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(54) **Improvements in or relating to fluid seals**

(57) A method is provided for manufacturing a moving part capable of providing a liquid seal. The method comprises the steps of: firstly forming the part from sintered steel; and secondly forming a layer of Fe_3O_4 on an un-ground surface of the part to provide a surface finish that is capable of providing a liquid seal.

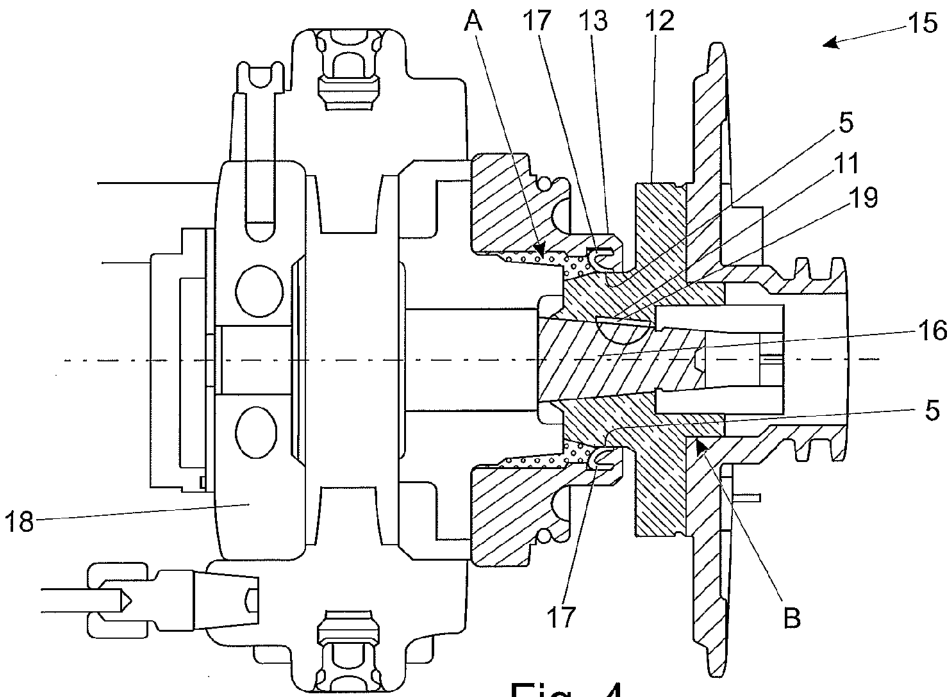

Fig. 4

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Description

[0001] The present invention is concerned with improvements in or relating to fluid seals and particularly, but not exclusively, to a method of manufacturing a part or component that has a surface capable of providing a fluid seal in use and to a part manufactured using such a method. The invention may find advantageous application in the manufacturing of rotating parts for vehicle engines that provide such fluid sealing surfaces. Aspects of the invention relate to a method, to a part, to a fluid seal surface, to a product, to a sealing arrangement and to a vehicle.

[0002] There are many different situations with the automotive industry in which there is a requirement to maintain a fluid seal, and particularly a liquid seal. Amongst these varied applications, there are several examples of situations in which a moving, typically rotating, part traverses a boundary between a liquid filled area and a non-liquid filled, or dry, area or between areas containing dissimilar liquids that must be kept separate. Examples of such parts are components for fluid pumps, crankshafts, engines or transmissions, amongst others.

[0003] These parts are typically made by machining from a single billet of material. The billet must have a minimum diameter that is at least equal to the maximum diameter of the finished part. This process produces a considerable amount of wastage in the form of scrap material. For complex shapes the machining process requires frequent resetting of the machine because it has to be re-set for each new face. Each face is then ground in order to give a sufficiently polished surface finish for a liquid seal to be formed. This process is very time intensive and is therefore costly. By way of example, the cost of grinding the part to produce the polished surface can constitute more than 50% of the total cost of manufacturing the part.

[0004] In order to form a liquid seal within an engine sub-system, the connecting part is typically provided with a sealing member or gasket typically formed from a plastics or polymeric material such as rubber. Any unevenness on the surface abutting the sealing member will result in the premature deterioration of the plastics or rubber seal. Many engine sub-systems are supplied as sealed units and, in these circumstances, it may not be possible to replace a damaged rubber seal. Instead, the entire sub-system must be replaced. Unevenness on the surface abutting the rubber seal may therefore decrease the effective lifetime of the sub-system as a whole. This may also adversely affect other sub-systems or components of the engine due to oil or coolant loss.

[0005] Alternatively, it is known to use a soft metal pressed insert fabricated from copper, phosphor-bronze or a similar material, in order to provide a sufficiently smooth surface to form an effective seal.

[0006] It is an aim of the invention to improve upon such known technology and manufacturing processes. Embodiments of the invention may provide a substantial reduction in the complexity and cost of the process by which such parts are manufactured. Other aims and advantages of the invention will become apparent from the following description, claims and drawings.

[0007] In one aspect of the invention there is provided a fluid seal arrangement (17) comprising a sealing member formed from a plastics or polymeric material and a part in sealing cooperation therewith, wherein the part is characterised by a body (10) formed from a sintered ferrous material having at least one surface (5) arranged to provide a fluid seal (17), in use, **characterised in that** the at least one surface (5)comprises an un-ground surface of the ferrous material coated with a layer of Fe304 wherein the part comprises one of a coupling (10) for a fuel injector (15), a viscous fan (70) for a cooling system of an internal combustion engine, a water pump (80), an oil seal retainer (90) for use in sealing the ends of a crankshaft (91), differential transmission, gearbox, axle, driveshaft (82, 92, 102), hub (13) or transfer case; and a vacuum pump (110) for use in the braking system of a vehicle.

[0008] The present applicant has identified that, contrary to established manufacturing practices, a fluid seal with satisfactory performance characteristics can be achieved simply by depositing a layer of Fe_3O_4 onto an un-ground surface of a sintered ferrous part without the need for grinding. This surprising technical effect permits the elimination of the normal step of grinding the surface to provide a polished surface and hence significantly reduces the complexity, time and cost of manufacturing the part.

[0009] The term "un-ground surface" as used herein is intended to mean a surface of the ferrous part which has been created by direct contact with the sintering tool and has not been subject to any further smoothing or polishing process, such as grinding, abrading or machining. This surface is also known as a raw surface or virgin surface of the part.

[0010] In addition, when a part is machined (for example by grinding), the performance of the part depends critically on the tolerances of each individual face. As each face has to be machined separately, each one could be subject to an error in the positioning of the grinding machine. In contrast, the sintering process of the present invention is a single step of sintering to produce the majority of the facets of the part. This results in a much more reliable process.

[0011] According to another aspect of the invention for which protection is sought there is provided a method of manufacturing a sintered ferrous part having at least one surface configured to provide a fluid seal, in use, the method comprising depositing a layer of Fe_3O_4 onto an un-ground surface of the part to as to form the at least one fluid sealing surface.

[0012] According to yet another aspect of the invention for which protection is sought, there is provided a method of forming a fluid seal surface on a sintered ferrous part, the method comprising depositing a layer of Fe_3O_4 onto an un-ground surface of the sintered ferrous part so as to form the fluid seal surface.

[0013] In an embodiment, the method comprises forming the part from a sintered ferrous material before depositing the Fe_3O_4 layer.

[0014] In an embodiment, depositing the Fe_3O_4 layer comprises steam treating at least the un-ground surface of the part. Alternatively, or in addition, the entire part may be subjected to steam treatment so as to deposit the layer substantially fully thereover.

[0015] According to a further aspect of the invention for which protection is sought there is provided a sintered ferrous part having at least one surface arranged to provide a fluid seal, in use, wherein the at least one surface comprises an un-ground surface of ferrous material coated with a layer of Fe_3O_4.

[0016] According to a still further aspect of the present invention for which protection is sought there is provided a method of manufacturing a moving part capable of providing a liquid seal, the method comprising the steps of forming the part from sintered steel and forming a layer of Fe_3O_4 to provide a surface finish on the sintered part that is capable of providing a liquid seal.

[0017] Furthermore, according to another aspect of the invention for which protection is sought, there is provided a moving part providing a liquid seal for use in a vehicle engine, the part comprising a sintered body and a steam treated surface finish for providing the liquid seal. The moving part may be a rotating part.

[0018] The method may comprise the step of machining complex structures, such as keyways, and screw threads within the part. These structures may be created separately from the sintering step since they generally do not include the surfaces that provide a liquid seal.

[0019] The part may be, inter alia, a coupling for an injection pump for a fuel injection unit in a diesel engine, a viscous fan from a radiator system of an internal combustion engine, a water pump for use in an internal combustion engine, an oil seal retainer for use in closing the ends of a vehicle crankshaft, an oil seal retainer for sealing a differential transmission, gearbox or transfer case or a vacuum pump for use in the braking system of a vehicle. Other examples will be recognised by the skilled person.

[0020] Within the scope of this application it is envisaged that the various aspects, embodiments, examples, features and alternatives set out in the preceding paragraphs, in the claims and/or in the following description and drawings may be taken individually or in any combination thereof.

[0021] The present invention will now be described, by way of example only, with reference to the accompanying drawings in which:

Figure 1 shows an axial view of a coupling for a fuel pump manufactured according to the method of the present invention;

Figure 2 shows a cross sectional view of the coupling shown in Figure 1;

Figure 3 shows a perspective view of the coupling shown in Figures 1 and 2;

Figure 4 shows the coupling of Figures 1 to 3 in context with those parts to which it is most closely attached, in use;

Figure 5 shows an exploded view of the relevant part of a diesel engine showing the position of the coupling of Figures 1 to 3, in use;

Figures 6a to 6e show a series of 30X magnification view of sintered parts with and without machining, pre- and post-durability testing;

Figure 7 shows a viscous fan from a radiator system of an internal combustion engine;

Figures 8a and 8b show exterior and interior views of a water pump for use in an internal combustion engine;

Figure 9 shows an oil seal retainer for use in closing the ends of a vehicle crankshaft;

Figure 10 shows an oil seal retainer for sealing a differential transmission, gearbox or transfer case; and

Figure 11 shows a vacuum pump for use in the braking system of a vehicle.

[0022] Sintering is a known method for making objects from a suitable material, usually a ceramic or metallic powder, by heating the material (below its melting point) until its particles adhere to each other and then applying pressure to the material by compressing it in a sintering tool. The sintering tool typically takes the form of a rigid metallic block with an internal cavity or void corresponding in shape to the desired final part. The sintering tool operates in a manner similar to that of a moulding tool whereby the material is forced into the mould cavity at high pressure so as to ensure that the material flows evenly into each void of the tool cavity. Once the material has cooled and set, the sintering tool is opened and the part removed.

[0023] Sintering is traditionally used for manufacturing ceramic objects and sintered bronze is frequently used as a material for bearings. Sintered bronze is suitable for use in bearings since its porosity allows lubricants to flow through it. In the case of materials with high melting points such as tungsten, sintering is used when there is no alternative manufacturing technique.

[0024] The designer of a metallic part may decide to use sintering over casting or billet machining for several reasons. The desired part may require a particular crystalline structure which cannot always be achieved with casting. The part may be desired in large numbers and so the percentage of material wastage associated with billet machining may not be cost effective. The dimensional tolerance of the part may be critical between two or more features of the part.

[0025] Sintering, on the other hand, produces parts with dimensional accuracy close to the original sintering tool with excellent repeatability. Some design features may not be created easily by sintering such as screw threads but once a part has been sintered further processes may be employed to achieve the desired final shape. Once a part has been sintered it may be machined, ground or cut as required to create surface features that cannot be formed by sintering alone.

[0026] In some instances, the function of a part may be enhanced if it is porous. For example if a part is bathed in lubricant, some of the lubricant will be trapped by the open pores of the sintered part which reduces wear. However, the porosity of ferrous sintered parts make them prone to corrosion if the part is exposed to water which may be unsuitable for some applications.

[0027] The surface of the sintered ferrous part created by the direct contact between the metallic powder forced into the tool and the surface of the tool cavity walls is known as the raw or virgin surface. In other words, the raw or virgin surface of the sintered part has not been subjected to any further smoothing and/or polishing treatment, such as machining, abrading or grinding, after removal from the sintering tool, and for this reason is referred to hereafter as the "un-ground surface". The un-ground surface tends not to be particularly hard-wearing and may be porous. If the final part must be non-porous or if greater durability is required then the ferrous part may require further surface treatment after sintering.

[0028] One such surface treatment is known as steam treatment. Steam treatment is a term of art referring generally to a coating technique that is typically employed when a part needs to be hardened. When handling ferrous parts, the technique generally results in the deposition of a uniform layer of Fe_3O_4, or "black iron" on the treated surface or surfaces of the part. Fe_3O_4 is a crystalline solid, meaning that a surface formed from Fe_3O_4 is hard and has a low porosity.

[0029] This deposition layer of Fe_3O_4 is typically 2 - 10 μm in thickness, but this may vary due to the quality of the un-ground surface or production process parameters such as temperature and duration of exposure to the steam. The resulting part has a black appearance and, in addition to an increased hardness, is also more resistant to corrosion than a similar part that has not been so treated.

[0030] The surface of a sintered ferrous part with a coating of Fe_3O_4 is far less porous as the Fe_3O_4 crystals are very fine and has the effect of uniformly filling the pores, especially in the crystalline surface structure of the sintered part. Steam treatment does not simply block the pores, but also increases the corrosion resistance and alters the mechanical properties, thus increasing the density, the hardness and the tensile strength of the sintered part and thus improving wear properties.

[0031] Conventional engineering practice requires that, for metallic parts, a fluid seal surface on a rotating part should be machined and ground to achieve the desired surface finish and fine dimensional tolerance required to provide a reliable fluid seal. However, the present applicant has discovered that, by suitable design of the sintering tool in the area of the fluid seal, the dimensional tolerances and surface finish of the sintered part are capable of providing a fluid seal once the part has been steam treated, without the need for an intermediate machining or grinding step.

[0032] The omission of the machining and grinding step for such a part represents a considerable time and cost saving and can additionally enhance performance of the part. For example, any machining or grinding process has to be carefully monitored to ensure sufficient repeatability and dimensional tolerances between features of the part such as mounting points and fluid seals. However, according to the present invention, the final shape of the part is achieved directly from the tool, thereby requiring fewer manufacturing operations and advantageously providing tighter dimensional tolerances.

[0033] Extensive testing by the applicant has been conducted to compare the fluid sealing performance of conventionally-machined parts and parts formed in accordance with the present invention and examples are shown in Figures 6a to 6e. This testing has shown that, contrary to conventional belief that a surface must be machined and/or ground in order to be suitable for a fluid seal, a reliable fluid seal may be produced by depositing a layer of Fe_3O_4 directly onto the un-ground surface created by the sintering tool.

[0034] The applicant has further discovered that the surface finish created by the deposition of Fe_3O_4 onto the raw sintered surface has been shown to be particularly suitable for use with sealing members made from plastics or polymeric materials such as rubber.

[0035] Provided that the surface quality of the sintering tool cavity is sufficiently smooth, the deposition of Fe_3O_4 directly onto the un-ground surface of the sintered ferrous part, for example by means of steam treating, produces a surface that is sufficiently smooth, durable and non-porous to form a reliable fluid seal without the need for additional machining or grinding processes. This represents a significant improvement in production efficiency as grinding a surface for a fluid seal represents a considerable proportion of the manufacturing time and cost associated with each part.

[0036] Figures 1 to 5 show a coupling 10 for the type of fuel pump used in a diesel engine. The coupling 10 is fabricated from, for example, M1040 steel. The grade of steel is chosen to be suitable for the particular application. The choice is a compromise between cost and weight to produce a part suitable for the mechanical loads whilst still providing a sufficient safety margin.

[0037] The coupling 10 is shown in isolation in Figures 1, 2 and 3. The coupling 10 comprises a flange 12 and a boss or hub 13. The flange and hub 13 both have a circular cross-section. The manufacturing tolerance on the diameter of the hub 13 is extremely tight to ensure that the coupling 10 is capable of interfacing to provide an effective seal. A fluid seal surface 5 is provided around the outer surface of the hub 13.

[0038] Figure 4 shows the coupling 10 in the context of a fuel pump system 15 including a fuel pump 18. Figure 5 contextualises the fuel pump system 15 within the engine as a whole. The coupling 10 is driven to rotate by the insertion of a shaft 16 into a keyed conical hole 11 with a keyway 19. The shaft 16 is also keyed and rotation results when a key is provided in joining the respective keyways 19. The hole 11 tapers through the hub 13 and at least partially through the flange 12. The coupling separates a liquid filled area A from a non-liquid filled area B. The seal is provided on the liquid filled side A of the coupling 10, by a rubber sealing member 17 which is seated on the fluid seal surface 5 of hub 13. Holes 14 are provided through the flange 12 to enable the coupling 10 to interface with other engine parts in the non-liquid filled area B.

[0039] The coupling 10 is manufactured in a three-step process: firstly, the body of the part is formed by sintering; next, the holes 14 in the flange 12 are tapped in known manner; finally, the entire part is steam treated to form the Fe_3O_4 coating. The steam treatment is used to harden the surface, reduce porosity and improve the corrosion resistance of the part. According to conventional processes, an additional grinding step would be performed on the part before the part is subjected to the steam treatment in order to provide a surface that is sufficiently smooth to provide a liquid seal. However, the present applicants have identified that the operation of steam treating the surface obviates the need for any grinding.

[0040] Looking at a microscopic level the cross-section of a machined part consists of a series of regularly spaced and shaped peaks. In contrast, the surface resulting from the steam treatment has an almost completely smooth surface with occasional microscopic peaks that result from random positioning of crystals. The smooth surface results in a much larger proportion of the surface area being available for interfacing with the seal. The occasional peaks are typically worn away by the interfacing part before any damage occurs to that part. The smooth surface provides an advantage for the lifetime of the seal.

[0041] In particular, the smooth outer surface of the hub 13 provides the fluid seal surface 5 and ensures that damage to the seal 17 is reduced, thus increasing the effective life of the fuel pump system 15.

[0042] Figures 6a to 6e show a series of 30X magnification view of sintered parts with or without machining, pre- and post-durability testing.

[0043] Figure 6a shows a sintered part at 30X magnification. The porous surface is readily apparent. This surface can be made more uniform by machining, as shown in Figure 6b. This provides a good surface finish, but the surface is porous and is therefore unsuitable for providing a liquid seal.

[0044] Figure 6c shows a non-machined sintered part that has been subjected to durability testing. The wear apparent on the surface is typical of that expected in such a test.

[0045] Figure 6d shows the sintered part of Figure 6a when it has been steam treated, i.e. in accordance with an embodiment of the present invention. The surface is very smooth and non-porous. The surface also has a very high surface hardness. Figure 6e shows the part from Figure 6d once it has been subjected to a durability test. It will be readily apparent that the wear is reduced in comparison with the sintered part shown in Figure 6c which had not been steam-treated.

[0046] Figure 7 shows a viscous-drive 70 for a cooling fan for an internal combustion engine cooling system. The viscous-drive 70 transmits drive to a cooling fan (not shown) from the engine in response to changes in the ambient air temperature surrounding the body of the viscous-drive. The viscous-drive 70 comprises a housing 71, a bi-metallic controller (not shown), an input shaft (not shown) and an output shaft 72. The input and output shafts 72 are separated by the viscous-drive 70. The housing 71 is secured to the engine or radiator by means of a suitable bracket. The housing 71 is positioned so as to be in the direct path of the air-flow from the cooling fan mounted to the radiator. The housing 71 is designed to transmit any changes in the air temperature to the bimetallic controller housed within the housing 71. The periphery of the housing is typically provided with a plurality of fins 73 to increase the surface area to optimise the controller's response to air temperature change.

[0047] The housing 71 further comprises at least two internal chambers (not shown), one accommodating the input shaft and one accommodating the output shaft 72. Fluid communication between the two chambers is via a valve (not shown) which is operated by the controller and a return orifice.

[0048] The input shaft has a first input or driven end and distal from the input end a plurality of vanes. The output shaft 72 has a driven end surrounded by a plurality of vanes. Distal from the driven end there is a drive end which is connected to the cooling fan. The vanes of the input and output shafts are surrounded by a viscous fluid contained within the housing. Fluid communication between the first and second chamber is permitted only via the control valve and the return orifice.

[0049] The input shaft is driven by the engine, typically via a belt and pulley. There is no communication of drive from the input shaft to the output shaft if the engine temperature is too low as the bi-metallic controller holds the valve closed.

[0050] Once the ambient air temperature exceeds a given threshold the bi-metallic controller opens the valve, allowing a circuit of fluid communication between the two chambers. A viscous fluid in the first chamber may then pass from the first chamber to the second via the valve, applying a force to the drive vanes on the output shaft and causing the fan to turn. Oil returns to the first chamber via the return orifice thus creating a fluid circuit.

[0051] A reliable seal must be provided between the housing and the input and output shafts to prevent a loss of fluid which would be detrimental to performance.

[0052] Figures 8a and 8b show, respectively, the exterior and interior of a water pump 80 for an internal combustion engine cooling system. The water pump 80 is used to pump coolant around the engine and between the engine and the heat exchanger or radiator. The water pump 80 comprises a housing 81, a drive shaft 82 and an impeller 83. The impeller 83 is connected to the drive shaft 82 which is driven by the engine. The drive shaft 82 rotates on bearings and must have a reliable fluid seal between the drive shaft 82 and the housing 81 to prevent loss of coolant or the fluid communication of coolant on one side of the housing 81 and engine lubricant on the other side of the housing 81 which would be detrimental to the engine.

[0053] Figure 9 shows an oil seal retainer 90 designed to be affixed to an end of the crankshaft 91 of a vehicle. An engine crankshaft 91 is provided with a primary output end (not shown) and distal from the primary output end is a secondary output end 92 or ancillary drive end. The output from the engine is taken from the primary output end of the crankshaft 91 which drives the vehicle via a gearbox.

[0054] Additionally, engine ancillaries such as power steering pumps, cooling fans, alternators, air conditioning compressors and vacuum pumps are driven by the crankshaft 91 via a pulley and serpentine belt system from the secondary output 92.

[0055] The crankshaft 91 is supported by bearings within a crankcase (not shown) which contains lubricating oil for the crankshaft 91 and the other moving parts of the engine. The pulley for the ancillary drive is mounted on the end 92 of the crankshaft 91 that is supported by bearings in the crankcase and protrudes through the crankcase to outside of the engine.

[0056] A reliable oil seat is vital at the point where the crankshaft 91 protrudes through the end bearings of the crankcase to support the pulley, to prevent the loss of lubricating oil from the crankcase.

[0057] Figure 10 shows an oil seal retainer 100 for sealing differential transmission systems, gearboxes and transfer cases. These devices supply drive torque from an engine to the road wheels of a motor vehicle.

[0058] Such devices typically comprise a housing 101, gears mounted on rotating shafts 102 supported by bearings set into the housing 101. The rotating parts require lubrication which is typically provided by a lubricant such as oil contained within the housing 101.

[0059] Rotating shafts 102 often need to protrude from the housing 101 in order to receive drive from the engine or another gear drive, or to output drive to the road wheels or another gear drive.

[0060] The housing 101 not only provides suitable support for the bearings that carry the rotating shafts 102, but also serves to contain lubricating oil and keep out foreign bodies such as dirt that could damage the gears.

[0061] It is vital to provide a reliable oil seal at the point where the rotating shafts 102 protrude from the housing 101 to prevent loss of lubricating oil or the ingress of dirt which would adversely affect the performance of the device.

[0062] Figure 11 shows a vacuum pump 110 for the braking system of a motor vehicle. The vacuum pump 110 is typically driven via a pulley driven by a serpentine belt on the side of the vehicle engine. The vacuum pump 110 typically comprises a housing 111, a drive shaft 112 and an impeller (not shown). The impeller is connected to the drive shaft 112 which is driven by the drive pulley on the outside of the housing 111. The drive shaft 112 rotates on bearings and must have a reliable fluid seal between the drive shaft 112 and the housing 111 to prevent loss of vacuum or lubricant. The loss of vacuum or lubricant would be detrimental to the performance of the braking system.

[0063] The rotating parts designed to provide liquid seals in each of the systems illustrated in Figures 7 to 11 respectively can be made using a two-step process of creating the part by sintering and then steam treating the sintered part in order to deposit a layer of Fe_3O_4 that creates a surface that is sufficiently smooth, non-porous and durable to provide the liquid seal without requiring machining or any additional surfaces to create the seal. A third, machining step is required only to create any internal shapes such as screw-threads or keyways that cannot be created using sintering, which step can be carried out before or after the step of steam treating the part.

[0064] Although the embodiments described herein relate to rotating parts, the skilled man will appreciate that parts designed to execute, for example, reciprocating or linear motion also require the smooth surface finishes provided by the method of the present invention. In addition to the disclosed embodiments, therefore the skilled person will recognise numerous other parts or components to which the present invention may be applied. The present application is intended to cover all such additional, non described embodiments and applications with express reference to applications within the field of engines and transmissions for vehicles such as automobiles, aircraft and watercraft. Furthermore, the present invention is equally applicable to non-moving parts wherein a fluid sealing surface is required between relatively moving parts.

[0065] The present invention provides a surface quality on the steam treated sintered part that greatly enhances the service life of the sealing member, this is especially the case for polymeric sealing members. The present invention provides a durable sealing surface as shown in Figure 6. The improvement in durability of both the fluid seal surface of the sintered part and the sealing member greatly enhances reliability and reduces service cost and complexity.

Claims

1. A fluid seal arrangement (17) comprising:

a sealing member formed from a plastics or polymeric material and a part in sealing cooperation therewith, wherein the part is **characterised by**:

a body (10) formed from a sintered ferrous material having at least one surface (5) arranged to provide a fluid seal (17), in use, **characterised in that** the at least one surface (5)comprises an un-ground surface of the ferrous material coated with a layer of Fe_3O_4;
wherein the part comprises one of:

a coupling (10) for a fuel injector (15);
a viscous fan (70) for a cooling system of an internal combustion engine;
a water pump (80);
an oil seal retainer (90) for use in sealing the ends of a crankshaft (91), differential transmission, gearbox, axle, driveshaft (82, 92, 102), hub (13) or transfer case; and
a vacuum pump (110) for use in the braking system of a vehicle.

2. A fluid seal arrangement (17) according to claim 1, wherein the sealing member is formed from a plastics or polymeric material and the part is formed from a sintered ferrous material, the part being movable relative to the sealing member and having at least one fluid seal surface (5) in sealing cooperation therewith, wherein the at least one fluid seal surface (5) comprises an un-ground surface of the ferrous material coated with a layer of Fe_3O_4.

3. A fluid seal arrangement (17) as claimed in claim 1 or claim 2, wherein the part or product is rotatable, slidable or reciprocable relative to the sealing member.

4. An engine, a transmission or a vehicle having a fluid seal arrangement as claimed in any of claims 1 to or 3.

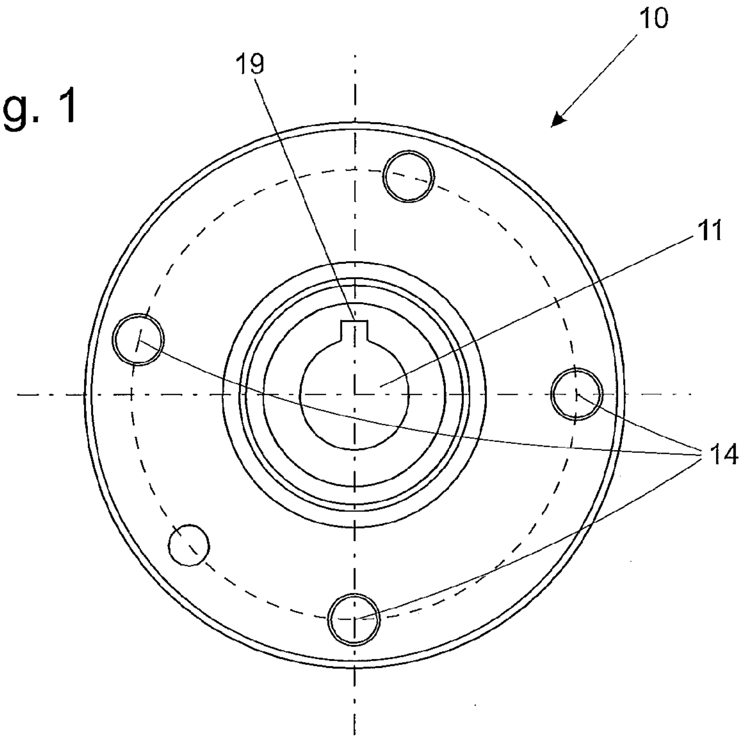


Fig. 1

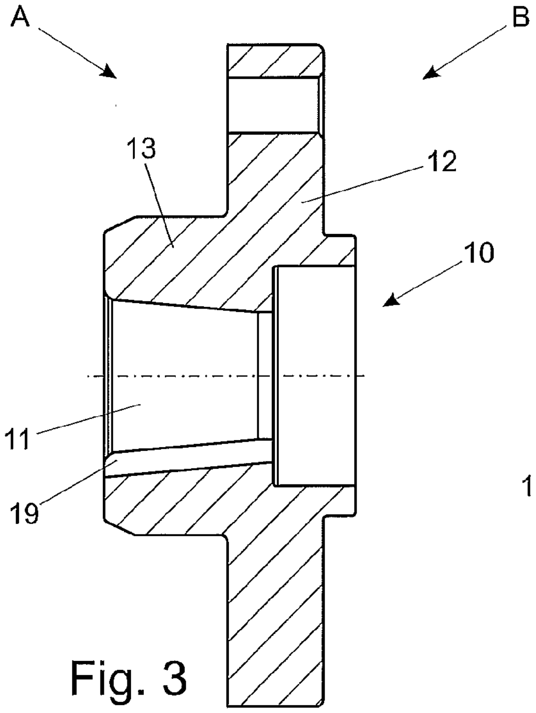


Fig. 3

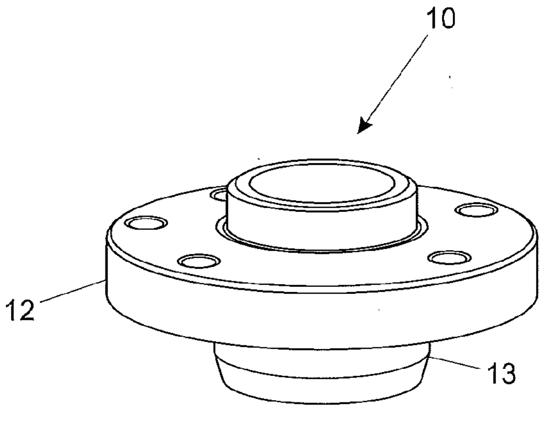


Fig. 2

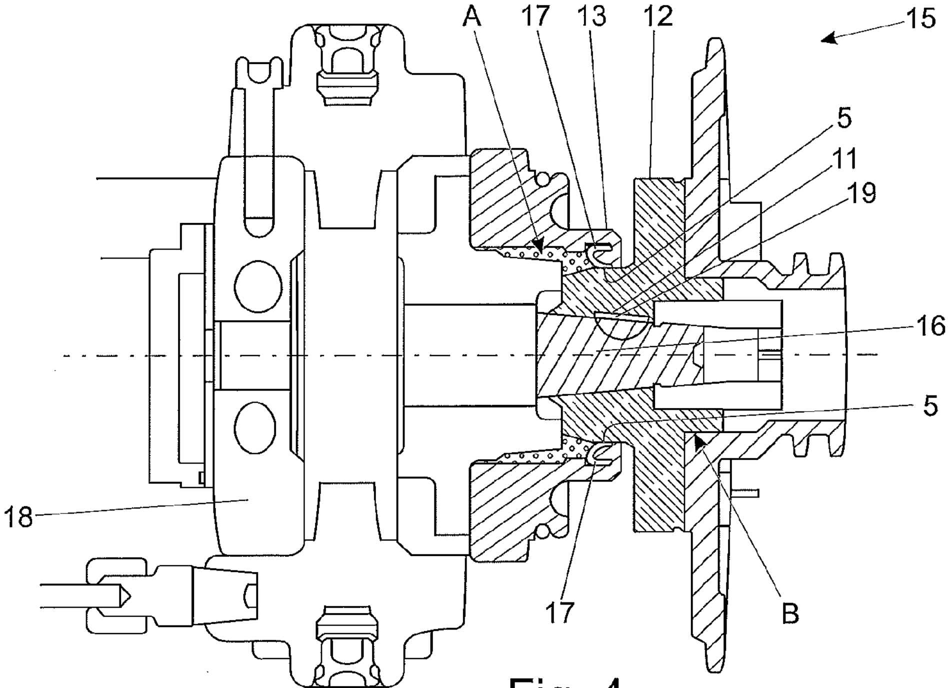


Fig. 4

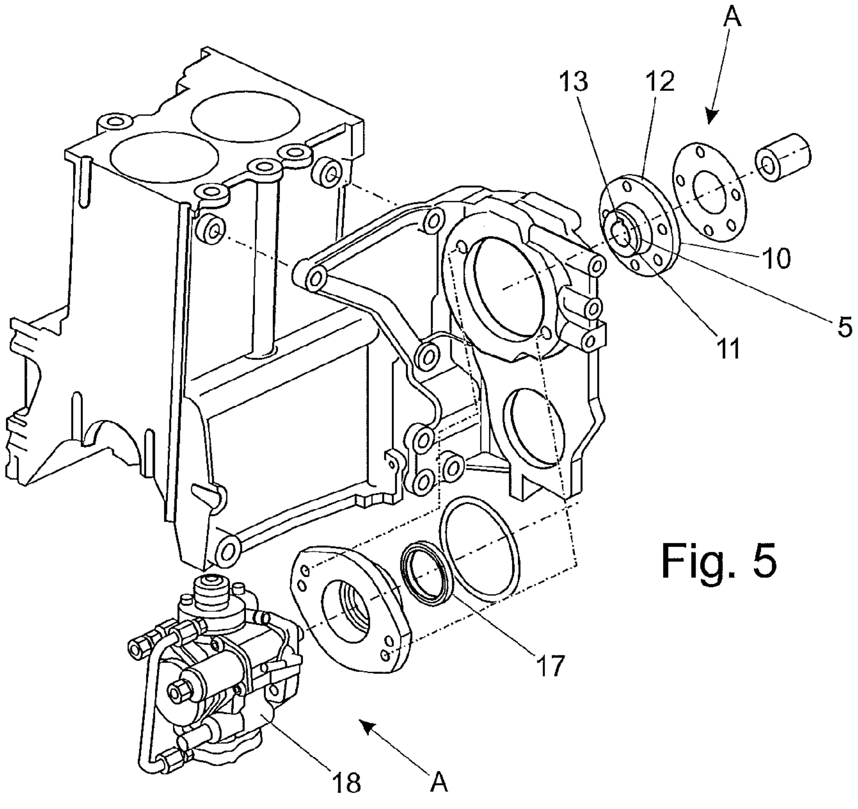


Fig. 5

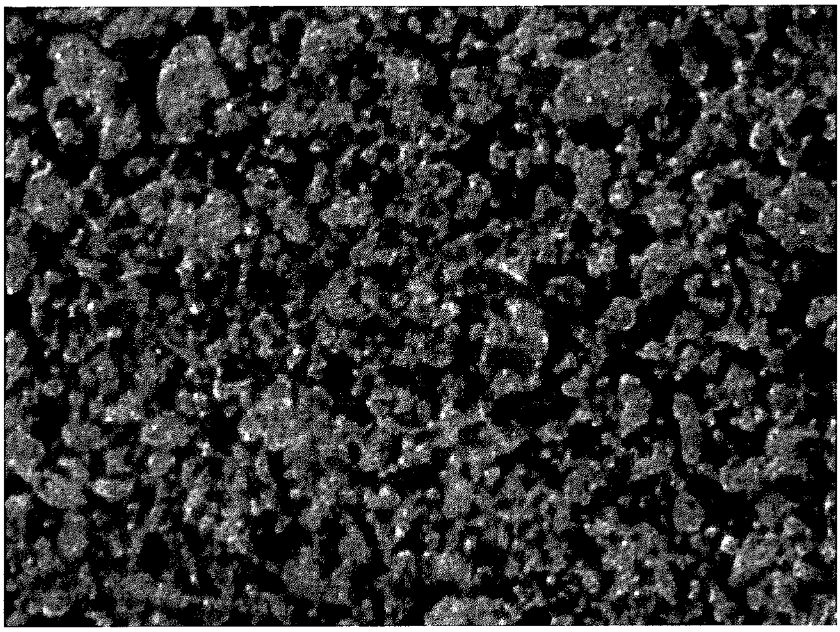


Figure 6a

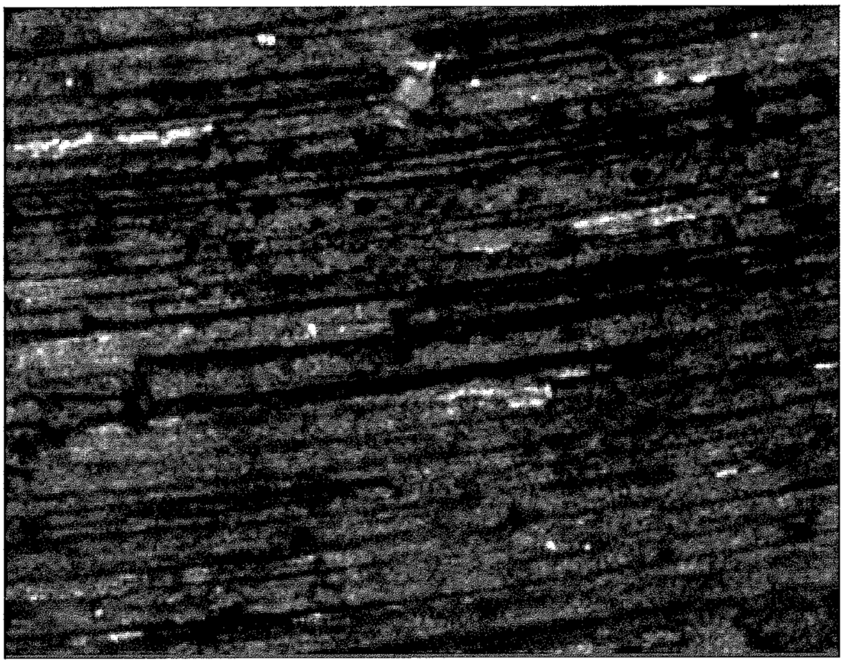


Figure 6b

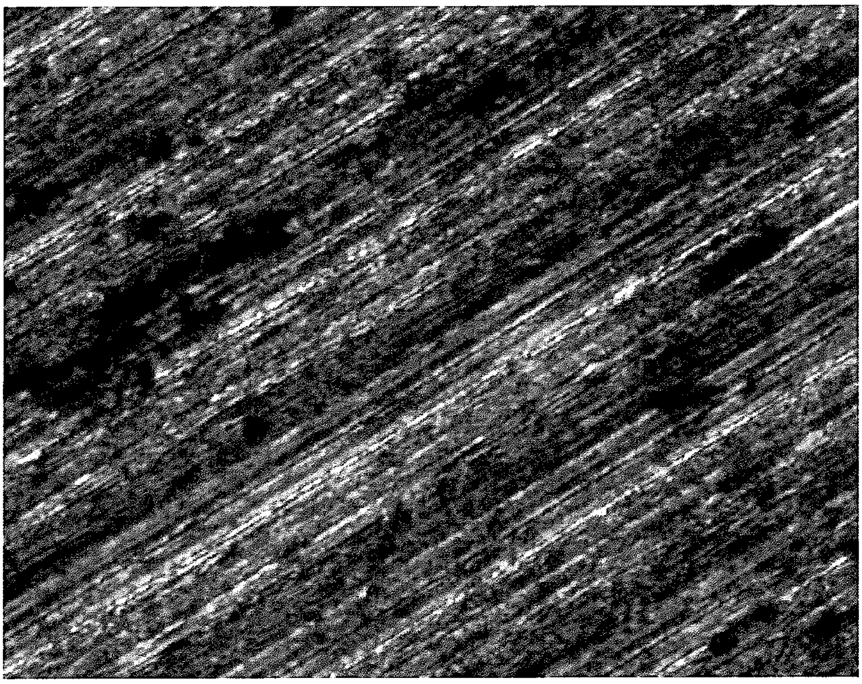


Figure 6c

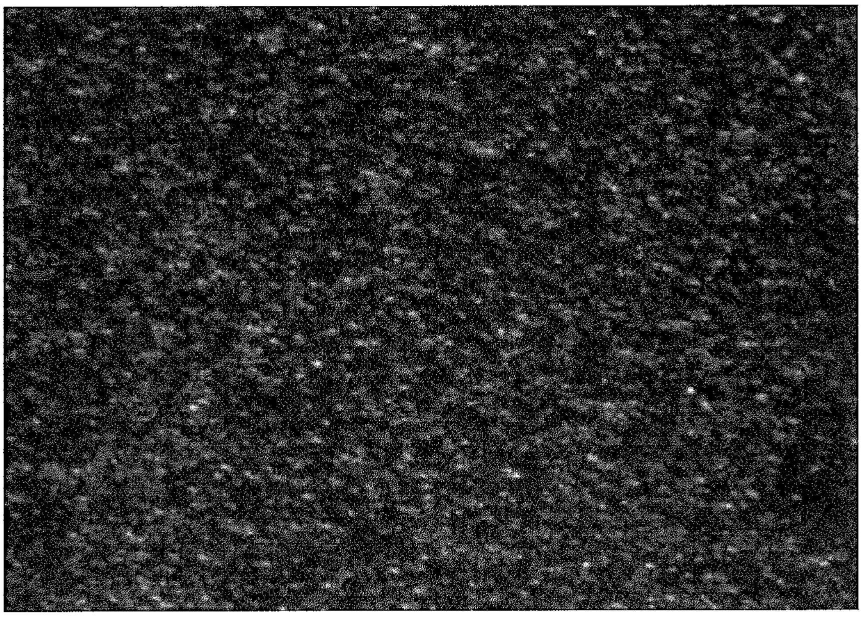


Figure 6d

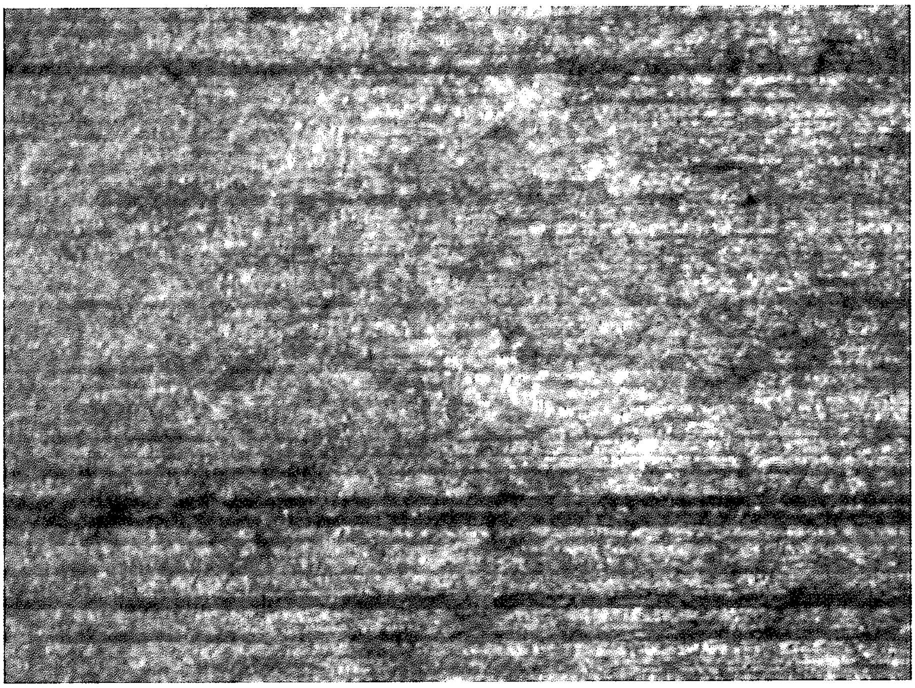


Figure 6e

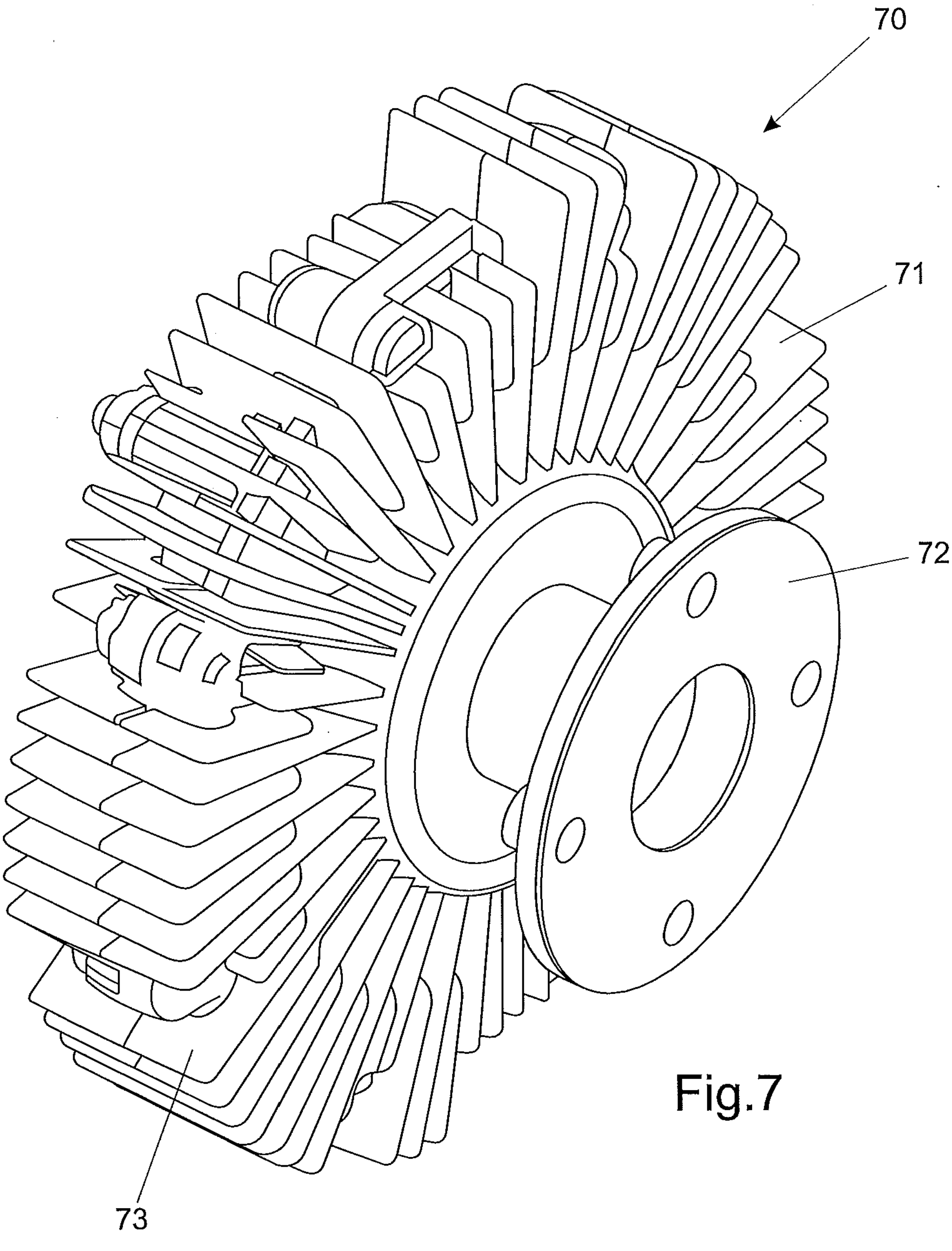


Fig.7

Fig.8a

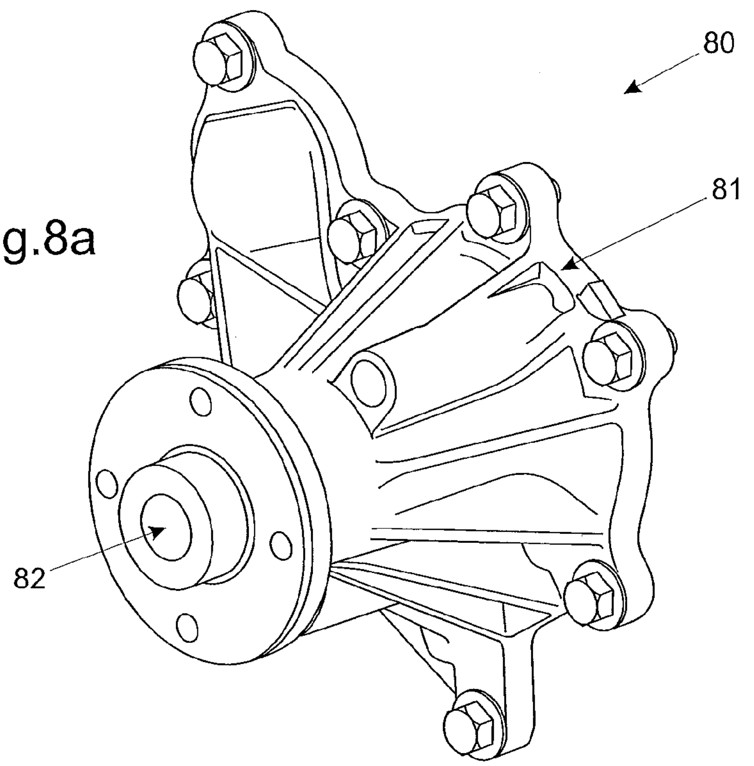
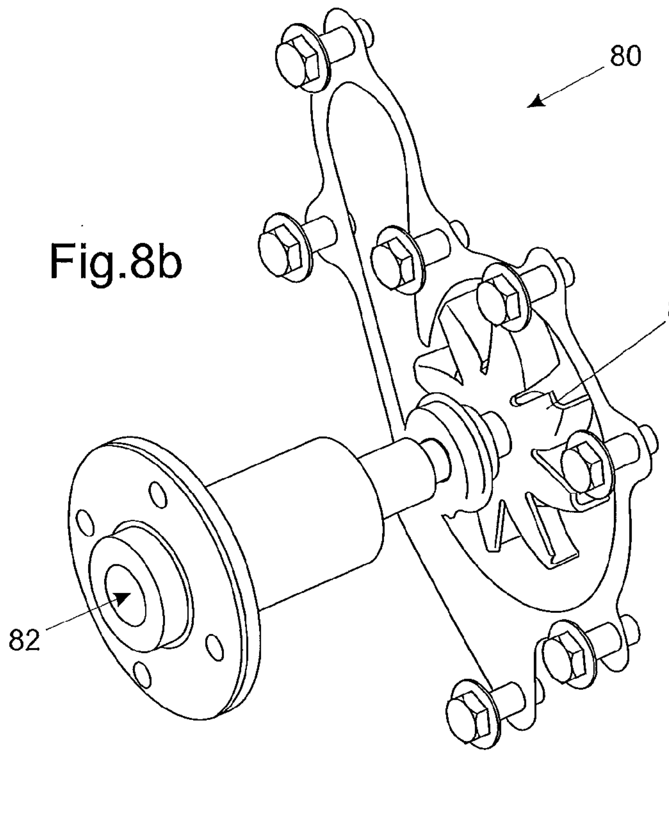


Fig.8b



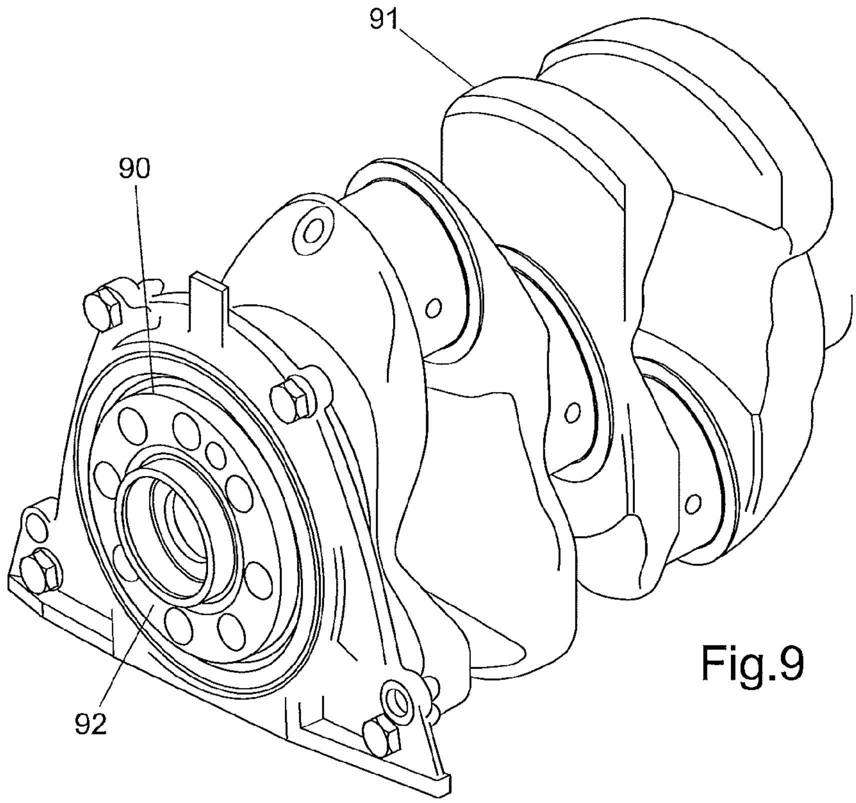


Fig.9

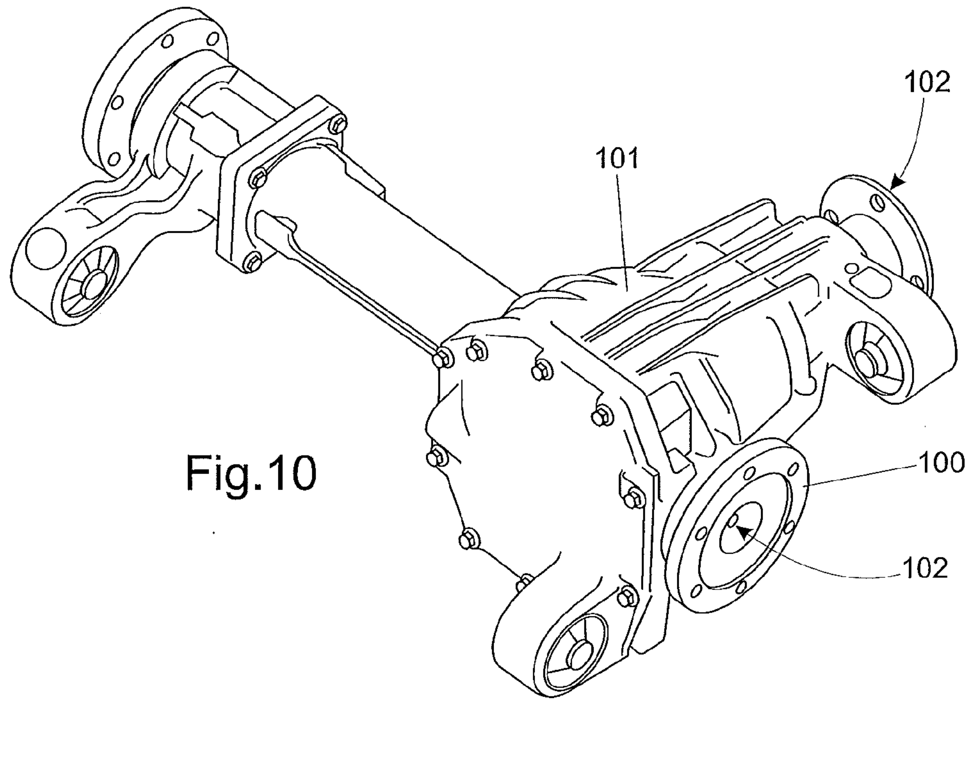


Fig.10

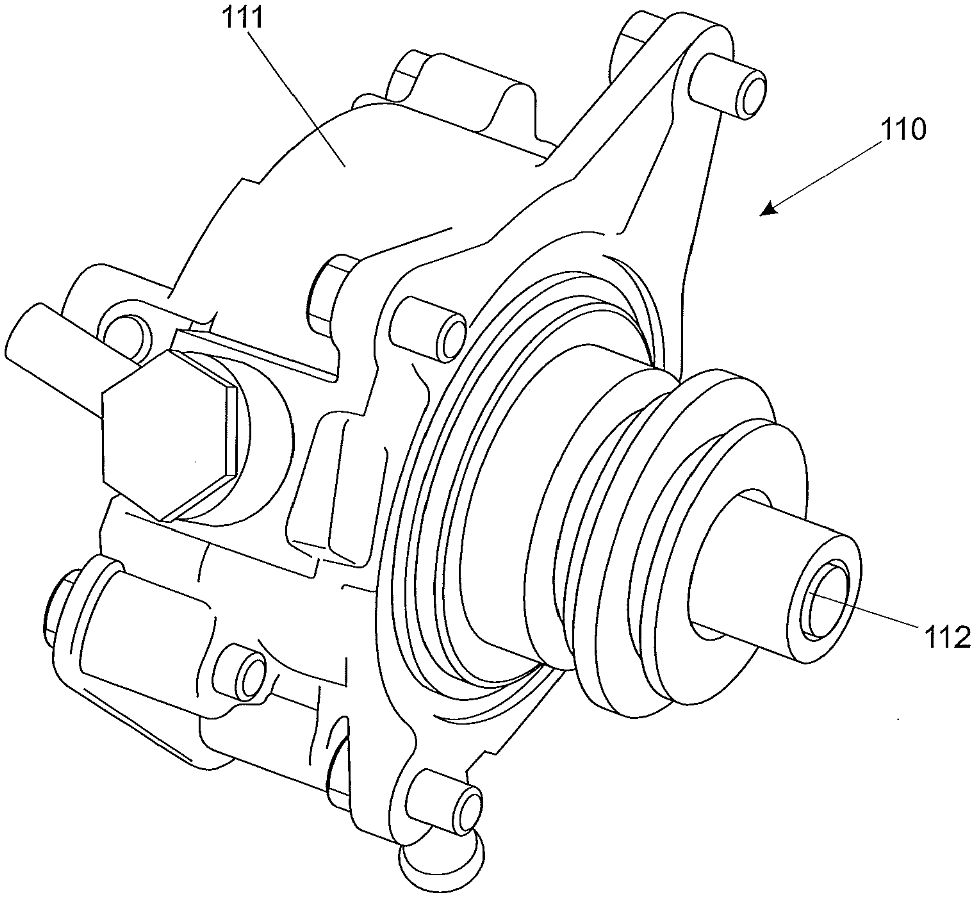


Fig.11