Sept. 18, 1962 P. B. ARCHIBALD 3,054,147

METHOD FOR SOLVENT-ISOSTATIC PRESSING

Filed Dec. 30, 1960

Fig. 1.

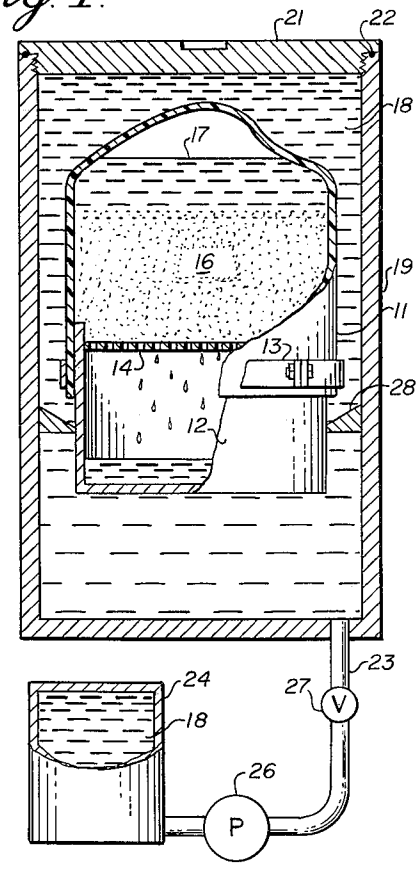


Fig. 2.

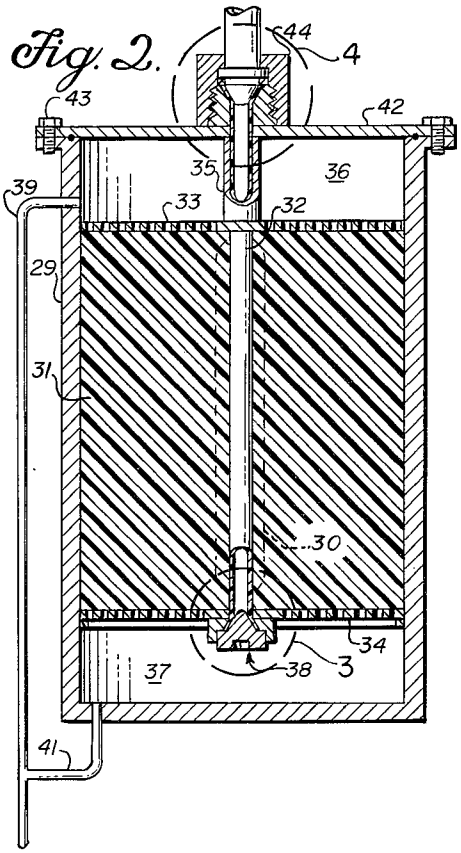


Fig. 4.

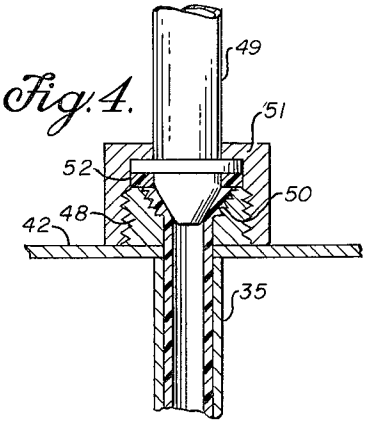
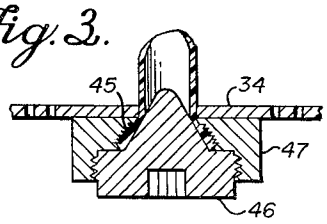


Fig. 3.



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METHOD FOR SOLVENT-ISOSTATIC PRESSING

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This invention relates to the production of compacted bodies and, more particularly, to a method for the production of large compacted bodies by a low pressure pressing operation.

Ram-and-die pressing operations are usually limited to the production of relatively small objects, since the dies required for large objects are prohibitively expensive. In order to produce large compacted bodies, isostatic pressing is usually resorted to. In the practice of conventional isostatic pressing, the material to be compacted is placed in powder form inside a strong flexible bag. The bag is evacuated, sealed, and immersed in a fluid medium contained within a pressure vessel. Pressure is then applied to the fluid by means of a pump external to the vessel, and the applied pressure is transmitted uniformly by the fluid over the surface of the flexible bag. The bag, in turn, compresses the powder within it to produce a homogeneous compacted body. Although a useful technique, isostatic pressing suffers from the disadvantage that it must be performed at high pressure in order to adequately compact the powdered material. Pressures of 10,000 to 50,000 pounds per square inch are commonly used in industrial isostatic pressing. High pressure operation requires heavy thick-walled apparatus, and conventional isostatic presses are generally large expensive pieces of equipment.

The present invention provides a method for conducting isostatic pressing operations at pressures substantially below those required by the prior art. Briefly, the method of the invention comprises the addition of a limited amount of suitable solvent to the powder which is to be compacted isostatically. The solubility of the powder granules in the solvent is highest at the points of greatest mechanical stress, i.e., the points of contact between adjacent granules. Therefore, the solvent effects a localized solution at these contact points. When this saturated solvent flows into the void spaces in the powder, wherein the mechanical stress is less than that at the contact points, the dissolved material is redeposited in the void spaces. The resulting decrease in total void space enables the pressing operation to be carried out at a much lower pressure than would be possible in the absence of any solvent. A pressure of 2,000 p.s.i. is typical in the practice of the invention; this corresponds to 5–10% of the pressure commonly used in conventional isostatic pressing. As a result of the low pressures employed, the present process makes possible the use of relatively light and inexpensive pressing equipment. Apparatus for performing the method of the invention is described hereinafter.

Accordingly, it is an object of the invention to provide a method for producing large homogeneous compacted bodies.

Another object of the invention is to provide a method for reducing the pressures required in isostatic pressing operations.

The invention will be described with reference to the accompanying drawings, of which:

FIGURE 1 is a cut away view of an apparatus for conducting solvent-isostatic pressing;

FIGURE 2 is a cross sectional view of an apparatus for the production of tubular compacted bodies by solvent-isostatic pressing;

FIGURE 3, is an enlarged view of plug assembly 38 shown in FIGURE 2; and

FIGURE 4 is an enlarged view of pressure coupling 44 shown in FIGURE 2.

Referring now to FIGURE 1, there is shown a flexible bag 11, which is securely clamped at its open end to a rigid container 12 by means of an annular clamp 13. A finely perforated plate 14, of a diameter equal to that of container 12, is disposed across the open end of the container, and is secured to the walls thereof. Within flexible bag 11 are disposed a granular charge material 16 and a solvent 17 in mutual contact. The assembly of flexible bag 11 and the contained solvent-granular material charge, and rigid container 12 is immersed in a fluid medium 18 contained within a pressure vessel 19. A tightly fitting threaded plug 21 and a pressure gasket 22 seal the opening of vessel 19, and a fluid conduit 23 communicates with the interior thereof. Fluid medium 18 is supplied to the pressure vessel from a fluid reservoir 24 connected in series with a high pressure pump 26. A pressure regulator 27 is disposed between pump 26 and vessel 19, so that a constant pressure can be maintained in the vessel's interior. Support ring 28 serves to position the bag-container assembly within the pressure vessel.

In the operation of the apparatus illustrated in FIGURE 1, a measured volume of solvent 17 is poured into flexible bag 11, and the granular charge material 16 is added thereto. The bag is clamped to rigid container 12, and the assembly is inverted and positioned within vessel 19 by means of support ring 28. Pump 26 is switched on, and hydraulic fluid from reservoir 24 is forced under pressure into vessel 19. When the pressure in vessel 19 has reached the desired value, regulator 27 is adjusted to maintain this pressure. The increased fluid pressure inside vessel 19 constricts flexible bag 11, thereby causing solvent 17 to percolate through the charge material 16, dissolving the granules at their contact points and redepositing the dissolved material in the void spaces of the powder. As compaction proceeds, the solvent is ultimately squeezed completely through and thereafter from the charge material and subsequently flows through perforated plate 14 into the closed end of rigid container 12. Granules of the charge material are prevented from falling into container 12 by the filtering action of plate 14. After the charge material has been under pressure for the required length of time, the pressure in vessel 19 is released and the compacted object is recovered from bag 11.

The main advantage of the solvent-isostatic pressing process over conventional isostatic pressing is that the presence of a solvent in the material to be compacted enables the pressing operation to be performed at a much lower pressure than would be possible in the absence of any solvent. The solubility of the material to be compacted is greatest at the points of contact between adjacent granules when the granules are under mechanical pressure. Therefore, when pressure is applied to the charge material, the solvent effects a localized solution at the contact points between adjacent granules, and redeposits the dissolved material in the regions of least mechanical stress, i.e., the void spaces between the individual particles. As the applied pressure is increased, the solubility of the granules at their contact points correspondingly increases, and the available void space becomes filled with redeposited material. It is apparent that the

solvent produces a degree of homogeneity in the resulting compact that is virtually impossible to achieve in conventional "non-solvent" isostatic pressing operations.

Typical pressing times at room temperatures are on the order of 10–50 hours, however, in many cases, the time required to compact the charge can be substantially reduced by carrying out the pressing operation at elevated temperature. This can be done by heating the hydraulic fluid in the reservoir, or by disposing an immersion-type heater in the pressure vessel. The bag-container assembly shown in FIGURE 1 is a closed system, so that the pressing operation can be conducted at temperatures well above the atmospheric boiling point of the solvent.

The relative proportion of solvent to charge material is critical to the successful operation of the pressing process. If insufficient solvent is added to the charge material, the solvent will not distribute uniformly among the granules, and the resulting compact will contain considerable void space. In addition, the length of time required to produce a satisfactory compact is considerably increased. If, on the other hand, too much solvent is added before pressing, a semi-fluid slurry is formed which results in the production of a badly deformed compact. It has been found that the optimum solvent concentration lies in the range of 5–15% by weight of the dry charge material.

The only restriction on the chemical composition of the solvent to be used in the process of the invention is that there be no chemical reaction between the solvent and the charge material. Satisfactory compacts have been obtained using water, acetone, ethanol, methanol, dioxane, and various mixtures thereof as solvents. If the material to be compacted is very soluble in the particular solvent selected, then substantial amounts of the charge will dissolve in the solvent and, unless recovered therefrom, will be lost. However, this loss of charge material can be minimized by pre-saturating the solvent with the charge material, and using this saturated solvent in the pressing operation. Since the solvent is already saturated with the charge material, a dynamic equilibrium between solvent and solute is set up, the rate of solution into the solvent being equal to the rate of deposition out of the solvent. In this manner, the use of a saturated solvent will minimize material losses in the solvent, while still effectively decreasing the void space in the finished compact.

In many cases, mixtures of organic and inorganic materials can be successfully solvent-isostatically pressed, provided that a solvent can be found in which either the organic or the inorganic component is appreciably soluble. For example, ammonium nitrate is soluble in acetone to the extent of 17 grams per 100 milliliters of acetone. This solubility affords a means whereby ammonium nitrate can be pressed in admixture with any organic or inorganic material. In the solvent-isostatic pressing of mixtures, as well as in the pressing of single compounds, the loss of charge material in the solvent can be minimized by saturating the solvent before use in the pressing operation. For the pressing of mixtures, the solvent is first saturated separately with each of the pure components comprising the mixture. This insures that the solvent will not dissolve any of the components selectively during the pressing operation.

It will be apparent from the foregoing discussion that the application of pressure to the charge mixture causes the individual granules therein to consolidate themselves into a closely packed structure. The closest packing of a particulate mass is obtained when the diameters of the particles therein are distributed according to a Gaussian frequency distribution, i.e., where there are many different particle diameters symmetrically distributed about a mean diameter. For purposes of the present invention, such a particle distribution can be obtained through ball milling or grinding the particles in a mortar. This procedure insures that the particles will pack together with a minimum of void space when pressure is applied. It has been found that the optimum mean particle diameter for solvent-isostatic pressing lies between six and ten microns. Powders having mean particle diameters substantially greater than ten microns require pressing times which may be too long to be practicable.

Although the main advantage of solvent-isostatic pressing is that it can be performed at pressures substantially below those required by conventional isostatic pressing, it is sometimes advantageous to employ a high pressure press in order to reduce the pressing time. In such cases, the addition of solvent before pressing will still produce a denser and more homogeneous compact than would be obtained by a non-solvent pressing at the same pressure.

The process of the invention can be used to advantage for the pressing of many different materials. The only restrictions on the nature of the charge material are that it can be obtained as a crystalline powder preferably having a mean particle diameter of six to ten microns, and that a suitable solvent is available for the powder. Materials which have been successfully pressed include; crystalline organic compounds, e.g., trinitrobenzene, inorganic salts such as ammonium nitrate, and mixtures thereof with powdered metals, specifically, powdered aluminum.

The solvent-isostatic pressing process can be used for the production of variously shaped compacts by simply disposing a suitably shaped mandrel within the flexible bag, and disposing the solvent and charge material around the mandrel. Aluminum mandrels have been used to press hollow hemispheres, cylindrical shells, and other simple geometrical shapes. An apparatus for the production of tubular compacts is illustrated in FIGURE 2, wherein there is shown a tubular housing **29** enclosing a solvent-moistened charge material **31**. An inflatable elastomeric tube **32** is axially disposed within housing **29**, and is surrounded along its length by charge material **31**. The inflated position of tube **32** is indicated by the dotted outline **30**. Perforated plates **33** and **34** are disposed within housing **29** so as to divide the interior of the housing into three chambers; an upper solvent recovery chamber **36**, a middle chamber containing the solvent-moistened charge material, and a lower solvent recovery chamber **37**. A supporting sleeve **35** is fixed to the upper surface of plate **33**, and tube **32** passes therethrough. Lower plate **34** rests on an annular shoulder formed into the interior wall of housing **29**, and upper plate **33** rests directly on the charge material **31**. The lower end of tube **32** is secured to plate **34** by means of a pressure plug assembly **38**. Solvent recovery pipes **39** and **41** communicate with chambers **36** and **37** respectively. A flanged cover **42** is bolted to the open end of housing **29** by means of circumferentially disposed bolts **43**. The lower surface of cover **42** bears against the upper end of sleeve **35**. Inflatable tube **32** passes through cover **42** and is secured thereto by pressure coupling **44**. The coupling also serves to connect tube **32** to an external source of hydraulic fluid under pressure (not shown). Tube **32** is preferably fabricated from a synthetic elastomer, e.g., neoprene.

A detailed view of plug assembly **38** is shown in FIGURE 3. A threaded conical plug **46** is screwed into a correspondingly threaded boss **47** which is secured to the lower surface of plate **34**. The conical surface of plug **46** bears against tube **32** and deforms the tube into serrations **45** formed in boss **47**. Tube **32** is thereby tightly sealed at its lower end to plate **34**.

A detailed view of pressure coupling **44** is shown in FIGURE 4. An externally threaded female fitting **48**, having a conical bore, is fixed to the outer surface of cover **42**. A male fitting **49**, having an external taper corresponding to the conical bore of fitting **48**, bears against inflatable tube **32**, and deforms the tube into serrations **50** formed into the bore of fitting **48**. A coupling nut **51** adjusts the pressure of fitting **49** against tube **32**, and hence adjusts the tightness of the pressure seal. An O ring **52** is disposed between fittings **48** and **49**. The ex-

ternal source of hydraulic fluid under pressure is connected to fitting **49**.

In the operation of the apparatus illustrated in FIGURES 2–4, tube **32** is first attached to plate **34** by means of plug assembly **38**. Plate **34** is lowered into place inside housing **29** until the plate rests on the annular shoulder formed into the interior wall of the housing at its lower end. A solvent-moistened charge material **31** is packed into the housing surrounding tube **32**, and the tube is threaded through plate **33** and attached sleeve **35** which are placed on the upper surface of the charge material. Cover **42** is then bolted on and tightened until the lower surface of the cover exerts a slight pressure against the upper end of sleeve **35**. Hydraulic fluid under pressure from the external reservoir is admitted to tube **32** via coupling **44**. Tube **32** inflates to the position indicated by the dotted outline in FIGURE 2 and, in so doing, compresses the charge material against the interior wall of housing **29**. Solvent is squeezed out of the charge material and flows through plates **33** and **34** into the solvent recovery chambers **36** and **37**. Solvent is recovered from the system through pipes **39** and **41**. After all the solvent has been squeezed out of the charge material, and the hydraulic pressure has been released, the resulting tubular compact is recovered.

The apparatus shown in FIGURE 2 is especially suited to the production of compacted bodies having symmetry about a central axis, e.g., rocket propellant grains. The interior wall of housing **29** can be suitably machined so as to shape the outer surface of the compacted body to any desired configuration. For example, if a cruciform configuration is desired, four longitudinal ribs can be fixed to the interior wall of the housing.

Further details of the solvent-isostatic pressing process are given in the following examples. In each of these examples, the apparatus illustrated in FIGURE 1 was employed for the pressing operation.

Example I

Chemically pure ammonium nitrate was ball milled for three hours. The resulting powder was examined under a microscope, and the average particle diameter was determined to be eight microns. Twenty-four grams of the ammonium nitrate powder were added to two cubic centimeters of methanol, and the mixture was pressed at room temperature for twenty hours at 2,000 pounds per square inch. The density of the resulting ammonium nitrate compact was 1.70 grams per cubic centimeter, corresponding to 98.7% of the theoretical maximum density (1.725 gms./cc).

Example II

Ten grams of ammonium perchlorate powder, having a mean particle diameter of nine microns, were mixed with one gram of a 1:1 solution of acetone in methanol. The mixture was pressed at room temperature for sixty hours at 2,000 pounds per square inch. The density of the resulting ammonium perchlorate compact was 1.948 grams per cubic centimeter, corresponding to 100% of the theoretical density.

Although several embodiments and examples of the invention have been described herein, these are intended to be merely illustrative, and various modifications can be made therein without departing from the spirit and scope of the invention as defined in the following claims.

What is claimed is:

1. A process for compacting materials, comprising the steps of, contacting a finely granulated material with a solvent in which said material is at least partially soluble, placing said granulated material and the contacting solvent into a deformable container, simultaneously compacting said material and separating the solvent therefrom by applying hydrostatic pressure to the exterior of said deformable container, and recovering said compacted material from within said deformable container.

2. A process for compacting materials, comprising the steps of, contacting a crystalline powder with a solvent in which said powder is at least partially soluble, placing said powder and the contacting solvent into an elastomeric bag in communication with a rigid container, sealing said bag, immersing said bag with the contained powder and solvent in a fluid medium contained within a pressure vessel, exerting force against the exterior surface of said bag by applying hydrostatic pressure to said fluid medium whereby the powder inside the bag is compacted and the solvent separated therefrom, and recovering said compacted body from within said elastomeric bag.

3. The process defined in claim 2, wherein the solvent is selected from the group consisting of water, methanol, ethanol, acetone, dioxane, and mixtures thereof.

4. The process defined in claim 2, wherein the weight of the added solvent lies between 5 and 15% of the dry weight of the powder.

5. The process defined in claim 2, wherein the average particle diameter of the powder lies between 6 and 10 microns.

6. The process defined in claim 2, wherein the powder is a crystalline inorganic salt.

7. The process defined in claim 2, wherein the powder is a mixture of a crystalline inorganic salt and a powdered metal.

8. The process defined in claim 2, wherein the powder is a crystalline organic compound.

9. The process defined in claim 2, wherein the powder is a mixture of a crystalline organic compound and a powdered metal.

10. A process for compacting materials, comprising the steps of, saturating a solvent with a material to be compacted, disposing said saturated solvent in contact with said material in the form of a crystalline powder, placing said powder and the contacting solvent into an elastomeric bag in communication with a rigid container, sealing said bag, immersing said bag with the contained powder and solvent in a fluid medium contained within a pressure vessel, exerting force against the exterior surface of said bag by applying hydrostatic pressure to said fluid medium whereby the powder inside the bag is compacted and the solvent separated therefrom, and recovering said compacted body from within said elastomeric bag.

11. A process for compacting materials, comprising the steps of, contacting a crystalline powder with a solvent in which said powder is at least partially soluble, placing said powder and the contacting solvent into an elastomeric bag in communication with a rigid container, sealing said bag, immersing said bag with the contained powder and solvent in a fluid medium contained within a pressure vessel, heating said fluid medium, exerting force against the exterior surface of said bag by applying hydrostatic pressure to said heated fluid medium whereby the powder inside the bag is compacted and the solvent separated therefrom, and recovering said compacted body from within said elastomeric bag.

12. The process defined in claim 11, wherein the fluid medium is heated to a temperature above the atmospheric boiling point of the solvent.

13. A process for producing compacted bodies of tubular shape, comprising the steps of, contacting a finely granulated material with a solvent in which said material is at least partially soluble, packing said material and the contacting solvent around and in contact with an inflatable member disposed within a rigid housing, expanding said inflatable member so as to cause the exterior surface thereof to bear against and compact said granulated material into a substantially tubular body, and recovering said compacted tubular body from within said rigid housing.

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