .

RH conditions are not independent of temperature, it is thus necessary to verify if the RH results observed are not affected by temperature change and if it is indeed an effect linked with air water vapor content. Fig. 6 shows the impact charge measured as a function of normal velocity for both target materials at two different temperatures. Despite the small temperature variation between the two series, a remarkable decrease of roughly 50% of the average impact charge measured is observed for both materials.

There are only few articles in literature investigating the links between temperature and triboelectric charging. Temperature variations are relatively small so it is unlikely that the impact charge decrease should be caused by the evolution of mechanical parameters such as rigidity. A temperature difference of a few degrees cannot account for a contact surface modification sufficiently large to explain the 50% impact charge decrease. Furthermore, an increase in temperature would cause a diminution of mechanical rigidity which would normally lead to a bigger contact surface, which is expected to influence the impact charge in the opposite direction. Thus, an electrical parameter explanation is more likely to explain the results observed in Fig. 6.

As temperature increase, a quick assumption would be to assume that more energy is available for charge transfer, which should lead to an impact charge increase. However, it is expected that charge mobility would increase as well. Thus, the impact charge decrease observed in these results could be caused by higher surface conductivity promoting recombination of charges during separation. Prior works on very different configurations have reported a similar impact charge decrease and associated the observed results with charge relaxation [25, 37].

Note that the temperature effect on triboelectric charging observed in these results cannot be accounted by a simultaneous variation of air water vapor content:

- RH variations showed no effect on glass/PU triboelectric charging while the influence of temperature is more important on glass/PU than glass/PTFE.
- The two parameters influence the impact charge in opposite directions. When the temperature increases at constant RH, it implies a more important water vapor content in surrounding air. Hence, increasing the temperature should affect impact charge the same way as RH while it is not the case.

This is why only two entirely different mechanisms can explain the observed effects of RH and temperature over triboelectric charging.

#### 4. Conclusion

The effect of two antistatic additives on contact electrification of glass beads against PU have shown remarkable results in a configuration of a very short single contact ( $\sim 50 \mu\text{s}$ ), depending on the mass of additive added on the surface of the beads. The antistatic effect of additive A, an organic oil, appears to be heavily dependent on RH conditions, with no impact charge reduction observed at low RH. On the other hand, additive B, a quaternary ammonium salt, shows a strong antistatic effect entirely independent of RH conditions in the framework of this study. The absence of relation between RH and charge decrease in the case of additive B needs to be investigated further in future work in order to determine if it is due to longer stabilization of water at the surface of the bead or if it is indeed independent. The antistatic effect is enhanced further when both additives are used conjointly, with antistatic effects appearing at a lower mass and achieving a lower impact charge in similar mechanical conditions. This suggests that the additive A helps achieving a more homogeneous and efficient repartition of additive B at the surface of the bead.

The effects of RH and temperature over triboelectric charging were also investigated in the case of glass/PU and compared with a second configuration glass/PTFE, in order to decorelate the action of additives over triboelectric charging from the action of water. These results are also important because little reports can be found in literature investigating RH and temperature on a single impact electrification configuration. No impact charge variation was measured at various RH conditions between 5 and 45% for glass/PU, while the impact charge measured in the case of glass/PTFE increased up to 30%. The stark difference in behavior with RH between the two material pairs is believed to be caused by the fact that at least two entirely different charge transfer mechanisms are involved in these cases. Note that no decrease of impact charge was observed at high relative humidity conditions, but it is planned to perform measurements at higher RH conditions than 50% with the experimental setup to complete the results obtained in this work. On the other hand, a temperature increase of only a few degrees caused an important decrease of impact charge of roughly 50% for both cases, which has been interpreted as an increase in the relaxation mechanisms during separation linked with charge mobility at the surface of both solids, which is connected to temperature.

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