(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: 0 045 564 A2

(12) # EUROPEAN PATENT APPLICATION

(21) Application number: 81302995.6

(22) Date of filing: 01.07.81

(51) Int. Cl.[3]: B 28 B 3/20
B 28 B 17/00, B 28 B 11/16

(30) Priority: 29.07.80 GB 8024695

(43) Date of publication of application: 10.02.82 Bulletin 82/6

(84) Designated Contracting States: BE DE FR GB IT NL SE

(71) Applicant: IMPERIAL CHEMICAL INDUSTRIES PLC
Imperial Chemical House Millbank
London SW1P 3JF(GB)

(72) Inventor: Benbow, John Jones
22 Mill Hill Lane
Northallerton N Yorkshire(GB)

(72) Inventor: Selwood, Alan
High Heighington Hall Heighington
Darlington Co Durham(GB)

(72) Inventor: Ralph, Donald Leslie
51 Malvern Road
Billingham Cleveland(GB)

(74) Representative: Chapman, Kenneth Hazel et al,
Imperial Chemical Industries PLC Legal Department:
Patents Thames House North Millbank
London SW1P 4QG(GB)

(54) **Extrusion.**

(57) Extrusion, especially of an aqueous ceramic compositions, is carried out under the influence of mechanical vibrations, preferably at ultrasonic frequencies in the range 15000 - 25000 Hz. This has advantages such as increasing the rate of extrusion at a given level of motive power or decreasing the extent to which compositions need to be preconsolidated before extrusion. Application of vibrations to the extrusion die facilitates cleaning after use. Extrudates can be cut cleanly by applying the vibrations to a cutting tool.

EP 0 045 564 A2

Extrusion

This invention relates to extrusion and in particular to a method and apparatus by which extrusion can be carried out under a degree of control not easily attained before.

According to the invention in its first aspect a method of extrusion comprises causing an extrudible material to flow through a die under the influence of mechanical vibrations.

As apparatus for carrying out this method the invention comprises an extrusion die, means to cause flow of an extrudible material through the die and means for applying mechanical vibrations to the material whereby to influence its flow through the die.

For the purposes of this specification the term "extrusion" includes both ordinary extrusion in which the material is pushed through the die by for example a ram or auger and also the operation in which its consistency is such that it can be pulled through the die or assisted by pulling, that is, so-called "pultrusion".

The effect of the vibrations is to decrease the force required to cause flow into and through the die. The mechanism by which this takes place is not known at present. It could, for example, result from loosening structures within the material, or by (for mixtures) liberating a low viscosity liquid or even gas at the interface of the material and the die. The invention is not however, limited to any particular mechanism.

Various practical results flow from using this aspect of the invention. In the simplest, less motive power is used. For the same power consumption the temperature of the material can be lower or (in the case of a material containing a volatile liquid) its viscosity can be greater and thus its liquid content can be lower, thus decreasing the energy consumption in drying the extrudate. In favourable conditions a composition normally extrudible only by ram owing to its high viscosity can by the use of the invention be auger-extruded: thus continuous operation is made possible. In the ceramics field it is expected to be found that so-called "high viscosity" compositions will be easier to extrude. Finally it is expected that certain ceramic compositions normally difficult to extrude uniformly in long lengths will no longer be so.

The extrudible materials to which the invention is believed to be applicable include molten polymers with solid fillers, rubber-like compositions, viscous solutions, and dispersions of solids in liquids possibly containing dissolved or swollen polymers. The invention is especially applicable to aqueous ceramic compositions containing for example one or more of the following solid phases: magnesia, alumina, silica, titania, zirconia, chromia and iron oxide and mixtures or compounds of two or more of these, for example cordierite and/or compounds capable of reacting together to form cordierite. If a hydrophilic polymer is present it is suitably one or more of the following: starch, cellulose and other carbohydrate ethers and esters, polyvinyl alcohols, ethers and esters, proteins, acrylic polymers, polyalkylene oxides. The molecular weight of the polymer can be chosen to suit the viscosity and organic content required. Instead or in addition other, non-polymeric, hydrophilic substances can be present, for example glycols, glycerol, sugars and sugar alcohols and ethers and esters thereof.

The invention is especially useful for that class of compositions that flow or deform plastically under shear during extrusion but become substantially rigid when not in motion.

Particular useful results of using the invention are that the following compositions can be extruded more uniformly and/or more rapidly than would otherwise be easily practicable:

(a) compositions containing a high proportion of clay as solid phase;

(b) compositions free or almost free of clay (under 5% $^w/w$ of the solids);

(c) compositions in which the particle size distribution leaves voids between larger particles under-filled or over-filled by smaller particles; and

(d) compositions requiring thorough preconsolidation for example by several passages through a pug-mill, to ensure adequate or constant homogeneity.

An example of a substantially clay-free composition to which the invention is applicable is one containing a perovskite type compound such as barium titanate or compounds reactable to such a compound on heating: it is important to minimise the clay content in order to ensure a positive coefficient of electrical resistance in the resulting extruded ceramic article.

In a die having discrete primary channels feeding into a unifying zone such as an annulus or a honeycomb, the vibrations may speed up flow through the primary channels, thus permitting the unifying zone to be kept full. In favourable conditions the vibrations so applied may make it unnecessary for the primary channels to have a larger aggregate cross-sectional area than the unifying zone. Alternatively the unifying zone land length can be decreased, thus decreasing further the pressure-drop through the die. If the vibrations are applied to the unifying zone and/or to any pooling area between the primary channels and the unifying zone, they can, in favourable conditions, assist unification by resolving non-uniformities caused the "memory" effect of anisotropic constituents of the composition being extruded.

In any extrusion operation a body of extrudible material is caused to undergo a change in cross-sectional shape and usually a decrease in cross-sectional area. The vibrations are applied preferably to at least one zone in which such a change or decrease is taking place. In such a zone preferably at least 10% $^{w}/w$ of the composition is moving with a component of velocity transverse to the direction of movement before entering that zone. Thus the vibrations are applied very suitably to the downstream zone, say 1 - 5 cm deep, of the barrel of a ram extruder or an auger. Another such application is to a pooling area or chamber of a multi-tube extrusion die.

In a further variant of the method the vibrations are applied differentially across the cross-section of the die or extrudate, to promote non-linear flow of extrudate. This can be done by using a highly directional beam of vibrations or by locally damping the vibration of the die. By this means it is possible to produce bent extrudates that can be readily wound into spirals or helices.

A useful property of the apparatus is the ease with which the die can be cleaned by immersing it in a liquid in which the extruded material is soluble or dispersible and applying the vibrations. Thus the invention provides a method of cleaning an extrusion die; it is especially valuable when the composition extruded is one that becomes substantially rigid when not in motion or when the die is complicated, as for extrusion of a honeycomb.

The frequency range of the vibrations effective to assist extrusion is large and is limited only by the need to match the applied vibrations with the natural frequencies of the apparatus used or the composition being extruded and to avoid the inconvenience and energy waste resulting from action on other media, including personnel. The acoustic range of 20 to 30000 c/s (Hz) can be considered typical of the usable frequencies, and the range 15000 - 25000 is highly convenient, and is especially effective for extruding compositions comprising a solid dispersed in a liquid. Vibration having more than one dominant frequency can be used, possibly applying a different frequency in different zones of the barrel, die and extrudate.

According to a further feature of the first aspect of the invention the method of extrusion includes stopping the flow by interrupting the vibrations and cutting the extrudate while it is stationary. By this means it is possible to make a cut exactly at a desired angle to the direction of extrusion and, in a multiple or honeycomb extruder, to cut all the extrudate at the same length. Since the composition can be stiffer than could be extruded in the absence of vibrations, the extrudate is firmer and less liable to distortion by the transverse force exerted by the cutting tool.

Since the interruption of vibrations applies an almost instantaneous brake to the flow of extrudible composition, the extruder should be mechanically designed to accommodate this without damage. For this purpose the drive to a ram or auger can include a slipping clutch or magnetic clutch, possibly synchronised with the interruption of vibrations. As an alternative an extruder barrel can include several outlet zones separately subjectible to the vibrations, so that the total throughput can be kept constant. Further, a single set of driving gear can drive several extruder barrels, a constant number of which are extruding at any instant. If desired, the braking effect can be accommodated by a damping system or energy-storage system such as a spring or air-buffer. If an auger is used, it can be one designed to permit slip or leakage internally during the interruption.

According to the invention in its second aspect a method of extrusion comprises causing an extrudible material to flow through a die and cutting the resulting extrudate by applying to it a vibrated cutting tool.

As apparatus for carrying out this method the invention comprises an extrusion die, means to cause flow of an extrudible material through the die, means for applying to the resulting extrudate tool means to cut the extrudate, and means to apply vibration to the tool during cutting.

Generally the frequency ranges are the same as for the first aspect of the invention. The vibrated cutting means can evidently be operated in synchronisation with the vibration source and other mechanical arrangements.

EXAMPLE 1

Effect of vibrations on extrusion force

A dry composition consisting of

alpha alumina	grade 1200	2000 g
	600	1850 g
	320	1150 g
Wyoming bentonite		550 g
Water-soluble starch		415 g

was mixed thoroughly in an orbital mixer for 18 minutes, then moistened by spraying with 820 g of water over 2 minutes. The wet mixture was passed twice through a 50 mm laboratory pug-mill to give a plastic mass. The mass was extruded from a ram barrel 1 inch in diameter through a die 6.2 mm in diameter and 5.5 mm long at a range of levels of applied force. It was found that a force of at least about 150 kg was necessary to effect any movement of the composition. This was then repeated with 100 watts of ultrasonic irradiation applied from a crystal transducer matched to an aluminium horn, suitably dimensioned so as to resonate at 19,800 hertz. The minimum force was now found to be only about 40 kg and at higher force levels the extrusion speed was much greater. The ram speeds as affected by ultrasonic vibrations are shown in Table 1.

Table 1

Ram speed cm min^{-1}	0.5	1.0	5.0
Extrusion force kg, without vibration	230	265	340
with vibration	145	210	290

An applied force above about 300 kg is evidently necessary to overcome the resistance to shear in the absence of vibrations, but a force of 250 kg is sufficient when the vibrations are applied. At an applied force of 140 kg or less extrusion was stopped almost instantaneously by stopping the vibrations and re-started almost instantaneously by re-starting them.

EXAMPLE 2

Effect of vibrations on extrudate cutting

For a simulation of this effect a block of the composition described in Example 1 was moulded on a movable platen fixed to the cross-head of an Instron testing machine. To the sides of a cutting blade was fixed an ultrasonic transducer similar to that used in Example 1. One end of the blade was supported on a load cell on a fixed platen beneath the movable platen. The movable platen was then advanced downwardly so that the blade penetrated the block at a speed of 1 cm min^{-1}, which speed was maintained constant. The resistance to penetration was measured by the load cell. The transducer was switched on and off at intervals. The resistance to penetration during application of vibrations was found to be substantially constant at a low level irrespective of the depth of penetration. In the absence of vibrations the resistance increased proportionally to the depth of penetration, from an initial value already substantially higher.

The same effect was observed with a penetration speed of 5 cm min^{-1}.

It is evident that the vibrations enable a blade to move rapidly through the extrudate, with low applied force and thus a low tendency to distort the extrudate. For many applications it is expected that a clean cut will be obtained even with a moving extrudate. If the extrudate is momentarily stationary as in Example 1, clean cuts even of relatively soft, otherwise fast-moving extrudates is possible.

EXAMPLE 3

(a) Example 1 was repeated but using a composition containing the alumina mixture, no clay or starch and, as hydrophilic polymer, a xanthan gum to the extent of 3% $^{w}/w$ on the alumina. Extrusion was carried out at 3 levels of applied force, resulting in 3 levels of extrusion rate. The effect of ultrassonic vibration was very large, as shown in Table 2.

Table 2

	Extrusion rate g min^{-1}		
Vibration intensity W	low force	medium force	high force
0	0	20	35
50	-	-	218
90	608	-	-
150	-	500	-
200	-	-	1200

(b) Example 3(a) was repeated with the following differences:

Extrusion aid "Zusoplast PS1" in place of starch;

Auger extruder with die having 36 holes 3.6 mm in diameter;

Vibration frequency 15900 Hz.

Using an applied force producing extrusion at 150 g min^{-1} in the absence of the applied vibrations, it was observed that this rate was increased to 850 g min^{-1} when the ultrasonic vibrations were applied at an intensity of 200 watts.

(Zusoplast PS1 is an organic polymer extrusion aid supplied by Zschimmer and Schwarz of Lahnstein am Rhein, Federal German Republic).

EXAMPLE 4

Example 3(b) was repeated but using the following composition:

Talc	2005 g
kaolin	2300 g
alumina mixture (as Example 1)	695 g
water soluble starch	1100 g
water	1100 g

and pug-milling it once. The effect of the vibrations was to increase the extrusion rate from 470 to 565 g min^{-1} or (at a higher applied force) from 500 to 740 g min^{-1}.

To clean the die it was immersed in water and subjected to the vibrations. The composition became very rapidly dispersed into the water and the die was substantially clean after 1 change of water.

EXAMPLE 5

Example 1 was repeated but using a 25.4 mm square honeycomb die having 512 cores, each of a right angled triangle cross-section. (The die was made from a single block of steel by making 15 cuts in each direction parallel to the sides, then making 15 diagonal cuts in each direction, so as to pass through alternate intersections of the cuts parallel to the sides. Each 8-fold intersection was in line with a feed channel from the opposite face of the block). The rate of extrusion was 12 cm min^{-1} without and 18 cm min^{-1} with the ultrasonic vibrations.

Note: each of the compositions used in the above Examples contained over 40% $^{w}/w$ inorganic solids and flowed plastically when under shear but became substantially rigid when not in motion.

PA/KHC/MP
19 June 1981

1. A method of extrusion which comprises causing an extrudible material to flow through a die under the influence of mechanical vibrations.

2. A method according to claim 1 in which the extrudible material is an aqueous ceramic composition.

3. A method according to claim 1 or claim 2 in which the extrudible material is a member of the class of compositions that flow plastically under shear during extrusion but became substantially rigid when not in motion.

4. A method according to any one of the preceding claims in which the vibrations are applied to the composition at a zone in which a change in cross-sectional area is taking place.

5. A method according to any one of the preceding claims in which the frequency of the vibrations is in the range 15000 - 25000 Hz.

6. A method according to any one of the preceding claims which comprises stopping the flow by interrupting the vibrations and cutting the extrudate while it is stationary.

7. An apparatus for carrying out a method according to any one of the preceding claims which comprises an extrusion die, means to cause flow of an extrudible material through the die and means for applying mechanical vibrations to the material whereby to influence its flow through the die.

8. A method of cleaning an extrusion die after use which comprises immersing it in a liquid in which the extruded composition is soluble or dispersible and subjecting it to mechanical vibrations of frequency 20 - 30000 Hz.

9. A method of extrusion which comprises causing an extrudible material to flow through a die and cutting the resulting extrudate by applying to it a vibrated cutting tool.

10. An apparatus for carrying out a method according to claim 9 which comprises an extrusion die, means to cause flow of an extrudible material through the die, means for applying to the resulting extrudate tool means to cut the extrudate and means to apply vibration to the tool during cutting.