Separation Science and Technology

Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

The Problem of Tube Breakage in Zone Refining, and a Solution

C. J. Doherty[a]; W. G. Pfann[a]
[a] BELL LABORATORIES MURRAY HILL, NEW JERSEY

To cite this Article Doherty, C. J. and Pfann, W. G.(1973) 'The Problem of Tube Breakage in Zone Refining, and a Solution', Separation Science and Technology, 8: 5, 593 — 597
To link to this Article: DOI: 10.1080/00372367308057049
URL: http://dx.doi.org/10.1080/00372367308057049

SEPARATION SCIENCE, 8(5), pp. 593–597, 1973

The Problem of Tube Breakage in Zone Refining, and a Solution

C. J. DOHERTY and W. G. PFANN

BELL LABORATORIES
MURRAY HILL, NEW JERSEY 07974

Abstract

The causes of tube breakage due to expansion of the charge during zone refining of chemical compounds are discussed. A new type of rotation-convection zone refiner which eliminates such tube breakage is described. Its main feature is that the molten zones fill slightly less than half the cross section of the tube, thereby producing solids with axial holes.

INTRODUCTION

In the zone refining of many materials that expand on melting, organic compounds in particular, a number of practical problems have arisen which reduce the efficiency of the refiner or increase the difficulty of its operation. One particularly annoying problem has been the breakage of the glass container when the container was in the form of a tube having its cross section filled with material to be refined. Breakage, due to expansion, commonly occurs when a new molten zone is formed at the closed end of a filled tube. Another common cause of breakage is the increase in volume of a molten zone due to heater fluctuation as it travels along a tube filled with charge-material.

Historically, the zone refining of organics began with horizontal open boats. The above problems of tube (boat) breakage were minimal. There was, however, a problem of matter transport arising from the expansion on melting, which caused material to accumulate at the front end of the boat (backward migration). This matter transport problem was easily resolved by raising the front of the boat to an empirically determined angle which prevented backward migration (*1*).

However, it was difficult in an ordinary horizontal boat to obtain short zone lengths, a desideratum in zone refining. Natural convection widened the zones at the top, leading to a "trapezoidal" zone shape. For this and other reasons, many chemists turned to vertical tube zone refining.

But vertical tube zone melting has disadvantages as well (*1*). Natural convection makes it hard to get short zones. Downward moving zones eventually create a porous solid with voids that cause irregular zone movements. Upward moving zones are subject to the breakage problems mentioned in the opening paragraph. To avoid tube breakage when a zone formed at the bottom of a tube, movable plugs of various forms were devised. These, too, had disadvantages: increased complexity, contamination, and gradual migration of the charge out of the refiner as the end-plug was forced further and further downward.

Recently an advanced form of zone refiner was invented, called the rotation-convection refiner (*2*). It used a horizontal tube, filled with charge material, which rotated slowly about its own axis, thereby achieving shorter zone lengths and interzone spacings than were theretofore possible. However, that refiner, having a filled cross-section, was also subject to backward matter transport and tube breakage, and movable plugs were commonly used.

APPARATUS

This paper describes an improved zone refiner which utilizes the rotation-convection principle (and thereby retains its advantages), but which completely avoids backward matter transport and tube breakage.

The new refiner rotates about its axis, as the refiner in Ref. 2. However, the total amount of material in the tube is less by a critical amount than that required to fill the tube, and the tube is tilted at a critical angle from the horizontal such as to prevent a net backward transport of matter. The height of the molten zone at the freezing interface must be less than the tube radius, so that the solid frozen out is disk- or cylinder-shaped, and has an axial hole. This is illustrated in Fig. 1 for one molten zone (without the tilting feature shown).

Although the zone occupies less than half the cross section, it produces, because of the slow rotation of the tube, a cylindrical solid filling the entire cross-section, except for the axial hole. We call it the "doughnut refiner" for obvious reasons. The total amount of material in the tube is

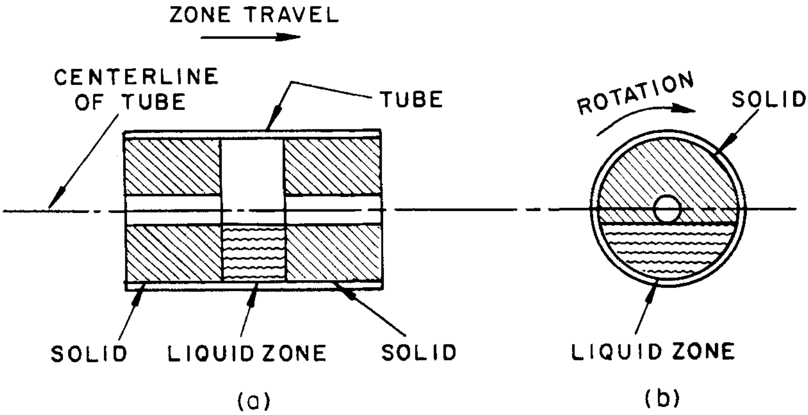

FIG. 1. Sketches of the "doughnut" rotation-convection refiner. (a) Longitudinal diametral section. (b) Cross section through molten zone.

determined by the number and length of molten zones present in a manner readily calculated, the essential requirement being that there be an axial hole in the solid.

If the refiner is operated horizontally, there will be backward matter transport. However, by raising the front end of the tube to a critical angle, a condition of zero net backward transport is achieved. The axial holes make this condition attainable. The critical angle is readily determined by experiment. Thus we have a zone refiner characterized by short zone lengths, short interzone spacings, enhanced natural convection, zero net matter transport, and no breakage of the tube either by formation of the first zone or by unintentional increase of zone length as the zone traverses the tube.

Heaters and coolers can be similar to those in the refiner of Ref. *2*, or they can be of simpler form, for the requirements are less critical. For example, they could be of a generally semicircular form beneath the tube.

EXPERIMENTAL RESULTS

Rotation-convection zone refiners, using the "doughnut" variation were built using glass tubes of 7/8 in. bore and 1-1/2 in. bore. Operation was completely successful over periods up to 100 reciprocations. The two main chemicals used were orthoterphenyl and naphthalene, each containing a small amount of oil red dye. To be absolutely certain that there could be no movement of the plug at the beginning of the charge, a glass disk was sealed into the glass tube. Rotation rates were 3–4 rev/min; the advance rate about 1 cm/hr.

The usual problems of formation of voids and their movement toward the end of the charge were completely absent. The 10-heater refiner was readily operated with axial holes about 1/4 in. in diameter, and with molten zone lengths from one-quarter to one-third of the tube diameter. We found it very difficult to obtain a good photograph of the refiner in action. However, in Fig. 2 is shown a 1-1/2-in. diameter charge of naphthalene for which the refiner was stopped after eight reciprocations and frozen in place. There can be seen the glass seal at the beginning, a length of clear white naphthalene, and an accumulation of dark dye at the end. Normally it would be preferable to end the operation either by cycling off the reciprocating heaters in sequence (*1*) or by simply drawing the entire charge out of the heater area.

As compared with the filled tube rotation-convection refiner, the doughnut refiner has the following pros and cons:

1. The effective zone length is far shorter than the actual zone length because the molten zone occupies less than half the cross section. This implies that the ultimate purification for a charge of given length should be greater, but it also implies that the rate of movement of impurity in the early stages of refining will be slow.

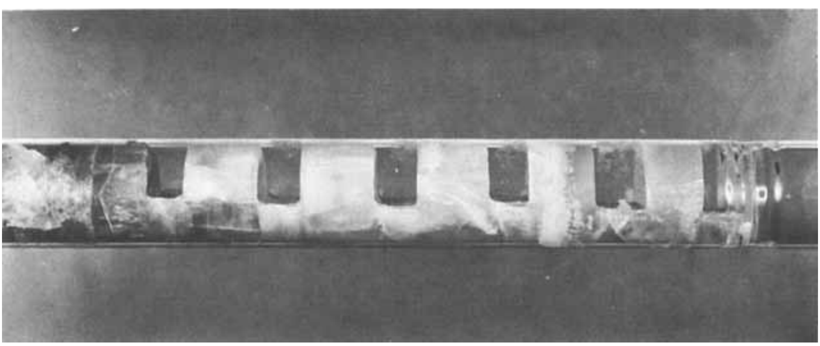

FIG. 2. One and one-half inch diameter solidified charge of naphthalene containing oil red dye after 8 reciprocations of "doughnut" rotation-convection refiner.

2. There is probably more convection in the molten zone in the doughnut refiner.

3. On the other hand, the impurity-rich layer of liquid adjacent to the freezing solid is withdrawn from the liquid and then later returned to it. This probably reduces the effective distribution coefficient.

4. Since there is a continuous open space along the entire refiner, the segregation of impurities with high vapor pressure would be retarded (*3*).

REFERENCES

1. W. G. Pfann, *Zone Melting*, 2nd ed., Wiley, New York, 1966, pp. 47 et seq.
2. W. G. Pfann, C. E. Miller, and J. D. Hunt, *Rev. Sci. Instr.*, *37*, 649–652 (1966); W. G. Pfann, U.S. Patent 3,423,189 (January 21, 1969).
3. O. Madelung, *Z. Phys.*, *162*, 508 (1961).

Received by editor April 19, 1973