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ATOMIC ENERGY OF CANADA LIMITED

L'ÉNERGIE ATOMIQUE DU CANADA LIMITÉE

AGGLOMERATION TECHNIQUES FOR THE PRODUCTION OF SPHERES FOR PACKED BEDS

TECHNIQUES D'AGGLOMÉRATION POUR LA FABRICATION DE BILLES DE LITS PARTICULAIRES FIXES

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Chalk River, Ontario

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AGGLOMERATION TECHNIQUES FOR THE PRODUCTION OF SPHERES FOR PACKED BEDS*

by

J.D. Sullivan

*Work funded by Atomic Energy of Canada Limited Research Company and Canadian Fusion Fuels Technology Project

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RÉSUMÉ

Un type intéressant d'enveloppe de régénération de fusion est celui qui représente un céramique à lithium sous forme de billes tassées d'une manière désordonnée. Les billes ont un diamètre de 3 mm et 0,3 mm. Dans le présent rapport, on examine les techniques employées pour fabriquer des billes céramiques à l'échelle industrielle. Les techniques examinées comprennent la granulation par culbutage et mélange, l'extrusion et l'agglomération en briquettes et boulettes. On en conclu qu'on peut fabriquer la quantité nécessaire de billes de 0,3 mm avec la technique d'agglomération par culbutage d'une poudre d'alimentation. On fabriquera les billes de 3 mm de diamètre par un procédé d'extrusion, de tranchage et de laminage.

Matériaux pour combustibles
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ABSTRACT

One attractive fusion-breeder-blanket design features a lithium bearing ceramic in the form of spheres packed into a random array. The spheres have diameters of 3 mm and 0.3 mm. This report surveys techniques used to produce ceramic spheres on an industrial scale. The methods examined include tumbling and mixing granulation, extrusion, briquetting and pelletizing. It is concluded that the required quantities of 0.3 mm diameter spheres can be produced by the tumbling agglomeration of a feed powder. The 3 mm diameter spheres will be made using a process of extrusion, chopping and rolling.

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AGGLOMERATION TECHNIQUES FOR THE PRODUCTION OF SPHERES FOR PACKED BEDS

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1. Preface

One attractive fusion-reactor breeder-blanket design features a lithium bearing ceramic in the form of spheres packed into a random array [1]. The spheres have diameters of 3 mm and 0.3 mm. The starting material for these spheres will be a ceramic powder, and methods will be needed to make megagram quantities of spheres from these powders. The general industrial term to describe the process of bonding together fine powders to form larger shapes is agglomeration. There are a number of different methods used in agglomeration, including agitation, briquetting, extrusion and pelleting. The suitability of these methods to produce ceramic spheres will be considered below. This report gives results from the literature and from laboratory-scale experiments carried out using some of the more promising agglomeration methods.

2. Background

There is a considerable background in the technology of agglomeration in the iron and steel, coal, food and pharmaceutical industries. The feed powders for these processes are typically larger than 50 micrometres, and often larger than 100 micrometres. There is little current application of the agglomeration of sinterable ceramic powders in the 0.1- to ten-micrometre size range. The handling characteristics of these fine powders are quite different from the coarser traditional powder products.

The effect of feed powder size on product size can be seen in Figure 1, which shows agglomerated particle size as a function of the number of revolutions of a drum agglomerator (this type of process is described below). Data are shown for a feed powder less than 50 micrometres in diameter, and for a feed of between 75 and 125 micrometres. The size of the agglomerated granules decreases with a decrease in the size of the feed powders. It can be concluded that if a one micrometre feed powder were used, the agglomerated particle size would be proportionately smaller than those shown in Figure 1.

There is a theoretical limiting sphere size which is practically achievable by simple agglomeration, given the feed-powder size distribution. To understand this, consider a process in which the agglomerating spheres pick up material from the mass of feed powder in discrete amounts (in fact, the discrete quanta being picked up are the feed powder particles themselves). In such a model, the time rate of volume increase is a constant ($dV/dt=constant$). Thus the diameter of the agglomerated particles is proportional to the cube root of the tumbling time. Consider the case of the agglomeration of a powder that forms spheres of approximately 200 micrometre diameter in one day (a typical result for a powder of micrometre dimensions). Assume that enough powder is added to the process that the spheres grow in volume by the same amount each day. A plot of the diameter of the spheres as a function of time, for this process, is shown in Figure 2. The process quickly reaches a point of diminishing returns and the sphere size levels off. Even after a month the predicted size would be less than one millimetre. The processes of nucleation of new spheres and erosion of agglomerated spheres would cause the actual sphere diameter to be less than that calculated above. Feed particles are incorporated onto the surfaces of agglomerated granules and it could be debated whether the rate of granule volume increase is constant or is proportional to granule surface area. In either case, the sphere size levels off and there is a practical limit to the size of spheres which could be produced by this process.

3. Agitation

The process of agitation is one in which a feed powder is placed in a tumbling or turbulent environment. As the particles tumble and impact, they clump together to form agglomerates. Devices which operate on this principle include both tumbling and mixing agglomerators.

3.1 Tumbling Agitation

Tumbling agitators include devices such as pan and drum agglomerators. A schematic of a pan agglomerator is shown in Figure 3. The feed powders are placed in the drum or tilted pan and the enclosure is rotated at such a rate that the powders tumble inside the enclosure (shown schematically in Figure 3a). This brings the powders into intimate contact and allows them to build up into larger shapes. Binder can also be sprayed into the pan. Because of the random motion associated with this process, sphere-like objects are formed. The smaller spheres tend to sink to the bottom, so the process is self-classifying (Figure 3b).

This process has been employed using A16SG alumina powder[1], to produce spheres of high uniformity, approximately 200 micrometers in diameter, without the use of a binder [2]. The time required for this (batch) process is approximately 50 hours. 200 micrometres seems to be a practical upper limit in size for this process, using A16SG alumina, since the population of spheres remains constant after this time, and there is no tendency for any of them to increase in size beyond this limit.

To investigate the effect of binder addition on the agglomeration process, a rotary (drum) agglomeration experiment was carried out using A16SG alumina powder and water as a binder. A one week trial was carried out in which 60 g of A16SG containing 10 vol% water was placed in a rotating drum and tumbled for one day. From this agglomerated sample, 30 g were extracted, moistened with a fine mist and placed in a second drum, along with 30 g of dry fresh feed powder. This second drum was tumbled for one day, and the above process was repeated. A portion of the tumbled product

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[1] Aluminum Company of America, 1501 Alcoa Building, Chemicals Division, Pittsburgh PA 15219

from each day was retained. The intent was to use the spheres produced by one run to act as seed spheres for the next run. If no new spheres were nucleated by this process, the existing spheres should have grown by a constant volume each day and the average sphere size should have followed a curve as shown in Figure 2.

The products from days one, three and five were sized by sieving. A plot of the size distributions on these days is shown in Figure 4. There is no noticeable increase in sphere size, even after five days. A constant volume growth rate would predict that the diameters of the spheres on day five be 70% larger than on day one, and the observed growth rate is clearly less than that. In fact, the larger size fraction is seen to decrease slightly with time and the fraction between 125 and 250 micrometres is seen to increase slightly. This result indicates that a steady-state condition (erosion rate equal to agglomeration rate) is set up fairly rapidly (less than a day), and once this steady-state is reached, there is no tendency for particle size enlargement.

The growth rate of the agglomerating spheres should be dependent on the binding force between the particles, and thus should be affected by the quantity of binder added to the feed powder. A mixture of A16SG alumina containing 40 vol% water was made up and tumbled for 60 hours in a drum agglomerator. Spheres up to 1 cm diameter were formed, but the size distribution was quite large (diameters ranged over an order of magnitude).

With the exception of producing spheres of a size determined by the powder characteristics (ie. A16SG readily forms uniformly sized spheres of 200 micrometre diameter), it is difficult to accurately control the size of the product. Agglomerate size is dependent on powder characteristics and also is determined partially by binder content. For larger agglomerate sizes, the product has a rather wide size distribution.

3.2 Mixing

Other methods of agitating powders make use of intentional mixing through the use of paddles, pins or blades. A drawing of a mixer is shown in Figure 5 [3]. These devices have the ability to increase the rate of contact of the particles, and consequently to decrease the granulation time.

The product from such a process has a rather wide size distribution, which can range over an order of magnitude in diameter. An example of the size distribution of ceramic powder agglomerated in an Eirich mixer is shown in Figure 6 [4]. The decrease in processing time in this type of agglomerator, over a simple pan agglomerator, can be seen in this figure. A typical processing time in the Eirich mixer is approximately 10 minutes, whereas for the pan agglomerator it may be hours. The size distribution of the feed powder, which ranges from 40 to 230 micrometres, is also shown in Figure 6. The maximum product size is approximately 5 mm, and there does not appear to be an increase in product size with tumbling time. It would be expected that the product formed from a smaller feed powder (1 micrometre) would be proportionally smaller, and this process would not be easily employed for the production of 3 mm spheres.

4. Extrusion

Tumbling and agitation methods rely on random contact between particles to form agglomerates. Extrusion is a process which forms the particles into a large mass during the initial stages of the procedure. The powder is plasticised by the addition of lubricants, binders and solvents, and then the clay-like plastic mass is forced through a nozzle to form an extruded shape. The driving force can be supplied by either a piston or a screw. A piston extruder is shown in Figure 7a. The requirements on the plastic mass are that it be able to flow through the nozzle, but the green extrudate must be strong enough to retain its shape.

This process cannot form spheres directly, since the product is a continuous rod. The extruded rod, however, can be cut into short cylindrical sections which can be tumbled, in a rotating pan or drum, rounding off the corners of the cylinders and forming spheres. This process is shown in Figure 7b. The size of the product is controlled by the size of the opening in the nozzle. Equipment which operates on this basis is made by Luwa Corporation[2] (marketed under the trade names Xtruder and Marumerizer) and is available in both laboratory and plant scale versions.

Laboratory trials, which imitated the steps used in the Luwa equipment, were conducted to assess the effectiveness of such a process to form spheres of ceramic powders. A16SG alumina was plasticised by the addition of water and starch. This plastic mass was extruded through a 3 mm diameter hole in a large bore plastic syringe to form a rod, which was cut into 3 mm lengths and tumbled in a pan to form spheres. The spheres were less round than those formed by drum agglomeration of powders, but were of suitable quality for use in a bed of packed spheres. The spheres sintered to only 70% of theoretical density, indicating the need for process improvement.

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[2] Luwa Corporation, P.O. Box 16348, Charlotte, North Carolina 28216

5. Briquetting

Briquetting, or roll compaction, makes use of counter-rotating rollers to compact the feed powder into a shape. Powder is fed into the nip between the rollers and released below the rollers in compacted form. A schematic drawing of this process is shown in Figure 8.

It is possible to form near spherical shapes from such a process by putting semi-spherical holes in the rollers. Very large forces are required to press the powder into the final shape (laboratory scale presses are about 220 kN, or 25 tons, and industrial presses can be up to 2.7 MN, or 300 tons [5]). The process is more energy intensive than that of extrusion and the rate at which spheres are made is set by the roller speed and the ability to deliver a controlled quantity of feed powder to the nip.

6. Tableting

The final form of processing considered here is that of tableting. In this case, the product is formed in a die with one or two punches which compact each tablet individually. The process is shown in Figure 9. This technique is very common in the pharmaceutical, powder metallurgy and ceramics industries. High-speed rotary presses are capable of production rates of typically 1000 parts per minute [5].

To assess the suitability of this type of process for the production of megagram quantities of ceramic, it is necessary to determine the effort needed to produce the required number of spheres. Consider only the larger size fraction in the fusion breeder blanket sphere bed (3 mm diameter). The total volume of the breeder blanket will be approximately 300 m^3. Of this volume, approximately 63% is occupied by the 3 mm spheres, for a total ceramic volume of 200 m^3 (for this size fraction). The volume of one sphere is 0.014 cm^3, so the total number of spheres is 1.4×10^{10}. At the (high) production rate of 1000 spheres per minute, it would require 10 000 days (27 years) of 24 hour per day production to fully charge the breeder bed. The smaller size fraction occupies one third the volume of the larger, but the individual spheres have one thousandth the volume of the larger spheres. As a result, the number of smaller spheres will be hundreds of times the number of larger spheres and tableting would be even less feasible for this size fraction.

7. Assessment

The goal of this exercise is to identify practical processes capable of producing large quantities of lithium bearing ceramic spheres in the size range 0.3 to 3 mm diameter.

Of the processes considered above, the most suitable for the formation of 3 mm diameter spheres would be extrusion. Although not precisely round, the spheres are of acceptable quality and have a reasonably narrow size distribution. Drum or pan agglomeration requires careful control over binder content and feed particle properties, and yields a product with a larger size distribution than that for the extrusion approach. Processes which form the spheres individually (briquetting or tableting) would require very high production rates to turn out the spheres in a reasonable time and are not feasible for this application.

The most suitable process for producing the smaller spheres would be pan or drum agglomeration of powders. Although it is rather difficult to control the size of the final product (increasing the size significantly above that formed in one day is not practical), this method is able to make large numbers of sub-millimetre spheres with good size uniformity in a reasonably short time period. Processing trials using tumbling agglomeration must make use of the actual powder to be agglomerated, since the product size is determined largely by the properties of the powder.

References

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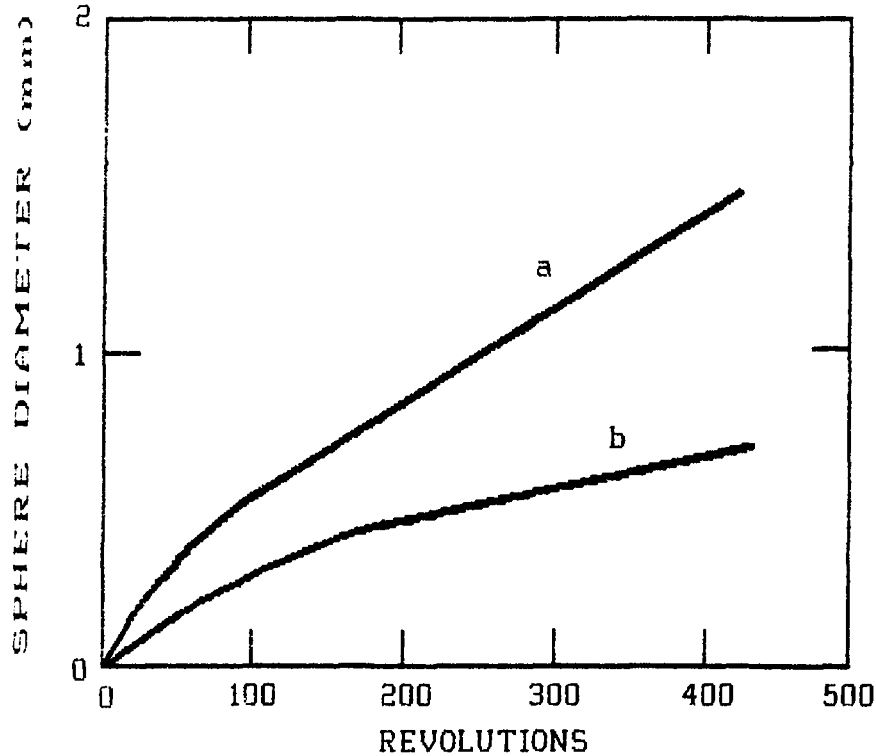

Figure 1. Effect of feed particle size on the growth rate of iron ore pellets in a laboratory balling drum. The feed powder for curve "a" ranged from 75 to 125 micrometres, the feed for "b" was less than 50 micrometres [3].

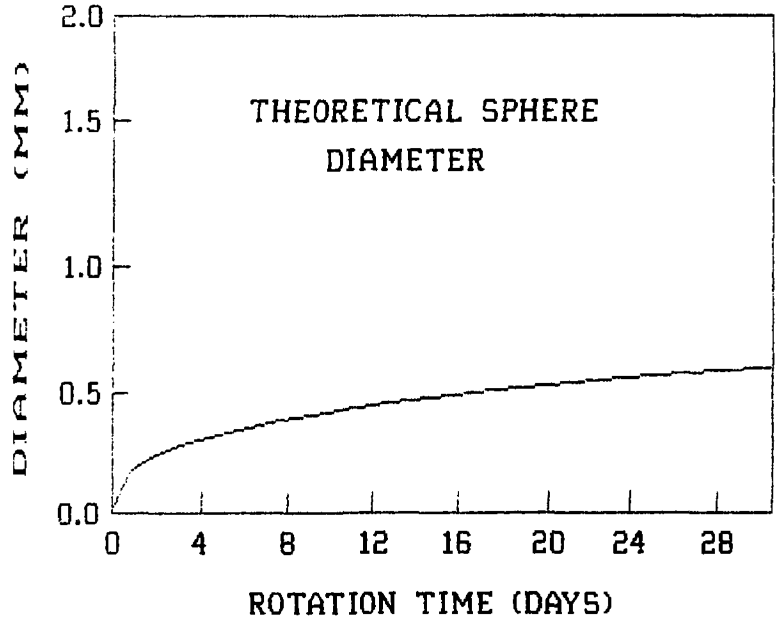

Figure 2. Theoretical sphere diameter as a function of rotation time assuming constant volume growth rate.

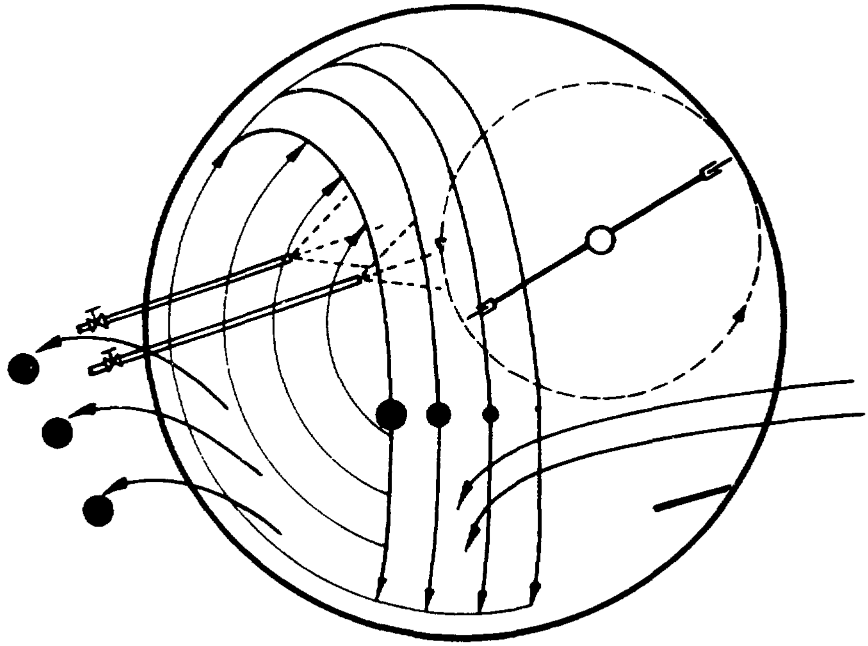

Figure 3a. Pan agglomerator showing powder feed (arrows) and binder addition.

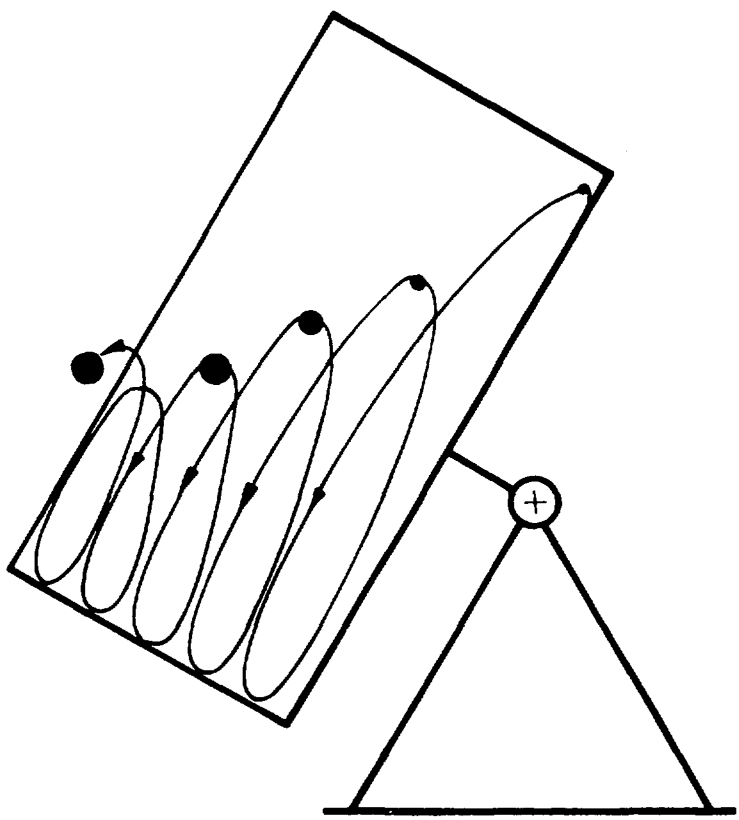

Figure 3b. Self-classifying feature of pan agglomerator.

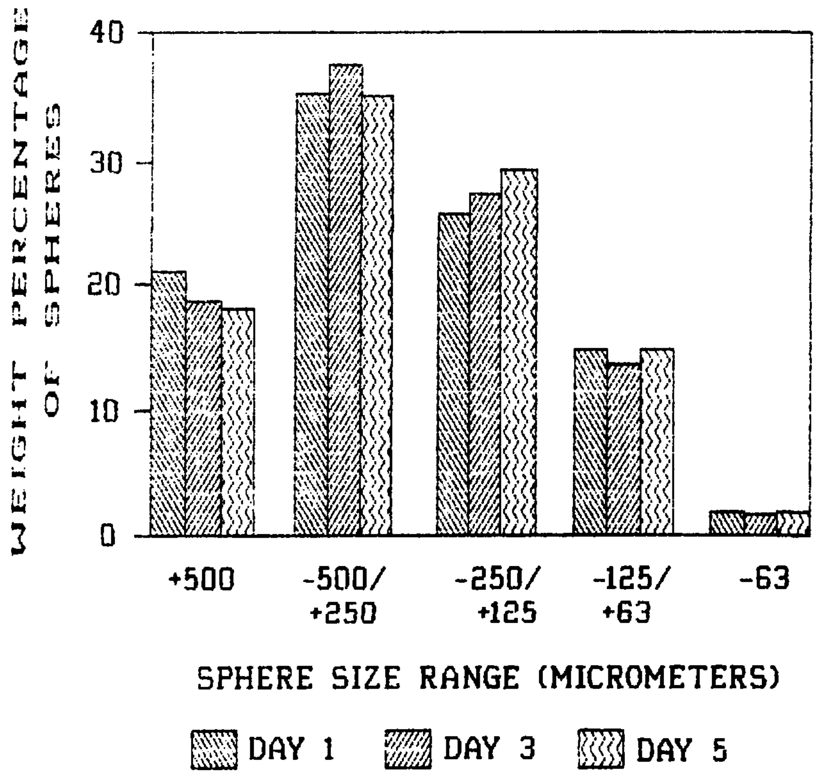

Figure 4. Sphere size distribution as a function of agglomeration time for A16SG alumina.

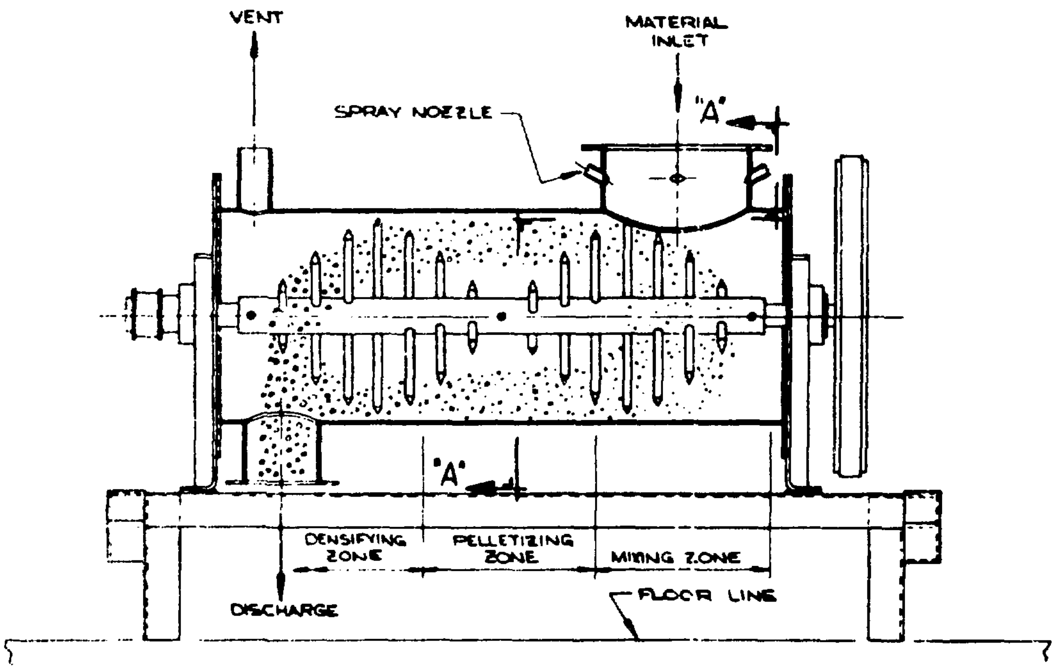

Figure 5. Diagram of a mixer showing powder feed, agglomeration zones and product discharge.

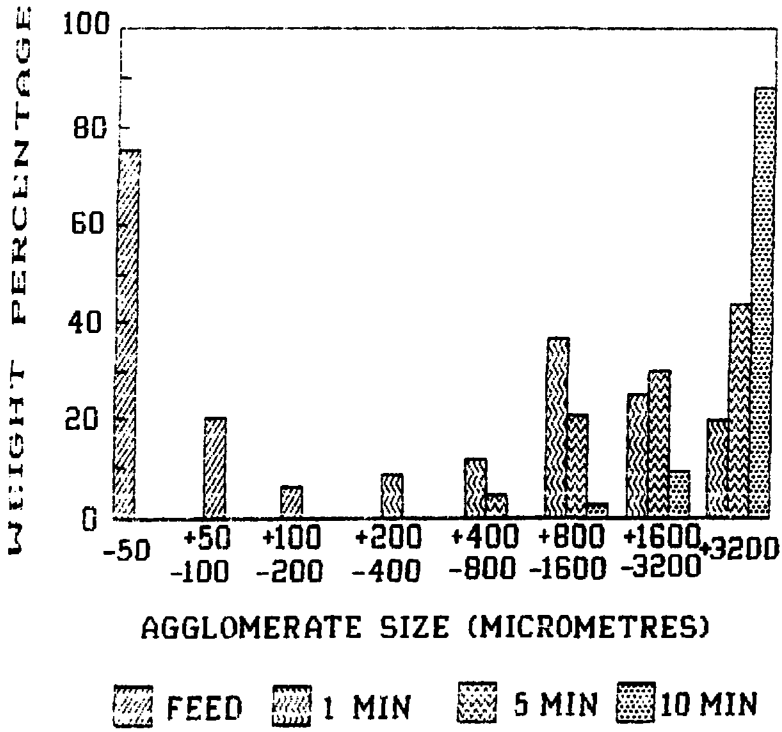

Figure 6. Particle size distribution in a ceramic powder, agglomerated in an Eirich mixer, as a function of mixing time. The moisture content is 14% [4].

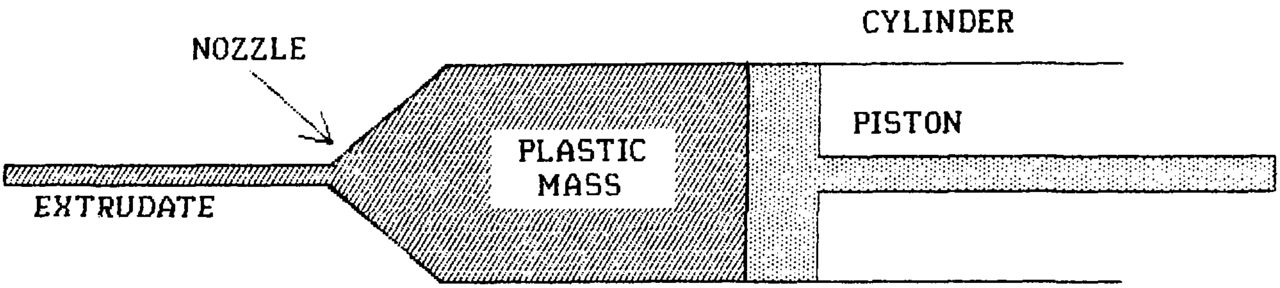

Figure 7a. Extrusion process using a piston extruder.

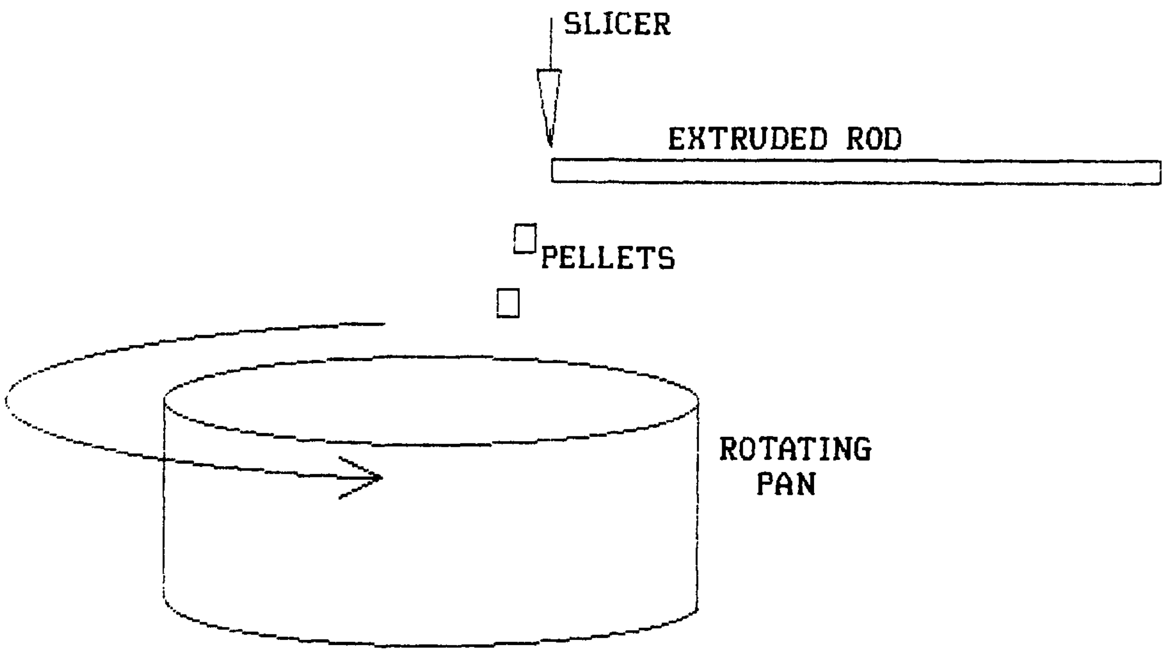

Figure 7b. Forming spheres from extruded rod in a rotating pan.

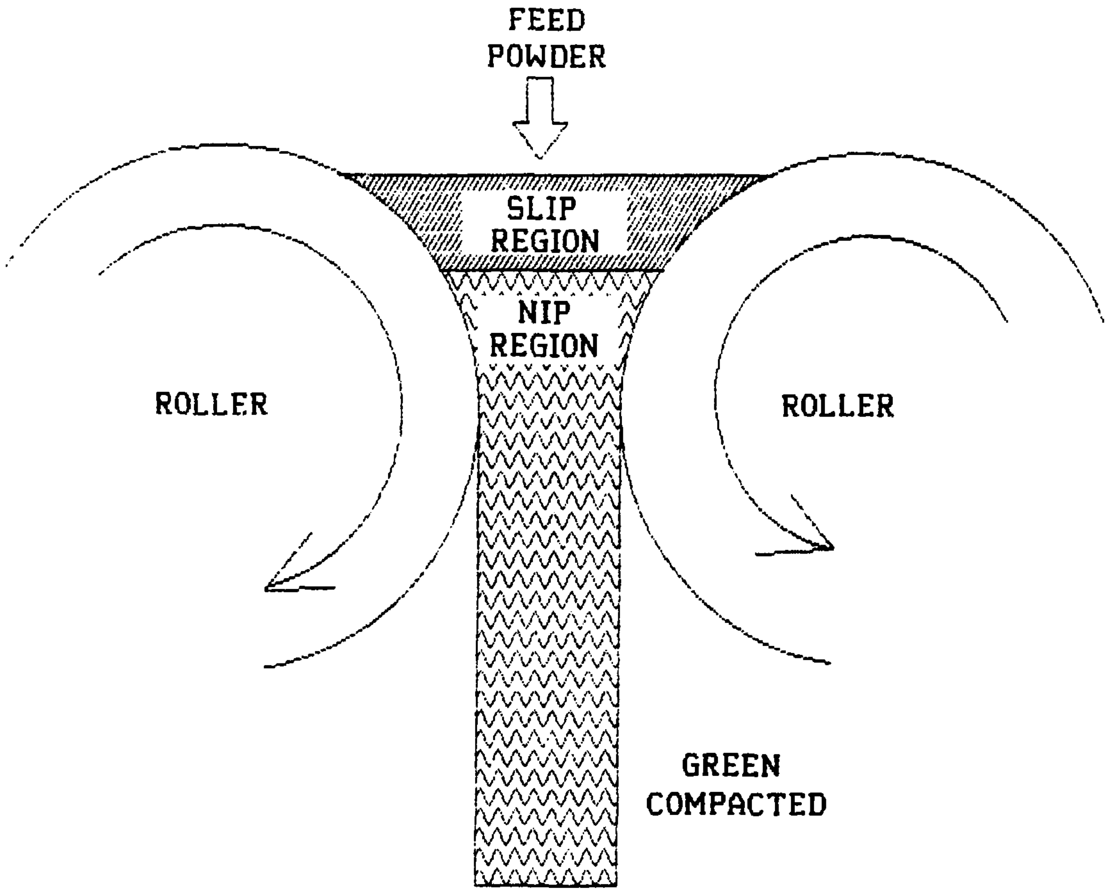

Figure 8. Briquetting of powder by roll compaction.

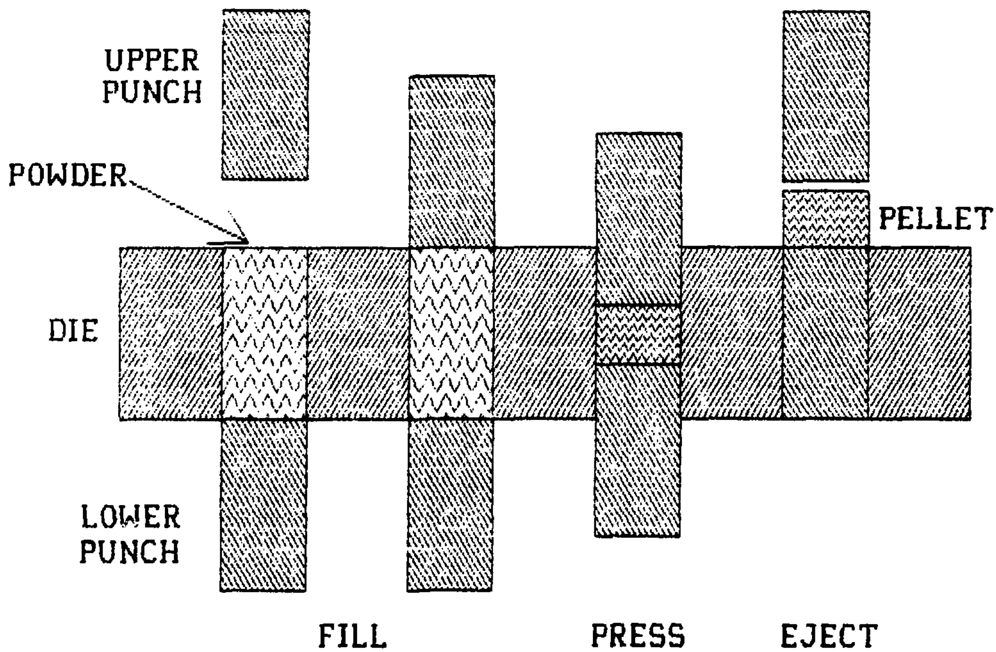

Figure 9. Tableting by double ended pressing of powder. Fill, press and ejection stages are shown.

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