COLLECTION OF NON-VOLATILE PARTICLES IN AN ELECTROSTATIC FILTER IN SUBLIMATION REFINING OF SUBSTANCES

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In sublimation refining of substances in an atmosphere of nitrogen or superheated steam, the non-volatile particles carried away by a hot gas mixture were collected in an electrostatic filter with an efficiency better than 99%. The mean migration velocity of the collected particles was calculated from experimental data using an expression due to Deutsch.

Sublimation is one of the most effective purifying operations. While formerly it was used primarily in laboratories, its large scale application appears promising today. One of major advantages of sublimation is the possibility of relatively perfect separation of organic volatile solid substances from inorganic non-volatile matter. Even in these cases, however, when very strict requirements are set on the purity of products, difficulties can occur due to the entrainment of non-volatile particles in the form of aerosols together with vapors of the sublimated substance. Filtration has been used to separate the dispersed solid particles from the hot gas mixtures. Glass wool or porous materials are used mostly in laboratories. These, however, have to be cleaned frequently since a layer of material on the filter increases the hydraulic resistance. It seemed therefore suitable to include an electrostatic filter into the sublimation apparatus, which could collect particles of the order $0·1\mu m$ with high efficiency.

The electrostatic filter is a device commonly used in industry to separate solid and liquid particles from gases with an efficiency as high as 99·8% (ref.[1]). Its principal part is the discharge electrode mounted in the axis of a cylinder functioning as a collecting electrode. A slab is used sometimes[2] as a collecting electrode. The discharge electrode is connected to a negative or positive pole of a high voltage DC source and the collecting electrode is earthed. If the voltage between the electrodes exceeds a certain critical value, a silent discharge, called corona (owing to the collision ionisation) appears around the discharge electrode. Further increase in the voltage between the electrodes changes corona into sparking.

Conditions for the existence of the corona around the electrode located in the axis of a cylinder can be determined by means of the relationship[1].

$$E = (\varepsilon_0 \varrho D_d/2) [1 + \sqrt{K_0/(\varrho D_d)}] \ln (D_t/D_d) . \qquad (1)$$

The values of the constants ε_0 and K_0 for air are 110 statvolt/cm and 0·18 cm respectively. When $D_t/D_d < 2·718$, the corona can no longer exist and only sparking occurs.

Ions of gas originated by the corona bind the particles of the dust and these charge particles move by the action of the electrostatic field toward the collecting electrode. The magnitude of the charge carried by a particle and its migration velocity in the electric field are directly proportional to the gradient of the electric potential. Published data[1] show that the migration velocity of all charged particles smaller than 1 μm is almost identical and governed by the Stokes law. The migration velocity of particles larger than 1 μm is directly proportional to particle diameter. A relationship due to Deutsch usually given[1,2] in the literature for the calculation of the dust collection efficiency in a tubular filter is

$$\eta = 1 - \exp (- 4\mu_e L/D_t w_e) . \qquad (2)$$

This relationship is apparently valid for a given size of particles only, since the migration velocity contained in it is directly proportional to the particle diameter. It can be used, however, for an approximate determination of the average migration velocity of particles toward the collecting electrode, provided that an empirically found overall efficiency of separation under given conditions is substituted into it.

As a rule, an increasing temperature generally impairs the efficiency of separation of dust as the critical voltage at the electrodes is lowered and the increased viscosity of gas decreases the velocity of particle motion. Sproull and Nakada[3] studied the effect of temperature and humidity on the specific resistance of the dust in an electrostatic filter. They established experimentally that the majority of dusts attain the maximum electric resistance in the range between 90°C and 115°C, and that humidity decreases its value particularly at this temperature. The increased content of moisture in the gas increases also the value of the potential initiating sparking, namely at the temperature above 260°C. The current of the corona is decreased by the presence of moisture[4]. The authors[5] conclude that high or low temperatures are more favourable for electrostatic separation than the medium temperature around 90°C. For most of the dusts the operation is more efficient and practicable at a higher content of moisture in the gas. The effect of increased voltage and lowered current is particularly advantageous at the temperature around 300°C, where the influence of moisture on the specific resistance of the dust is small.

We have used electrostatic separation of non-volatile particles from a hot streaming gas mixture in sublimation refining of organic substances, namely, terephthalic acid in a fluidized bed[6]. The requirements on purity of this acid are usually very stringent, particularly in its use for the production of polyester fibers. Nitrogen or superheated steam were used as carrier gases.

EXPERIMENTAL

Chemicals and Apparatus

Terephthalic acid containing 0·72% in one case and 0·1 in the other of potassium was subjected to sublimation purification. The size of the acid particles was 10 to 100 μm. Further we purified terephthalic, isophthalic and benzoic acids contaminated by compounds of other metals, the content of which are shown in Table II. Refined nitrogen[6] with an oxygen content less than 2

p.p.m., or water steam prepared from freshly distilled water, degased under reduced pressure, were used as carrier gases.

The *potassium content* was determined by titration using acetic acid solution of perchloric acid. Prior to the titration the sample was rid of terephthalic acid by sublimation in an aluminium block. The relative error of determination of potassium content around 10 p.p.m. was about 20%.

Apparatus

The electrostatic filter is shown schematically in Fig. 1. The collecting electrode is a glass tube provided with ground joints at both ends (1) and (2) to facilitate assembling of interior, discharge electrode. The discharge electrode is a wire (Kantal A) 0·8 mm in diameter mounted in a capillary passage of the quartz joint (1) by means of a silver bead fused of the end of the wire. At the inlet of the gas mixture (3), the wire is mounted in a quartz tube (4) functioning as an insulator. The other end of the tube is connected with a weight (5) outside the filter. The capillary passages in the joints (1) and (2) are sealed by glass wool. The inner surface of the tube is coated with a conductive layer. The electric contact of the collecting electrode (6) passing through the capillary is provided with a fused-on silver bead resting in a cavity in the interior wall of the tube. The glass tube is electrically heated from outside and insulated by a jacket of glass wool. At the exit of the gas mixture there is a ground joint (7). Even a very careful completion of the joint (3) between the sublimator and filter allows certain leakage of air into the apparatus, which in turn impairs the quality of the product as the sublimation takes place at high temperature. Therefore, we increased the length of the sublimator so that the upper part forms the collecting electrode of the filter. A scheme of this set-up is shown in Fig. 2. About 200 mm long discharge electrode is
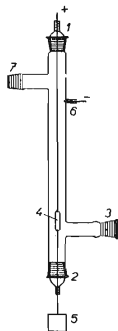

FIG. 1

Scheme of Electrostatic Filter
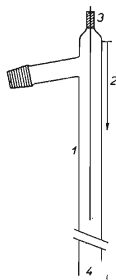 Explanation see in text.

FIG. 2

Scheme of Electrostatic Filter with Sublimator.
  1 Electrostatic filter; 2 heating; 3 Teflon passage; 4 sublimator.

mounted in the upper, narrowed section of the tube by means of a heat resistant teflon passage. The spherical cap of the collecting electrode including the narrowed throat for teflon passage is neither heated nor thermally insulated. The electric conductivity of glass at the temperature around 40—50°C is so small that the upper part of the electrode is an insulator itself.

To charge the filter, a DC source with the voltage ranging between 0 and 20 kV was used. The discharge electrode was connected to the positive pole and the negative, collecting was earthed. This arrangement was imposed by the construction of the source which did not enable reverse polarity.

RESULTS AND DISCUSSION

Since neither the composition, nor the size of the particles to be collected were known, we were not able to estimate their velocity in the electric field. Consequently, the dimension of the filter could not be determined either. On taking the gas velocity 0·5 m/s and the residence time of gas in the filter 1 s, we obtained, for a certain gas flow rate, the diameter of the separating tube 20—40 mm. Therefore, we used tubes 20 (Filter I) and 36 mm (Filter II) in diameter.

The voltage necessary for initiation of the corona discharge was calculated from Eq. (1) using constants for air and the relative density of the gas at 340°C. The respective voltages are 6·2 kV and 7·3 kV.

At first, the inner surface of the tube (collecting electrode) was silvered and the filter operated properly at the room temperature (it held up cigaret smoke). An increase in temperature to 200°C, however, resulted in sparking. Similar results were experienced with a semiconducting coat of the tube. It was found afterwards, that the electric conductivity of glass (Sial, Pyrex) at a temperature above 150°C allows sufficient potential to be formed at the electrodes under the conditions of the corona discharge. Consequently we used a clean glass tube, heated at the surface to the

TABLE I

Efficiency of Collection of Potassium from Terephthalic Acid

Inlet concentration of potassium[a] %	Potassium content in product p.p.m.	Gas velocity in sublimator m/s	Gas velocity in filter m/s	Overall efficiency of collection	Average migration velocity m/s
0·56	10	0·10	0·32	0·998	0·100
0·08	9	0·20	0·64	0·989	0·143
0·08	10	0·20	0·64	0·988	0·140
0·08	8	0·53	0·53	0·990	0·122

[a] Determined from the potassium content of product with disabled filter.

temperature of sublimation. Furthermore it was established that at the gas velocity 0·32 m/s the major portion of particles is collected at the collecting electrode within about 10 mm wide ring from the inlet, and that the upper limit of the range of collection is about 50 mm above the inlet. The effective length of the tube of the filters I and II was therefore changed to 100 mm and 180 mm respectively, to preserve the D_t/L ratio for comparison of the efficiency of both filters.

The maximum voltage available on the filter I at the temperature 340°C with nitrogen as a carrier gas and at gas velocity. 0·32 m/s (96% nitrogen, 4% terephthalic acid) was 8·4 kV. Using steam instead of nitrogen, the maximum voltage was 9·7 kV. The maximum voltages on the filter II were 11 kV and 13 kV when working with nitrogen and steam respectively. The voltage did not change when the gas velocity was doubled, neither did the sparking occur. The maximum voltage available on the electrodes of the filters is limited apparently by the electric conductivity of the end of the collecting electrode, heated to a substantially lower temperature than the rest of the electrode. In the atmosphere of superheated steam the maximum voltage is in both cases higher by about 18% than with nitrogen, which is in agreement with Sproull's observations.

Sublimation purification of terephthalic acid removes the traces of residual potassium present in the raw acid most likely in the form of potassium salts of terephthalic acid. Since potassium terephthalate is non-volatile, it should remain partly in the sublimator and its light particles be carried by the gas into the filter. Using the relationship[5] (3):

$$Re = Ar/[18 + 0.61 (Ar)^{0.5}], \tag{3}$$

we can calculated approximately the minimum velocity of entrainment of spherical particles under the conditions of sublimation. Let us assume that the density of en-

TABLE II

Collection of Compounds of Other Metals Concentration given in p.p.m.

Element		Fe	Zn	K	Al	Cu	Ni
Terephthalic acid	raw acid	1 400	33	1 200	85	34	160
	product	1·8	2	5	1·6	1·0	2
Isophthalic acid	raw acid	150	—	180	50	—	—
	product	2	—	5	2	—	—
Benzoic acid	raw acid	500	300	—	—	—	—
	product	2	2	—	—	—	—

trained material is 2 g/cm³. The calculated velocity of entrainment of spherical particles 100 μm in diameter in superheated steam will be 0·45 m/s, in nitrogen 0·35 m/s. For particles 50 μm in diameter in superheated steam and nitrogen 0·12 m/s and 0·076 m/s respectively. Particles 50 μm in diameter can therefore be entrained at the velocity of the gas mixture 0·1 m/s, while at the velocity 0·2 m/s particles larger than 60 μm.

Since we had no direct means of determination of the fraction of entrained particles, we performed first purification with disconnected filter and then, under the same conditions, with the filter. The difference in potassium content in the final product of both experiments gives the amount collected in the filter. The ratio of this value and of the potassium content of the first experiment (in Table I shown as inlet concentration of potassium) gives the overall efficiency of collection.

In experiments with the filter I, the velocity of gas mixture in the sublimator was 0·1 m/s or 0·2 m/s and the corresponding velocity in the filter 0·32 m/s or 0·64 m/s. The voltage was kept at 7 kV in nitrogen runs and 8 kV when steam was used. Starting with 0·1% of potassium in the raw acid, at the gas velocity in the filter 0·32 m/s, the potassium content of the product was less than 5 p.p.m. Here the error of determination is too large making thus the evaluation practically impossible. At the content 5 p.p.m. in the product, the overall efficiency of collection would amount to 0·994 and the average migration velocity to 0·081 m/s. At the velocity 0·64 m/s, the content of potassium in the product was in the range 8—10 p.p.m. 20% error in determination corresponds to about 5% error in the calculation of migration velocity. In experiments with the filter II, the velocity of the gas mixture in the sublimator and filter was 0·53 m/s, and the voltage 8 kV and 10 kV for nitrogen and steam respectively.

The average migration velocity toward the collecting electrode was calculated by means of Eq. (2). The results of experiments are summarized in Table I. The second column of the Table shows the averages of a larger number of repeated experiments. The efficiency of collection was practically independent of the kind of the carrier gas used. Analysis of collected particles proved that they consist of potassium terephthalate.

In a similar manner we proceeded in purification of acids contaminated by compounds of other metals. The sublimation of terephthalic acid took place at 340°C, isophthalic acid at 300°C and benzoic acid at 170°C. The results are summarized in Table II.

Analysis of the experimental data show high efficiency of the electrostatic filter for collection of non-volatile particles entrained by the carrier gas in sublimation refinement of substances in a fluidized bed. The values of the overall collection efficiency and from it calculated migration velocity of particles toward the collecting electrode at the corona voltage can be utilized for filter scale up.

LIST OF SYMBOLS

D_d outer diameter of discharge electrode   (cm)
D_t inner diameter of cylindrical collecting electrode   (cm)
E electrostatic potential difference giving rise to corona discharge (statvolt)
K_0 "energy-distance" constant for electrical discharge in gas   (cm)
L length of collecting electrode   (cm)
u_e migration velocity toward collecting electrode   (cm/s)
w_e average gas velocity in separator   (cm/s)
ε_0 electrical breakdown constant for given gas   (statvolt)
ϱ relative gas density related to its density at 0°C and 760 Torr
η collection efficiency
Ar   Archimedes group
Re   Reynolds group

REFERENCES

1. Perry J. H.: *Chemical Engineers' Handbook*. McGraw-Hill, New York 1963.
2. Štorch O.: *Průmyslová odlučovací zařízení*. Published by Státní nakladatelství technické literatury, Prague 1957.
3. Sproull W. T., Nakada Y.: Ind. Eng. Chem. *43*, 1350 (1951).
4. Zebrowski S. P.: Physik. Z. *33*, 727 (1932).
5. Todes O. M., Gorosko V. D. in the book: *Processy v Kipjascem Sloje*. (N. I. Syromjatnikov, V. F. Volkov, Eds). Metallurgizdat, Sverdlovsk 1959.
6. Vítovec J., Hančil V.: Chem. průmysl, in press.

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