AGGLOMERATE GRANULATION AS AN EQUILIBRIUM PROCESS

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1. INTRODUCTION

Agglomerate granulation is a process in which a mass of loose solid particles is transformed into a granular system of agglomerates by mixing it with a limited quantity of a liquid phase. The binding forces causing the particles to stick together are provided primarily by the capillary suction forces which are generated when the small voids between the particles are wholly or partly filled with liquid. The higher the degree of filling of the total void volume, the granules formed will consist of more primary particles and, in consequence, have a larger diameter. From many experiments described in the literature it can be derived that if the liquid phase content of such a granulate (y%wt) approaches the maximum degree of filling (y'), the (average) granule size will tend to infinity. Obviously, if the degree of filling equals zero the average granule size will be equal to the average size of the primary solid phase in its original form ($\bar{d}_p$).
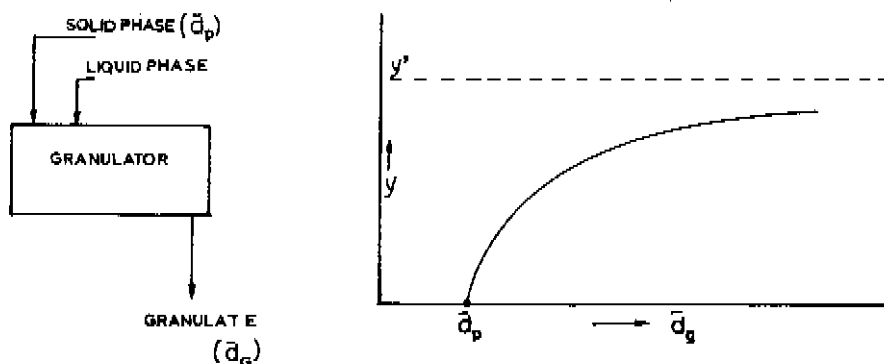A qualitative representation of the foregoing is shown in fig. 1.

SOLID PHASE ($\bar{d}_p$)

LIQUID PHASE

GRANULATOR

GRANULATE ($\bar{d}_G$)

Fig. 1

It is not our intention at this moment to deal in close detail with the correct mathematical relationship between $\bar{d}_g$ and its most principal determining factors $\bar{d}_p$, y and y', but exclusively proceed from the viewpoint that the average diameter of a granulate will increase with increasing liquid content of the granules.

In its simplest form this can be expressed as $\bar{d}_g = f(y)$.

In the above outline of the granulation process attention is directed primarily at the increase of the average diameter resulting from the increase in liquid content of the granulate. However, one should be well aware of the fact that an increase in liquid content will not only raise the average granule size but, in fact, shift the total sieve curve of the granulate to a higher level (fig. 2).
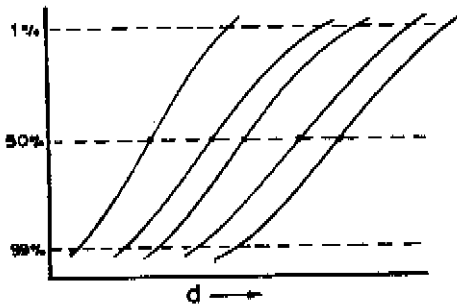

Fig. 2

As will be shown later, this is a factor of practical importance, because it provides a tool for controlling the proportion of "good product" fraction that can be obtained from the total amount of granulate. For the objective of granulation is not to produce a granulate of maximum average diameter, but a granulate containing a maximum proportion of "good product" (i.e. product coming within the specified diameter limits).

Briefly summarized, we may say, therefore, that:

- the relation $\bar{d}_g = f(y)$ denotes the degree to which the sieve curve of a granulate is shifted upon a change in liquid content,

- the total sieve curve indicates how strongly the "good product" fraction (between a and b mm) is influenced by change in $\bar{d}_g$.

It should be noted that whereas the factor $\bar{d}_g$ in the first relation is the resultant of the process in the granulator (granulate formation), in the subsequent stage this factor constitutes the cause of what is happening on the sieve (separation of the granulate into good product and off-size product, or recycle fraction).

We thus have:

$\bar{d}_g = f(y)$ (1)   where y = per cent of liquid phase in the granulate

$z = f(\bar{d}_g)$ (2)   where z = per cent of good product in the granulate.

2. GRANULATION UNIT

We shall now elaborate on the abovementioned relations with reference to a basic diagram (fig. 3). Only the granulator and sieve-crusher sections are shown; cooling and drying equipment has been omitted because this does not play an essential role in the establishment of equilibrium between:

liquid content ━━━▶ average diameter ━━━▶ good product fraction

      └ ─ ─ ─ ─relation 1─ ─┘   └─── relation 2───┘
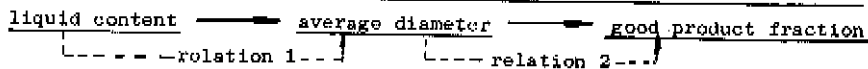
Under practical conditions the sieve curve may slightly change whilst the granulate is passing through the coolers and driers; although of minor significance, the influence of the responsible effects (after granulation, dust and rupture formation, etc.) can be assessed by determining the sieve curve in the cooled and dried product.

From figure 3 it can be concluded already that for the situation in a granulation unit to be in equilibrium, the total feed to the circuit (v) must be in balance with the total discharge (L). We shall have to examine therefore under what conditions this requirement will be satisfied. For this purpose the essential steps in the granulator and sieves will be considered separately.
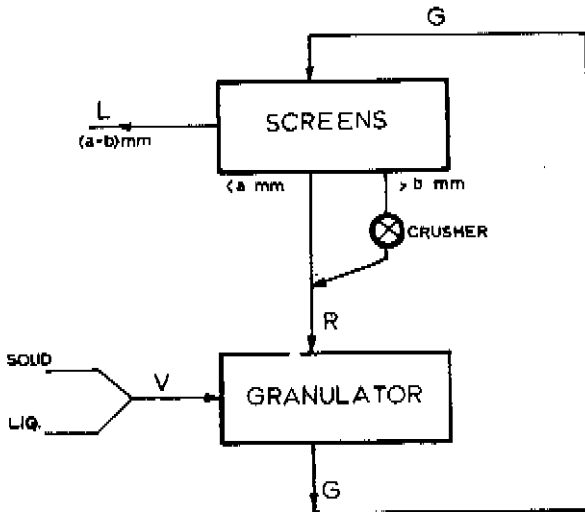
Fig. 3

V · weight of net feed (wholly or partly liquid)
G = weight of granulate (average diameter $\bar{d}_g$)
L = weight of end product (between a and b mm diameter)
R = weight of recycle product (dust plus crushed granules) - average
diameter $\bar{d}_p$

3. GRANULATOR ACTION

In the granulator, the net feed (wholly or partly liquid) is mixed
with the dry recycle stream. The liquid content of the resulting granulate
will increase as the proportion of the net feed in the granulate
increases. We point out, however, that the ultimate liquid content need
not be equal to the weighted average between the amounts of liquid in
the feed and recycle flows, because the liquid phase in the feed may,
after mixing with the colder recycle stream, give rise to crystallizing
or dissolving effects. The actual liquid content will be primarily
determined by the following three factors:
- H_2O content of the feed,
- solubility of the salt mixture,
- temperature in the granulator.

Although - and this applies particularly to fast-acting granulators - it is still an open question to what extent physical equilibrium will establish between the liquid and solid phases, we may postulate that the liquid content of the granulate is clearly related to:

. the feed-to-granulate ratio (V/G),

. the combination of the aforementioned three factors (further to be called the "feed conditions") in the actual situation.

In the light of what has been said above, the original relation $\bar{d}_g = f(y)$ ($\bar{d}_g$ is a function of the liquid content),

may be replaced by:

$\bar{d}_g = f(V/G)_c$ ($\bar{d}_g$ is a function of the proportion of feed in the granulate (V/G) and of the feed conditions (c)).

The graphical representation of this relation for various feed conditions is shown in fig. 4, to which we wish to add the following notes.

- A large number of experiments in the UKF-Stamicarbon pilot plants (output: appr. 100 kgs of granulate/h) and in commercial plants (output per unit: appr. 100-150 t/h) have shown that the relation $\bar{d}_g = f(V/G)$ is very well reproducible under constant operating conditions, and that the results of pilot plant runs are in very good agreement with those of experiments in commercial installations.

- It can be seen that the asymptotic trend of the original relation $\bar{d}_g = f(y)$ can still be clearly recognized in the shape of the relation $\bar{d}_g = f(V/G)$. The relative position of the three V/G lines (a, b and c) is clearly bound up with the fact that feed conditions providing a lower liquid content (a "drier feed" as represented by line c), demand a higher V/G ratio to enable the inter-particle voids to be completely filled with liquid than what is necessary under wet feed conditions

Fig. 4.
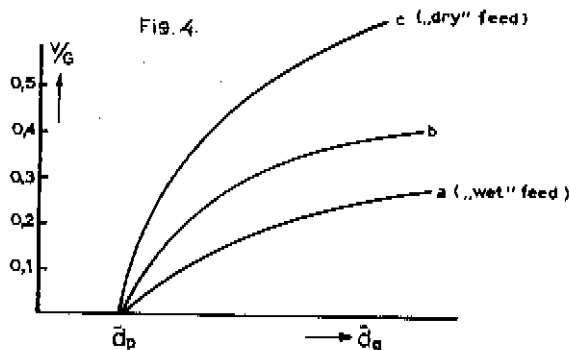
- It will be obvious that if V/G = 0, $\bar{d}_g$ may in all cases be considered equal to the average diameter of the recyle product $(\bar{d}_p)$.

In most plants $\bar{d}_p$ is approximately constant[*], because it is defined by the mesh size of the fine sieve and the setting of the crushers. It can, however, be varied to a limited extent by changing the crusher setting.

In summary, the relation between $\bar{d}_R$ and V/G can be basically influenced in two ways:

- by varying the "wetness" of the feed, which causes the V/G-line to hinge about its origin (fig. 5a),
- by varying the diameter of the recycle product, which causes the V-line to shift towards the left or the right (fig. 5b and 5c).
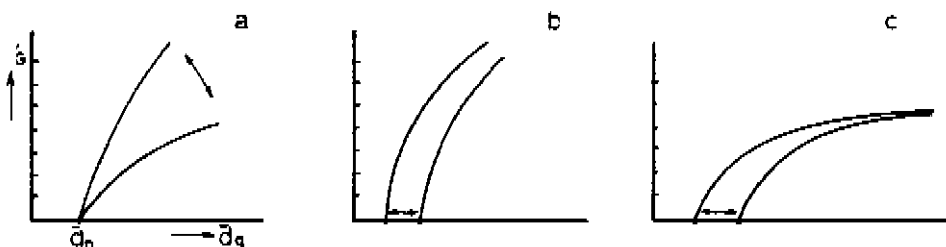

Fig. 5 (a,b,c)

4. SIEVE ACTION

The function of the sieves in a granulation line consists in recovering the "good product" fraction (granules within the specified diameter range a-b) as end product from the granulate.

If this fraction (L t/h) is withdrawn from a granulate flow of G t/h, the good product content of the gross granulate can be denoted as L/G.

We thus have two factors which relate to the composition of the granulate, namely:

[*] The recycle stream is made up of a portion of screen fines and a portion of crushed granules. Even if the average diameters in these two portions are constant (but unequal), the average diameter of the total recycle product $(\bar{d}_p)$ will depend on the fines-to-coarse ratio. Hence, a shift in this weight ratio will cause a shift in $\bar{d}_p$.

V/G = net feed content of the granulate, and

L/G = good product content of the granulate.

We have already seen that the equilibrium in the granulation unit is
reached and maintained only if and as long as V = L, or, referred to the
granulate, if and as long as V/G = L/G.

We also know that there exists a relation between V/G and $\bar{d}_g$
(V/G lines).

What we have to do now is to relate L/G to $\bar{d}_g$ in order to determine at
what value of d_g the factors L/G and V/G will be equal.

From mathematical statistics we know that:

- a plot relating the volume of a given diameter fraction to the average
diameter of the total mass passes through a maximum,

- if the sieve curve (approximately) conforms to the "normal
distribution"[*], the abovementioned fraction (L/G) will be maximum if
d_{50} ($\bar{d}_g$) equals the average of the upper and lower limits of that fraction.
So, if the limits are set at 2 and 4 mm, L/G will be maximum at
$\bar{d}_g$ = appr. 3 mm,

- the value of L/G at any given $\bar{d}_g$ is dependent on the standard
deviation (σ_g),

The relation between L/G and $\bar{d}_g$ for a given set of sieve meshes and two
different standard deviations, σ_1, and σ_2 in the granulate is qualitatively
illustrated in fig. 6.

The main factors governing σ_g will be discussed later.
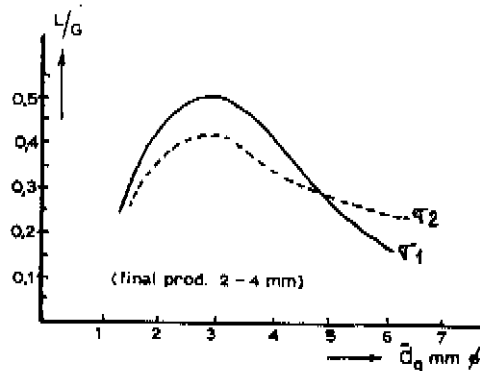

Fig. 6

[*]A distribution conforming to the Gauss curve. This conformity will exist
if a plot of the integral sieve curve versus the probability distribution
yields a straight line.

5. V/G AND L/G LINES COMBINED

We have already made it clear that

- the V/G line for a given feed and a given $\bar{d}_p$ indicates how $\bar{d}_g$ will increase with an increase of the feed ratio V/G,

- the L/G line shows how the "good product" content of the granulate (L/G) will increase with an increase of $\bar{d}_g$.

A plot of the two lines in one system of coordinates shows that the equilibrium V/G = L/G in the granulation unit will be established at the point where the lines intersect (fig. 7). The $\bar{d}_g$ value pertaining to that point is the equilibrium $\bar{d}_g$, while the corresponding L/G (= V/G) denotes the granulation efficiency under the conditions given.

This equilibrium establishes automatically, whatever the (absolute) quantity of feed supplied to the granulation unit, because the total quantity of granulate will adapt itself to the equilibrium point and the feed rate.
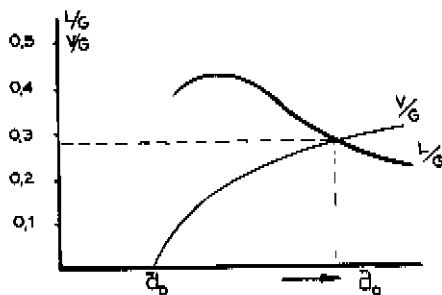

Fig. 7

Example:

If V = 35 t/h and V/G (= L/G) at equilibrium equals 0.28, G will establish at 35/0.28 = 125 t/h.

It is clear from this that under one given set of conditions (i.e. conditions governing the course of the V/G and L/G lines), there will be only one point[*] where the granulation system is in equilibrium.

[*]Under certain conditions, a 2nd and even 3rd equilibrium point may exist. This will be discussed later.

An increase, or decrease, in feed rate will raise, or lower, $\bar{d}_g$ for a short while only. After a few recycles this effect has completely levelled out, and the granulation system is operating again at the equilibrium $\bar{d}_g$ and the corresponding V/G (= L/G) value.

In the end the change in feed will be reflected exclusively in a change in loading of the unit. If it is desired to change the location of the intersection permanently, this can be done solely by shifting the V/G, or L/G, line into the direction desired (this will be discussed in more detail later).

Turning to the equilibrium diagram (fig. 7) we shall now deal with several aspects in somewhat more detail such as:

- the stability of the equilibrium point,
- the possibility of shifting the equilibrium point,
- suggestions for further granulation research.

6. STABILITY OF THE EQUILIBRIUM POINT

STABLE POINTS

    In the situation of fig. 7 the two lines intersect in one single point. This point of intersection may, depending on the course of the lines, be located either on the left, on, or on the right of the optimum (fig 8 a, b, c).
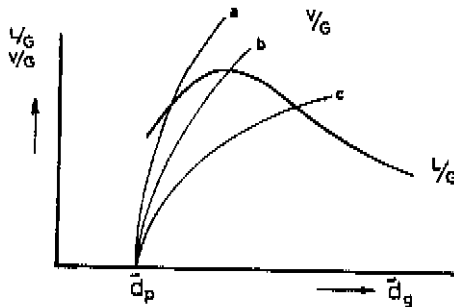

Fig. 8

In each of these cases the point of intersection is <u>stable</u>, in that sence that if any effect should cause the actual $\bar{d}_g$ to deviate from the equilibrium $\bar{d}_g$, the equilibrium will be automatically restored either to

the original point (if neither the V/G line nor the L/G line has been
changed) or to a new point (if one of the two lines has been changed).
An example of the first alternative is illustrated in fig. 9.

Here, V/G increases from A to B, whereupon $\bar{d}_g$ goes up from a to b; at
$\bar{d}_g$ = b we have L/G = C and, hence, V/G > L/G; the discharge from the unit
becomes smaller than the feed to it, with the result that the recycle
stream increases. As a consequence, V/G decreases again, so that $\bar{d}_K$ falls
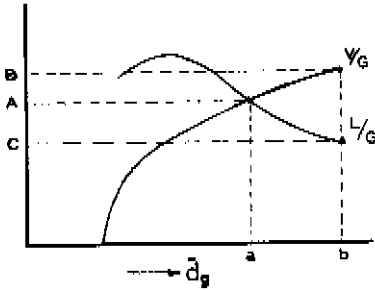off too, thereby sending L/G up again until V/G = L/G.

Fig. 9

In the situation of fig. 10 the wetness of the feed increases
(line (1) ⟶ line (2)). At constant V/G = A, $\bar{d}_K$ goes up from a to b,
and L/G decreases from A to B. We then also have V/G > L/G, the recycle
stream increases until V/G again comes to equal L/G.

It should be pointed out, however, that in this case the equilibrium has
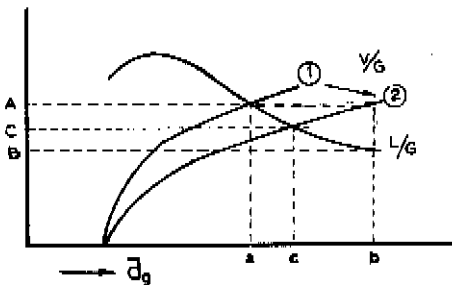permanently shifted from (a,A) to (c,C).

Fig. 10

Essentially similar shifts will occur when the intersection lies on the left of the optimum (fig. 11); in this case, however, shifting of the V/G line from 1 to 2 will cause the new equilibrium to establish at a higher instead of at a lower efficiency.

Sequence of the shifts:

1) V/G line from 1 to 2

2) $\bar{d}_K$ from a to b; V/G < L/G - recycle flow decreases,

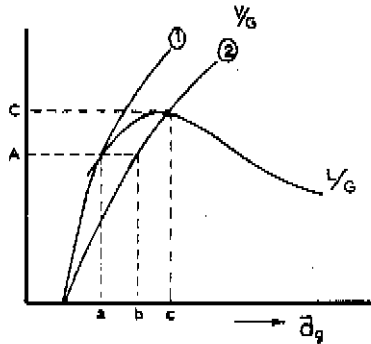3) $\bar{d}_K$ from b to c; V/G = L/G.

Fig. 11

In general we may say that an intersection on whose r.h. side the V/G line lies above the L/G line represents a stable equilibrium point.

UNSTABLE POINT

Depending on the course of the two lines (V/G and L/G) there may be more than one point of intersection, especially if $\bar{d}_p$ is rather small (very fine recycle product).

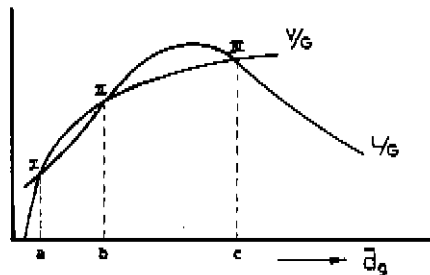We then have a situation as illustrated in fig. 12.

Fig. 12

I and III in this plot are stable points. Point II, however, is unstable.
When a deviation occurs, equilibrium will <u>not</u> return to this point, but
the deviation will increase in magnitude until the process regains a new
equilibrium, either in point I or III.

Point I is of little practical significance (very low efficiency;
negligible increase in granule diameter).

Point III, on the other hand, is a very real point; however, upon such
a change in V/G as will lower $\bar{d}_g$ to below b, the equilibrium will shift
to point I.

Clearly, a granulation unit operating in conformity with this diagram
is very difficult to control, especially if the causes of the above
phenomena are not understood.

INDIFFERENT EQUILIBRIUM

This type of equilibrium, which is characterized by a partial
overlapping of the V/G and L/G lines, is to be regarded as an
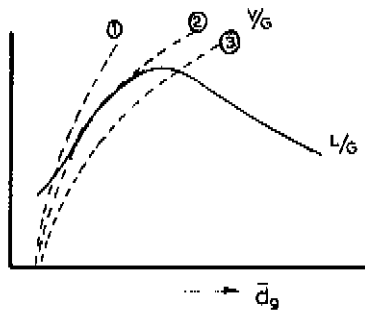intermediate between stable and unstable equilibria. (fig. 13 line 2)

Fig. 13

In the range where the V/G and L/G lines overlap, the process does not
react to changes in the V/G ratio. However, slight changes in the course
of the V/G line (fluctuations in the "wetness" of the feed) very soon
create a situation in which the stable equilibrium point may fluctuate
over a very wide range. Under these conditions the granulation behaviour
therefore fluctuates strongly and is difficult to control.

SEMI-UNSTABLE EQUILIBRIUM

A rather erratic granulation behaviour is also encountered in cases
where the stable equilibrium point establishes in the fairly flat section

of the V/G line.

Very minor deviations in the feed or recycle rate (resulting in a change
in V/G) already produce fairly strong shifts in $\bar{d}_g$ (see fig. 14). This is
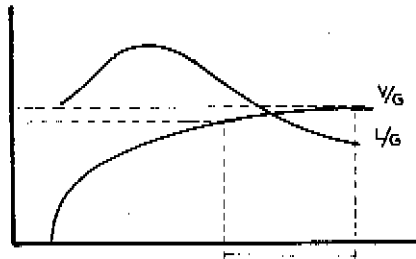especially apt to occur if the equilibrium point lies in the higher
diameter range.

Fig.14

7. POSSIBILITY OF SHIFTING THE EQUILIBRIUM POINT

Normally, it will be endeavoured to locate the intersection between
V/G and L/G in the top of the L/G curve, because the granulation
efficiency is highest there. Granulate diameters above or below this
range give rise to loss of efficiency. If in a given situation the
equilibrium should move too far away from the optimum, it can be
displaced towards the right or the left by changing the course of one of
the two lines.

SHIFTING OF THE V/G LINE

We have already seen that the V/G line can be shifted by varying the
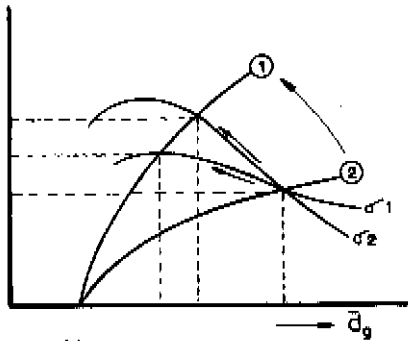"wetness" of the feed (fig. 15) or the value of $\bar{d}_p$.

Fig. 15

The degree to which this will influence $\bar{d}_g$ and the efficiency depends
not solely on the magnitude of the shift, but also on the course of the
(unchanged) L/G line.

SHIFTING OF THE L/G LINE

As stated in par. 4 the L/G line is defined by two factors: the lower and
upper limits specified for the end product (a and b mm) and the particle
size distribution (σ_g) in the granulate. The effects of these two
factors can be described as follows:

- the $\bar{d}_g$ value at which L/G passes through a maximum lies approximately
half way between the upper and lower limits where the separation is made
(a and b mm). We say "approximately" because many experiments have shown
that plotting of the integral sieve curve yields a straight line only if
the plot is made on a log. diameter scale. When such a sieve curve is
shifted towards the larger diameter range, the maximum L/G value will not
coincide with the arithmetic average of a and b, but with the logarithmic
average. However, since a and b lie close together, the difference is
rather insignificant.

Example:

a = 2, b = 4 mm; arithmetic average = 3.0 mm; logarithmic average = 2.8 mm.
As a rule, the limits of separation (a and b) are commercially prescribed,
which means that they allow only little opportunity of shifting the
optimum $\bar{d}_g$.

- Beside the fixed a and b values, there is only one other factor by
which the height of L/G is determined, namely the particle size

distribution (σ_g). A low σ_g (steep sieve curve) tends to increase the L/G maximum. We point out, however, that whereas the L/G maximum increases with falling σ_g, the L/G line falls off more sharply at $\bar{d}_g$ values on the right and the left of the optimum.

This explains why very steep sieve curves (often in combination with a high wetness of the feed) may go with granulation efficiencies that are equal or lower than those found in the case of flatter sieve curves. An example of this is shown in fig. 16.
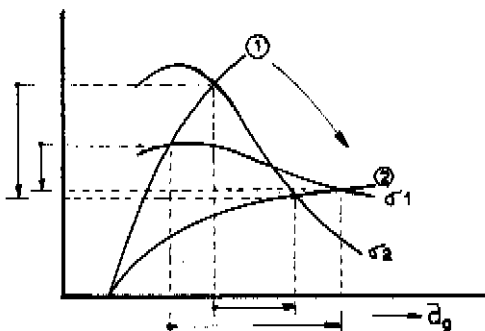

Fig. 16

It will be evident therefore that it is not possible to predict what will happen if a change is made in one of the two lines, or that such a prediction will be misleading if no information is available at the same time on the course of the other line.

We can now draw several conclusions from fig. 16, such as:

- reduction of σ_g (steeper sieve curve) in the case of a wetter feed (line 2) will have a negative effect on L/G in the equilibrium point, whereas this effect on V/G (line 1) will be just the reverse,

- reducing the wetness of the feed (V/G line from 2 to 1) if the granulate has a standard deviation σ_1 produces a minor effect on L/G, but a large effect on $\bar{d}_g$, whereas at σ_2 this will be just the other way about.

In general, it will be endeavoured, however, to raise the L/G maximum as high as possible (σ_g as small as possible). Therefore, it is important to find out the factors on which this σ_g depends.

Unfortunately, we are not in a position as yet to make any definite statements on this point. However, one aspect has come forward very

clearly in practically all experiments, namely the very high degree to
which the σ-value of the granulate is defined (or co-defined) by the
σ-value of the recycle flow. A steeper recycle sieve curve always gave
a steeper granulate sieve curve and, hence, a higher L/G maximum. Since
the sieve curve of the recycle flow is in many cases determined by the
performance of the coarse recycle crusher, the crusher type and crusher
capacity should be chosen with very great care.

The sequence: recycle sieve curve ⟶ granulate sieve curve with
increasing $\overline{d}_g$ ⟶ L/G line for two different sieve curves is shown in
appendix 1.

PRACTICAL CORRECTION OF L/G

We have seen that the L/G line shows the relation between the fraction
of good product sieved off from the granulate and the average granulate
diameter. However, in introducing this line, we tacitly proceeded from
two assumptions:

- the sieves separate 100 % of the good fraction from the granulate, and
- no good product nor unsieved granulate is recycled.

The L/G line dealt with so far is therefore to be regarded as a
theoretical L/G line, and should be corrected for these two effects in
practice.

It will be clear though that the good product fraction will never be
separated off completely, because the screens are not capable of making a
100 % sharp cut. The amount of good product obtained from the screens
is usually smaller than the amount conforming to L/G, the difference
being very highly dependent on the degree to which screens are loaded
(or overloaded), the fouling of the screens, the quality of the screen
decks, the length to width ratio of the mesh, the shape of the granules
etc.

This explains why the practical L/G line usually lies below the
theoretical line (fig. 17). The consequences this has for the equilibrium
of course depend again on the shape of the actual V/G line (see lines 1,
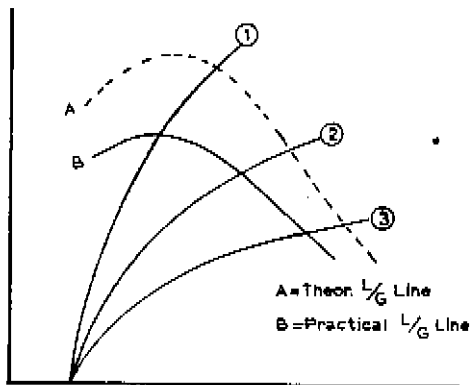2 and 3 in fig. 17).

Fig. 17

So much is clear, however, that improper sieve operation frequently has a
negative effect on the granulation efficiency.

Recycling of good product or unsieved granulate is frequently resorted to
in those cases where the equilibrium $\overline{d}_g$ lies at too high a level, since
this causes excessive fouling of the equipment. The effect on the L/G line
will be essentially the same as that produced by poor screen performance,
i.e. lowering of the L/G line.
If x % of the total granulate is recycled by bypassing the screens,
the practical L/G line will come to lie at an x % lower level than in the
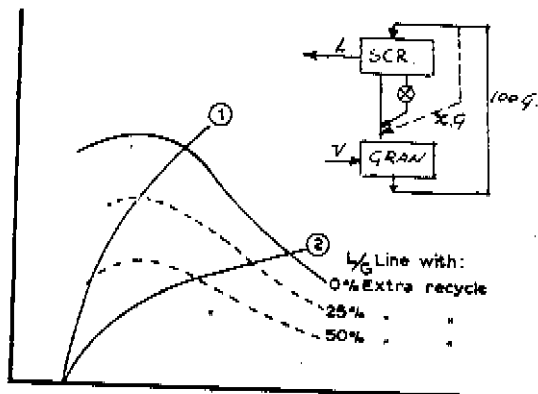event this additional quantity were left out of the recycle.

Fig. 18

So, this method of recycling can be resorted to if the equilibrium point has to be shifted to a lower $\bar{d}_g$. A remarkable feature encountered in this case is that the need of additional recycle increases with an increase of the normal recycle flow (from the screens). At equilibria corresponding to a high L/G ratio (small recycle flow), there is no need of enlarging the recycle flow (V/G line 1, fig. 18).

8. SUGGESTION FOR FURTHER GRANULATION RESEARCH

We have made it clear now that in a given situation the equilibrium point in a granulation system is defined by the combination of the V/G and L/G lines.

Experience in the UKF/Stamicarbon pilot plants (appr. 100 kgs granulate/h) has shown that under differing feed conditions these V/G lines can be very well determined and agree surprisingly well with the V/G line in large production plants (100 t of granulate/h).

Simultaneous determination of the L/G line, however, is equally essential as determination of the V/G line. For, the V/G line alone does not give any information about the equilibrium point that is to be expected, neither about its location nor about its character. Determination of this L/G line does not require any great effort because it can be constructed from the same basic information as is needed for constructing the V/G line.

Further, from the granulate sieve curve pertaining to a chosen V/G ratio, not only the value of $\bar{d}_g$ but also the amount of fraction intermediate between two separation limits (L/G theor.) can be read off.

However, an important requirement to be satisfied is that the sieve curve of the recycle product must fit in as closely as possible with that in the existing production plant or, if the plant is still in the design stage, with the sieve curve to be expected on the basis of the screens and crushers that have been chosen.

The assumption that the recycle flow will be identical to that produced in the pilot plant may be misleading, because the screens and crushers used there often differ altogether from those employed in practice.

Appendix 2 shows an example of a practical production run in which the V/G and L/G lines were determined for a N+P melt.

LIST OF SYMBOLS

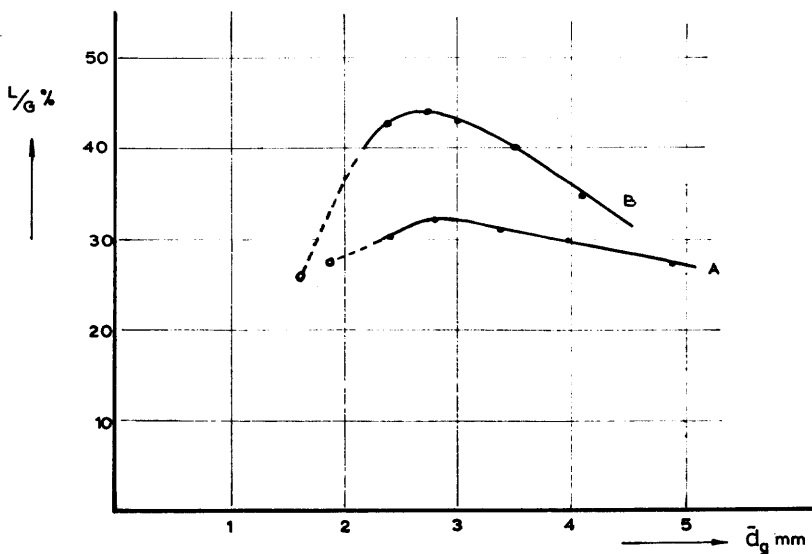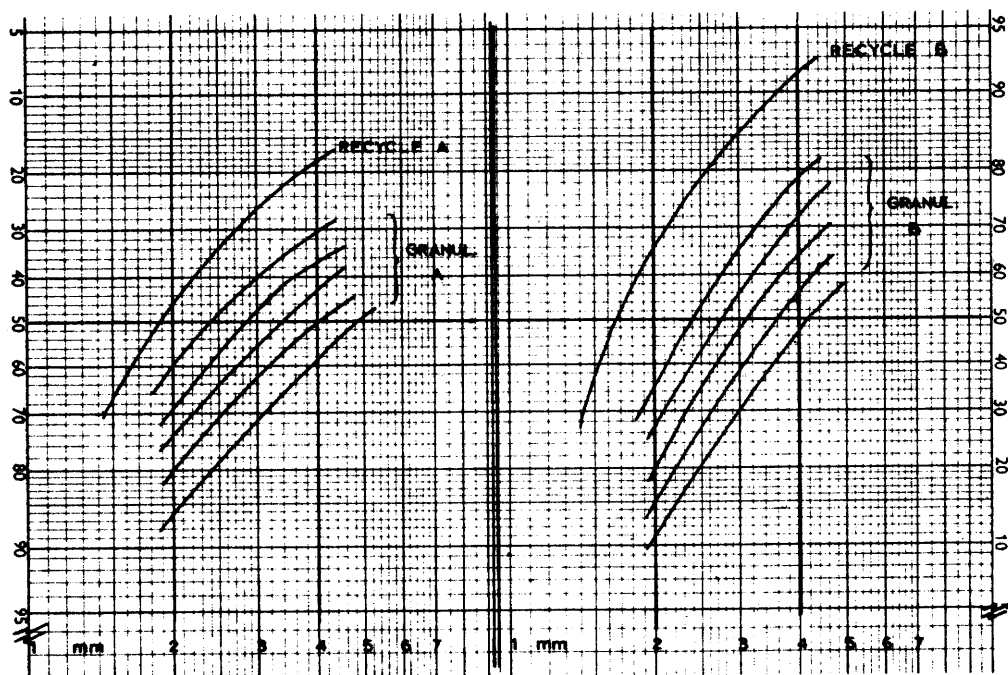V = net feed rate to granulator

R' = recycle flow rate

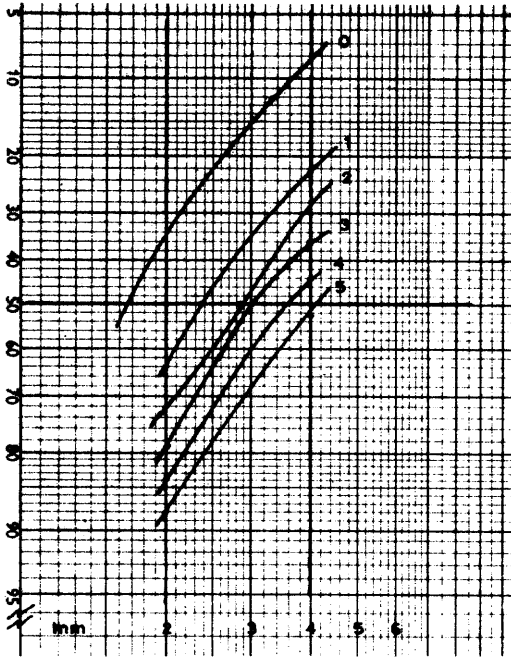G = granulato flow rate

L = end-product flow rate from screens

$\bar{d}_p$ = average diameter of solid phase, or recycle product to granulator

$\bar{d}_g$ = average granule diameter.

SIEVE ANALYSIS

SIEVE CURVES

Nº	$\bar{d}_g$	% > 2	% > 4	L/G theor.	L/G prac.	V/G
0	1.7	34	8	26	—	0
1	2.4	64	22	42	29	21
2	2.8	72	28	44	32	26
3	3.0	79	36	43	32	29
4	3.5	84	44	40	29	32
5	4.1	88	53	25	26	36