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Triboelectrification of granular insulating materials as affected by dielectric barrier discharge (DBD) treatment

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ABSTRACT

The aim of this paper is to point out the influence of dielectric barrier discharge treatment on tribocharging of granular insulating materials. Particles of Polyvinyl Chloride (PVC) and Polypropylene (PP) were subjected to an AC dielectric barrier discharge (DBD) plasma treatment in ambient airprior to tribocharging in a vibratory device. The charge to mass ratio was measured for treated and untreated materials. Electrostatic separation of a mixture of granular materials (PVC and PP) to measure the effectiveness of DBD treatment was evaluated by processing treated and untreated PVC/PP granular mixtures in a free-fall electrostatic separator. The obtained results clearly indicate that DBD has the capability to influence surface charging proprieties of polymer granular materials. In case of short treatment time, typically less than 3 s, a marked increase in the charge to mass ratios was observed for both PVC (about 35%) and PP (roughly 45%). In the same way, the quantity of DBD-treated materials, recovered after electrostatic separation, was increased by about 104% and 30% for PVC and PP, respectively, as compared to untreated case. The DBD treatment time is a key factor to increase the tribo electric effect.

1. Introduction

Triboelectricity is one of the earliest observed, but also less understood electrostatic phenomena [1–3]. Until recently, triboelectrification of insulating materials was perceived mainly as a source of electrostatic hazard [4–6]. Nowadays, the triboelectric effect is more and more employed as a charging mechanism in several electrostatic applications, such as: separation of polymer materials [7–9], energy harvesting [10,11], triboelectric based sensors [12,13]…etc. In such applications, the charge acquired by the materials has a major influence on the outcome of the process. Therefore, it is very important to enhance the tribocharging capability of the processed materials.

Tribocharging occurs when two different materials are separated after having exchanged electric charges during their contact [14]. In the process, one material acquires positive and the other negative charges. Despite the important number of studies devoted to triboelectricity, the mechanism of charging by contact (and friction) between insulators seems to be still unclear. Indeed, several issues are still poorly understood, like the nature of the charge carriers exchanged during contact, the role of friction, of contact pressure, of ambient conditions and so on. In this day, triboelectrification of insulators is tentatively explained by three mechanisms: electron transfer [15], ion transfer [16] and transfer of charged material between the bodies in contact [17]. Understanding these mechanisms would help to improve, or inversely eliminate, the triboelectric effect.

The electrostatic separation of granular insulating mixtures is the result of the combined action of mechanical and electric forces [18]. Therefore, the triboelectric charge carried by the granules should be as high as possible so that the electric Coulomb force could overcome the mechanical and gravitational forces and consequently better separate the constituents of the mixture. Previous carried studies on electrostatic separators have already tried to improve the tribocharging devices and optimize their operation. One solution, for instance, is to increase the charging time and air speed in the case of fluidized bed devices [19–23]. However, the possibilities offered by the adjustment of such parameters are

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limited: a too long charging time would reduce the hourly quantities of treated material and a too high air speed would increase the energy consumption at levels unacceptable for industry application.

Several research groups have used plasmas to modify the tribocharging properties of polymers [24,25]. Most plasma systems require a pump and a specific gas to operate. DBD might be a better option, as it can be conveniently generated in atmospheric air, using very simple electrode geometries.

The present paper is aimed at evaluating the influence that exposure of granular insulating materials to a dielectric barrier discharge (DBD) might have on the triboelectric charging process. Thus, two granular polymers, PVC and PP, were submitted to an AC DBD before being tribocharged in a vibratory device. The tribocharged materials were later introduced in a free fall electrostatic separator to evaluate the effect of DBD treatment on the selective sorting of the two constituents of the granular mixture.

2. Experimental procedure

2.1. Materials

The experiments were performed on two granular insulators, Polyvinyl Chloride (PVC) and Polypropylene (PP). The materials used in this study were provided by RECYMAP, a French company specialized in plastic recycling. The aspect of the used PVC and PP granular materials is shown in Fig. 1. The mixture used in the separation experiments was composed of 50% PVC and 50% PP.

2.2. Dielectric barrier discharge-DBD plasma system

Surface treatment using a dielectric barrier discharge was carried out in a plasma reactor composed of two circular aluminum plate electrodes (diameter: 70 mm, thickness: 12 mm) each of them covered by 3-mm thick glass plate (150 mm × 150 mm); the air gap between the two glass plates: 5 mm. The samples were placed on the lower glass plate. The electrode configuration is supplied by a high voltage amplifier (model Trek 30/20A) (Fig. 2).

2.3. Vertically-vibrated bed device

The charging of untreated and treated insulating particles was carried out in a stainless-steel box connected to the vibratory device (Fig. 3). The amplitude of the vertical oscillations was 1 mm, at a frequency of 50 Hz. In the metallic box, charge transfer is due to particle-particle and particle-wall collisions.

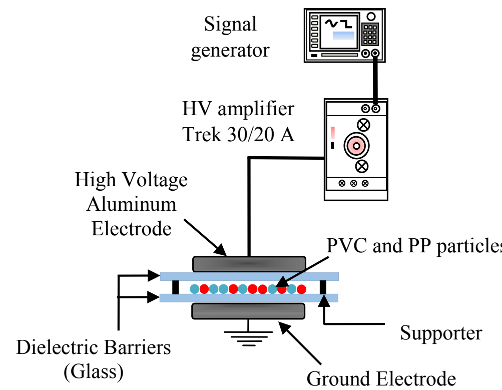

Fig. 2. Dielectric barrier discharge plasma system.

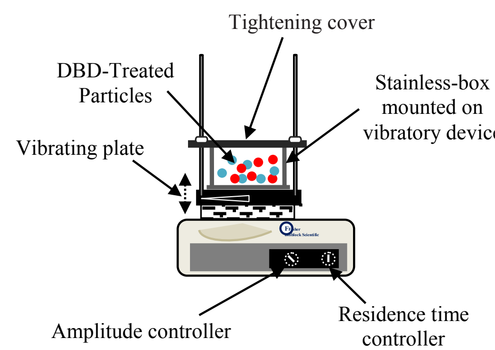

Fig. 3. Vibratory tribocharging device.

2.4. Free-fall electrostatic separator

The charged binary mixtures (either untreated or treated PVC and PP particles) were processed in a standard free-fall electrostatic separator under the action of the high intensity electric field generated by two vertical aluminum plate electrodes of opposite polarities connected at ± 30 kV, as shown in Fig. 4. The products were collected in three boxes placed at the lower end of the separator: two, adjacent to the electrodes, for recovering PVC and PP products, and one in the center, for middling.

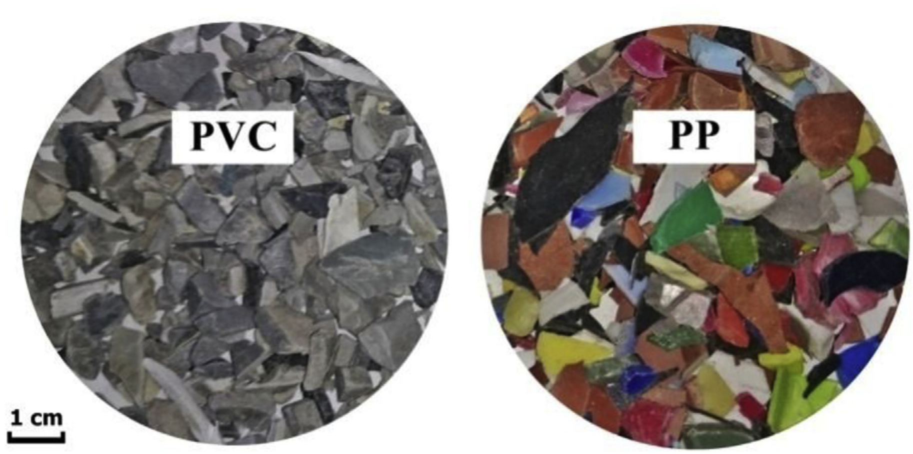

Fig. 1. Aspect of PVC and PP plastic particles.

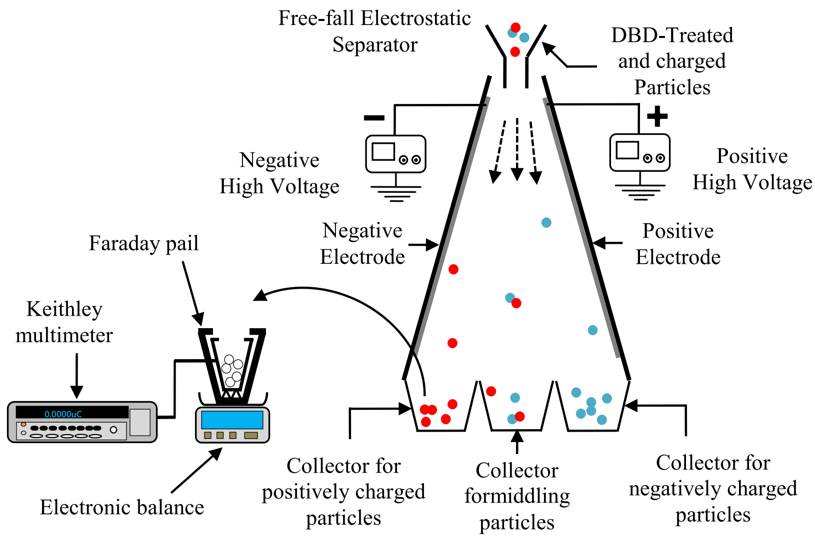

Fig. 4. Schematic representation of the free-fall electrostatic separator.

2.5. Methods

In a first set of experiments, 10 g samples of PVC and PP granules were charged separately in the vibratory device, for 4 min. The same amounts of PVC and PP granules were also separately treated in the DBD reactor, at 30 kV and 800 Hz, for several durations: 1s, 3s, 5s, 10s, 2min and 5min. The frequency was chosen such that to generate a homogeneous discharge and avoid the filamentary regime that becomes predominant beyond 800 Hz. At higher frequencies (tens of kHz), the plasma is likely to be too energetic for this particular application.

After treatment, the samples were then charged for 4min in the vibratory device. The charge of treated particles was measured before and after tribocharging with a Faraday pail, connected to an electrometer (model 6415, Keithley Instruments). In this way, it was possible to compare the charge generated by triboelectric effect with and without DBD plasma treatment.

The second set of experiments was carried out on a binary mixture composed of 30 g of PVC and 30 g of PP. Samples of this granular mixture were treated by DBD using the same AC high-voltage and frequency as before, and were maintained in the vibratory tribocharging device for the same duration (4 min). The charged granular mixtures were then introduced in a standard free-fall electrostatic separator. When separation was over, the mass and the electric charge of the two products recovered in the PVC and PP collecting boxes were measured. The charge acquired by the granules and the recovery rate of the two plastics were measured for both treated and untreated mixtures.

All the experiments were performed in ambient air (temperature: 21 °C–23 °C and relative humidity: 29%–32%) and triplicated. The electric charge of the samples was estimated by a Faraday pail connected to an electrometer (Keithley Instruments, model 6514), while the mass of the particles was measured with an electronic balance (resolution: 0.01 g).

3. Results and discussion

3.1. Triboelectrification of DBD-treated samples

The results of the tribocharging experiments are shown on Fig. 5.a and b, for PVC and PP granules respectively. In all but two of the experiments the range of the measured values was less than 10%. In those two particular cases, two additional experiments were performed and the average of five values is represented on the figure.

In all experiments, both PVC and PP particles acquire a negative charge when contacting the stainless-steel box walls. DBD treatment alone does not charge the granules significantly, whatever is the treatment time. This can be attributed to the alternating nature of the DBD; the charge communicated in half cycle is eliminated by the opposite charge in the next half cycle and so on. It can be concluded that AC-DBD plasma treatment does not have a direct influence on the surface charge.

Conversely, the triboelectric charge was enhanced as a result of DBD exposure. This suggests that DBD plasma produced a modification in particles surface properties. The results in Fig. 5 show that the DBD treatment time significantly influences the charge acquired by triboelectric effect. Indeed, in the case of 1s exposure to DBD, the charge of PVC granules increased from −64.44 nC for the untreated sample, to −76.31 nC after treatment, which represents an increase of more than 18%. As for the PP granules, the charge was increased from −18.19 nC for the untreated sample, to −42.79 nC after treatment, i.e. a charge increase by more than 135%. In the case of 5 min treatment time, the effect of the DBD is reversed; the charge of PVC decreases from −64, 44 nC for the untreated sample to the −5.5 nC. For PP, the charge decreases from −18.19 nC for the untreated sample to −10.28 nC.

From the results of Fig. 5, it can be concluded that the effect of DBD treatment is dependent on material but also on the treatment time. The increase of the treatment time has not a beneficial effect on the triboelectric charge. According to [26,27], granules exposure to DBD plasma at atmospheric pressure causes surface etching, as result of the breaking of some bonds and removal of molecular fragments from polymer surface. The etching effect is related to the density of the electric power transferred in the discharge and to the treatment time; it leads to the increase of surface roughness and H_2O molecules adsorbed at polymer surface, phenomena which are accompanied by increased wettability. The relative humidity seems to strongly affect triboelectrification mechanism of polymers [16,28]. Almost no contact electrification occurs at RH = 0%, it attains the maximum at about 30% and most often decreases at values beyond 40% [16]. Thus, modification of surface properties

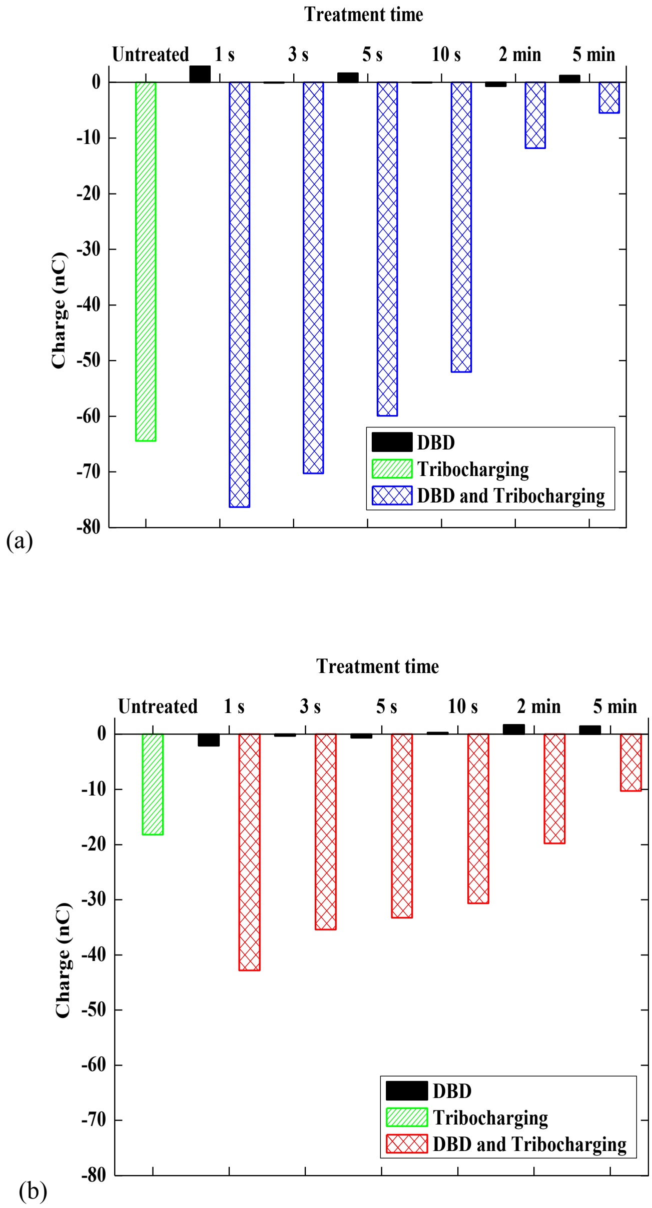

Fig. 5. Triboelectric charge of untreated and DBD treated particles: (a) PVC; (b) PP.

may facilitate material humidification at long time DBD treatment; under these circumstances the charge acquired by triboelectric mechanism may decrease, explaining the results given in Fig. 5.

The relative charge increase due to DBD treatment was computed as follows:

$$\Delta Q_t\,\% = (Q_{t\ (treated)} - Q_{0\ (untreated)})^{*}100/Q_{0\ (untreated)} \qquad (1)$$

where Q_0 is the triboelectric charge of untreated particles and Q_t the triboelectric charge after plasma exposure to a duration t. The Fig. 6 shows the variation of the charge increase as a function of DBD treatment time for the two materials PVC and PP.

The time parameter influences to a different degree the changes in surface properties observed for the two polymers. For both polymers, the relative triboelectric charge increase tends to diminish with increasing the duration of DBD treatment, and becomes negative for larger values of t. The positive values of ΔQ_t correspond to the situations in which the DBD enhances the

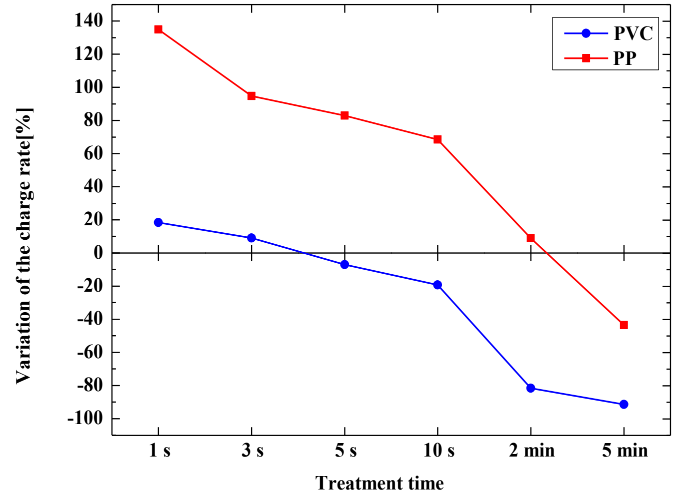

Fig. 6. Triboelectric charge variation rates of DBD plasma treated particles.

triboelectric charge. When $\Delta Q_t = 0$, it means that the DBD effect is null, the negative values of ΔQ_t correspond to the reversal of the DBD effect, i.e. the triboelectric charge is lower for the granules that were longer exposed to the plasma. The relative triboelectric charge increase of PP particles is higher than that of the PVC, for all DBD-treatment times. The time where the variation rate is zero corresponds to the maximum duration of the DBD exposure that causes positive effects. At longer exposure times, the surface modifications are accompanied by humidification that has an adverse effect on tribocharging. This critical value of DBD exposure time is about 4 s for the PVC and beyond 2 min for the PP granules. The difference between the DBD effects on the two materials is due to their different chemical structure, shape and surface roughness.

3.2. Electrostatic separation of DBD-treated mixture samples

The charge to mass ratios recorded for the two products recovered in the collector of a free fall separator after processing DBD-treated and triboelectricaly charged PVC/PP granular mixtures are shown in Fig. 7. The separation results as a function of DBD treatment duration are represented in Fig. 8.

When separately charged, the two materials, PVC and PP, are negatively charged. But, in the mixture the PVC has a negative

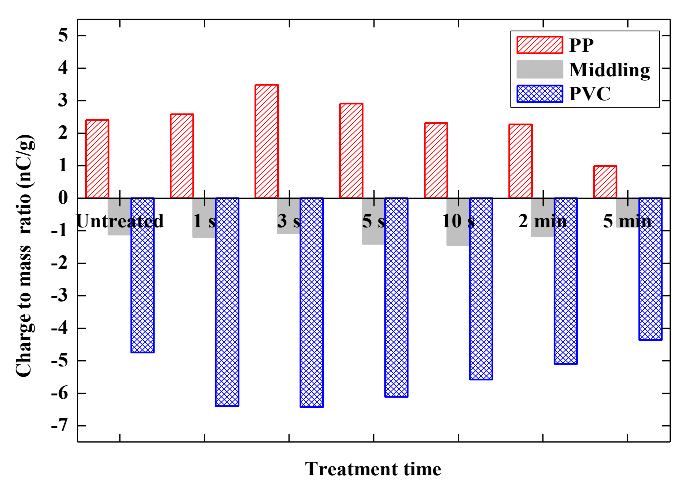

Fig. 7. Charge to mass ratio of products at different treatment times.

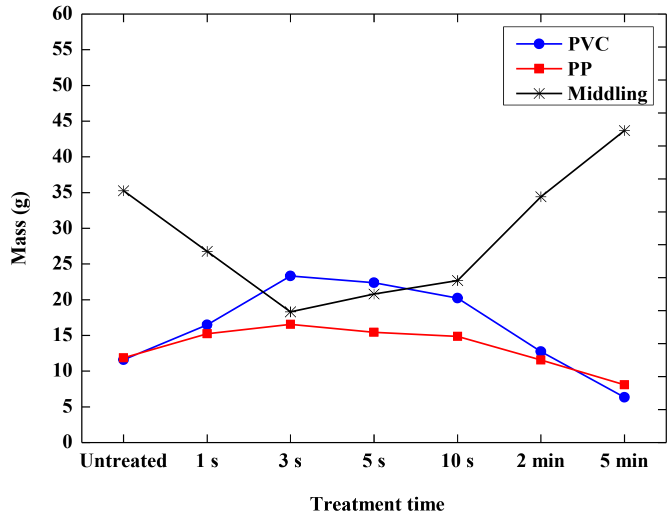

Fig. 8. Recovery of PVC, PP and middling products obtained after electrostatic separation from untreated and treated particles mixtures at different times.

charge and the PP a positive charge, because of contact between them. The separation results (Fig. 7) show that the effect of DBD treatment on the separation process is strongly related to the exposure duration. The maximum charge and best separation results were obtained at 3 s, a DBD treatment duration for which the recovered quantity was enhanced by more than 104% for the PVC and 30% for the PP, comparing to untreated case. However, at longer exposures, the influence of the DBD becomes less important, its effect on the tribocharging and separation may be even reversed (Fig. 8).

The composition of the middling product obtained in experiments with granular mixture at different treatment times is presented on Fig. 9. The purity was calculated as the ratio of the mass of pure material *mpc* and the total mass of the product *mtc* collected in a given box:

$$\text{Pur}\ (\%) = (\text{mpc}/\text{mtc}) * 100 \quad (2)$$

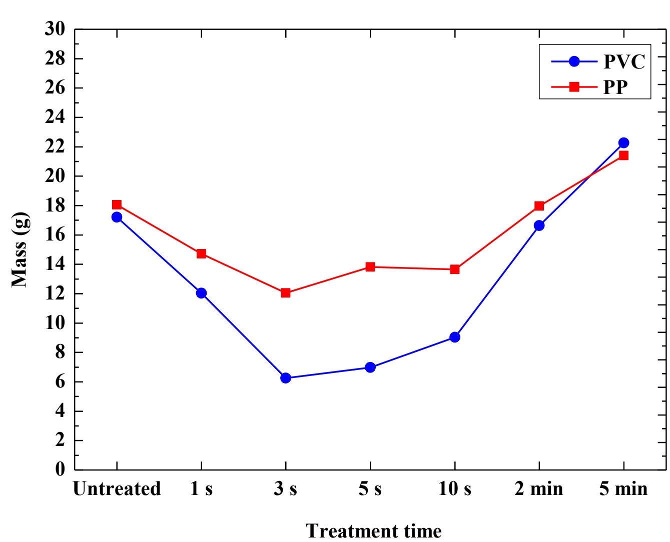

Fig. 9. Composition of the middling products (PVC and PP content) obtained after electrostatic separation from untreated and DBD treated particles mixtures.

Table 1
Purity (%) of the two recovered materials (PVC and PP).

	Untreated	1 s	3 s	5 s	10 s	2 min	5 min
PVC	79.12	79.63	78.95	81.75	80.71	81.31	82.3
PP	76.41	73.9	71.48	72.16	79.97	81.15	83.08

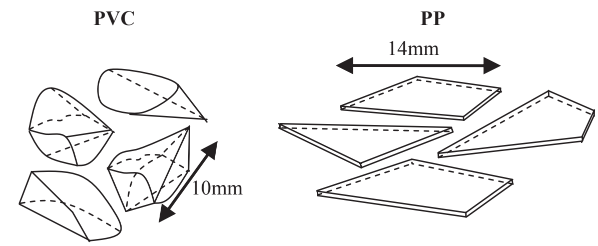

Fig. 10. Shape and size of PVC and PP plastic particles.

In the interval 3 s-10 s, the PP forms the most part of the middling product as it does not acquire higher charges, as the PVC (Fig. 7). However, outside this interval the quantities of the two materials in the middling become similar. This observation points out that the influence of the DBD is different from one material to the other and depends on the exposure time. The purity of the recovered products is not significantly modified by the exposure time (Table 1).

The difference between charging capability observed for the two materials (PVC and PP) can be attributed to their properties (conductivity, permittivity), chemical structure but also to their shape. Indeed, the particles characterized by different shapes and sizes have different contact areas with the plasma generated by the DBD, so they charge differently (Fig. 10).

No matter what the shape and size of the granules, their exposure to a DBD prior to triboelectric charging can improve the acquired charge and consequently the separation results if the exposure duration is appropriately selected. The obtained results indicate that a few seconds (less than 10) are sufficient to enhance the triboelectric effect.

4. Conclusions

DBD treatment of granular PVC and PP originating from industrial wastes influences the triboelectric charging and the electrostatic separation of the two polymers. Thus:

- Exposure of PVC and PP granules to a DBD prior to their charging by friction can strongly enhance the triboelectric effect.
- The duration of the DBD treatment plays a crucial role in the process. The best tribocharging effects and electrostatic separation results are obtained for relatively short exposures to DBD (typically 3 s).
- For treatment durations longer than 10 s, the effect of the DBD is reversed and the triboelectric effect is poorer.
- Under optimal operating conditions, the quantities of PVC and PP obtained from a DBD-treated granular mixture in a free fall electrostatic separator can be increased respectively by 104% and 30%, as compared with the untreated case.
- The effect of DBD treatment on the triboelectric charge varies from one material to the other.

References

[1] Herbert W. Meyer, History of Electricity and Magnetism, MIT Press, Cambridge, Mass, December 1971.

[2] L.H. Lee, Dual mechanism for metal-polymer contact electrification, J. Electrost. 32 (1994) 1–29, http://dx.doi.org/10.1016/0304-3886(94)90026-4.
[3] K. Nakayama, Tribocharging and friction in insulators in ambient air, Wear 194 (1996) 185–189, http://dx.doi.org/10.1016/0043-1648(95)06840-6.
[4] L.G. Britton, Static hazards using flexible intermediate bulk containers for powder handling, Process Saf. Prog. 12 (1993) 240–250, http://dx.doi.org/10.1002/prs.680120410.
[5] J.E. Vinson, J.J. Liou, Electrostatic discharge in semiconductor devices: an overview, Proc. IEEE 86 (1998) 399–418, http://dx.doi.org/10.1109/5.659493.
[6] Istvan Berta, Static electricity hazards: solid surfaces and gases, in: Jen-Shih Chang, Mercel Dekker (Eds.), Handbook of Electrostatic Process, 1995.
[7] A. Iuga, A. Samuila, R. Morar, M. Bilici, L. Dascalescu, Tribocharging techniques for the electrostatic separation of granular plastics from waste electric and electronic equipment, Part. Sci. Technol. 34 (2016) 45–54, http://dx.doi.org/10.1080/02726351.2015.1043675.
[8] G. Buda, A. Samuila, S. Atroune, M. Bilici, L. Dascalescu, Set point identification of a tribocharging process for mixed granular solids, J. Electrost. 71 (2013) 407–412, http://dx.doi.org/10.1016/j.elstat.2012.11.026.
[9] M. Miloudi, K. Medles, A. Tilmatine, M. Brahami, L. Dascalescu, Modeling and optimization of a propeller-type tribocharger for granular materials, J. Electrost. 69 (2011) 631–637, http://dx.doi.org/10.1016/j.elstat.2011.08.010.
[10] B.K. Yun, J.W. Kim, H.S. Kim, K.W. Jung, Y. Yi, M.S. Jeong, J.H. Ko, J.H. Jung, Base-treated polydimethylsiloxane surfaces as enhanced triboelectric nanogenerators, Nano Energy 15 (2015) 523–529, http://dx.doi.org/10.1016/j.nanoen.2015.05.018.
[11] P. Bai, G. Zhu, Y.S. Zhou, S. Wang, J. Ma, G. Zhang, Z.L. Wang, Dipole-moment-induced effect on contact electrification for triboelectric nanogenerators, Nano Res. 7 (2014) 990–997, http://dx.doi.org/10.1007/s12274-014-0461-8.
[12] Su Yuanjie, Zhu Guang, Yang Weiqing, et al., Triboelectric sensor for self-powered tracking of object motion inside tubing, ACS nano 8 (4) (2014) 3843–3850.
[13] Yi Fang, Lin Long, Niu Simiao, et al., Self-Powered trajectory, velocity, and acceleration tracking of a moving object/body using a triboelectric sensor, Adv. Funct. Mater. 24 (47) (2014) 7488–7494.
[14] W.D. Greason, I.M. Oltean, Z. Kucerovsky, A.C. Ieta, Triboelectric charging between polytetrafluoroethylene and metals, IEEE Trans. Ind. Appl. 40 (2004) 442–450, http://dx.doi.org/10.1109/TIA.2004.824439.
[15] A.F. Diaz, R.M. Felix-Navarro, A semi-quantitative tribo-electric series for polymeric materials: the influence of chemical structure and properties, J. Electrost. 62 (2004) 277–290, http://dx.doi.org/10.1016/j.elstat.2004.05.005.
[16] L.S. McCarty, G.M. Whitesides, Electrostatic Charging Due to Separation of Ions at Interfaces: Contact Electrification of Ionic Electrets, vol. 47, Angew. Chemie - Int. Ed., 2008, pp. 2188–2207, http://dx.doi.org/10.1002/anie.200701812.
[17] H.T. Baytekin, A.Z. Patashinski, M. Branicki, B. Baytekin, S. Soh, B.A. Grzybowski, The mosaic of surface charge in contact electrification, Science 333 (6040) (2011) 308–312.
[18] M. Remadnia, M. Kachi, S. Messal, A. Oprean, X. Rouau, L. Dascalescu, Electrostatic separation of peeling and gluten from finely ground wheat grains, Part. Sci. Technol. 32 (2014) 608–615, http://dx.doi.org/10.1080/02726351.2014.943379.
[19] W. Aksa, K. Medles, M. Rezoug, R. Ouiddir, A. Bendaoud, L. Dascalescu, Modeling and optimization of a separator for granular mixtures composed of multiple insulating materials, IEEE Trans. Ind. Appl. 51 (2015) 4743–4751, http://dx.doi.org/10.1109/TIA.2015.2445815.
[20] L. Dascalescu, O. Fati, M. Bilici, F. Rahou, C. Dragan, a Samuila, a Iuga, Factors that influence the efficiency of a fluidized-bed-type tribo-electrostatic separator for mixed granular plastics, J. Phys. Conf. Ser. 301 (2011) 012066, http://dx.doi.org/10.1088/1742-6596/301/1/012066.
[21] F. Rahou, A. Tilmatine, M. Bilici, L. Dascalescu, Numerical simulation of the continuous operation of a tribo-aero-electrostatic separator for mixed granular solids, J. Electrost. 71 (2013) 867–874, http://dx.doi.org/10.1016/j.elstat.2013.06.004.
[22] J. Li, L. Dascalescu, M. Bilici, Z. Xu, Numerical modeling of the trajectories of plastic granules in a tribo-aero-electrostatic separator, J. Electrost. 71 (2013) 281–286, http://dx.doi.org/10.1016/j.elstat.2012.10.012.
[23] W. Aksa, K. Medles, M. Rezoug, M.F. Boukhoulda, M. Bilici, L. Dascalescu, Two stage electrostatic separator for the recycling of plastics from waste electrical and electronic equipment, J. Electrost. 71 (2013) 681–688, http://dx.doi.org/10.1016/j.elstat.2013.03.009.
[24] J. Kodama, R. Foerch, N.S. McIntyre, G. S.P, Castle, Effect of plasma treatment on the triboelectric properties of polymer powders, J. Appl. Phys. 74 (1993) 4026–4033, http://dx.doi.org/10.1063/1.354447.
[25] S. Trigwell, D. Boucher, C.I. Calle, Electrostatic properties of PE and PTFE subjected to atmospheric pressure plasma treatment; correlation of experimental results with atomistic modeling, J. Electrost. 65 (2007) 401–407, http://dx.doi.org/10.1016/j.elstat.2006.10.004.
[26] K.G. Kostov, A.L.R. dos Santos, R.Y. Honda, P.A.P. Nascente, M.E. Kayama, M.A. Algatti, R.P. Mota, Treatment of PET and PU polymers by atmospheric pressure plasma generated in dielectric barrier discharge in air, Surf. Coat. Technol. 204 (2010) 3064–3068, http://dx.doi.org/10.1016/j.surfcoat.2010.02.008.
[27] M. Xi, Y.L. Li, S. yong Shang, D.H. Li, Y.X. Yin, X.Y. Dai, Surface modification of aramid fiber by air DBD plasma at atmospheric pressure with continuous on-line processing, Surf. Coat. Technol. 202 (2008) 6029–6033, http://dx.doi.org/10.1016/j.surfcoat.2008.06.181.
[28] K. Hiratsuka, K. Hosotani, Effects of friction type and humidity on tribo-electrification and triboluminescence among eight kinds of polymers, Tribol. Int. 55 (2012) 87–99, http://dx.doi.org/10.1016/j.triboint.2012.05.017.