

PLASTICS VISCOSITY CONTROL METHOD AND APPARATUSCROSS-REFERENCE TO RELATED APPLICATION

5 This is a continuation-in-part of US Patent Application No. 09/333,544 filed June 15, 1999 and entitled METHOD AND APPARATUS TO CONTROL VISCOSITY OF MOLTEN PLASTICS PRIOR TO A MOLDING OPERATION, which is incorporated here by reference.

10 FIELD AND BACKGROUND OF THE INVENTION

 The present invention relates in general to molding with polymers, and in particular to a new and useful apparatus and method for controlling the viscosity of various polymeric materials by shear thinning and/or
15 disentanglement, as well as special measures for controlling the viscosity of polymeric materials that are capable of crystallizing upon cooling at a temperature which prevents disentanglement to take place in the optimum conditions. This is the case for semi-crystalline polymers
20 such as Nylon 66, for example.

 It is well known for molding polymeric materials that the processing parameters (e.g. temperatures, pressures, flow rates, flow length etc.) are a direct consequence of the viscosity of the melt which is governed by the state
25 of entanglements of the macromolecules. The resin "melt index" usually characterizes the fluidity of the melt and can be used to specify a certain resin grade suitable for a given molding application. The melt index is a function of the molecular weight of the macromolecular chains and
30 their degree of entanglement. A high melt index

corresponds to a high flow resin. The mechanical performance of a molded product is also a strong function of its molecular weight characteristics, the longer the macromolecular chains the stronger and stiffer the final article. Unfortunately the high strength required of plastics during their usage often results in a lack of fluidity during the molding operation, with as a result, high operational molding costs and molding defects (weld lines, sink marks etc.). One accepted practice which is designed to compensate for this lack of control of melt fluidity includes decreasing the molecular weight of the macromolecules being molded. Although this decreases the viscosity, improving the flow length significantly, it often results in a reduction of the mechanical properties, in particular strength and stiffness. Moreover, this practice cannot be used in those instances wherein the final molded product has to be very small and/or thin, such as in thin-wall injection molding applications.

Resin suppliers have succeeded in providing the plastic industry with means to lower the viscosity to ease up processing or increase melt elasticity by blending grades of different molecular weights. The problem with that solution is that the mechanical performance of the lower molecular weight polymers is also severely reduced, a compromise for better processability which processors have to pay.

The industry would welcome a process which allows the decrease of viscosity of plastic melts without the need to change the molecular weight of the resins, with the added advantage of a reduction in the number of grades a resin manufacturer has to offer.

Shear thinning of plastic materials is well known and is used practically to lower the viscosity of melts during

the filling stage of injection molding by increasing the speed of the injecting piston. This is particularly useful in the case of thin wall injection molding where considerable forces are required to fill the mold when the viscosity of the melt remains quasi-Newtonian.

It is also well known that shear thinning can be obtained, at a given temperature, by either increasing the shear rate or the frequency of oscillation of the melt at constant amplitude of oscillation. In particular, it is well known that the viscosity of a plastic melt can be reduced by shear thinning induced by vibration. See US Patent 4,793,954 to Lee; J.P. Ibar, "Melt Viscosity Reduction of Plastics by Vibration during Filling in Injection Molding", ANTEC 1997, Toronto, SPE Reprints (1997); and J.P. Ibar, "Smart Processing of Plastics Through Vibration Controlled Shear Thinning and Orientation", 1997 ASME International Mechanical Engineering Congress and Exposition. Reprint MD-Vol. 79, pp 223-348, 1997.

Shear thinning does not invoke the same mechanism of viscosity reduction than what is required to produce long term viscosity reduction by disentanglement. See, for example, US Patent 5,885,495 to Ibar. Shear thinning results from the elastic cooperativity of the network of interactions between parts of the macromolecules called conformers. The viscosity reduction is instantaneous and only prevails under vibration, i.e. it ceases if the vibration ceases. Shear thinning can be useful, however, and can be optimized or favored in a plastic according to the present invention, in ways that are not known or obvious to the person of ordinary skill in this field.

Viscosity reduction induced by vibration-shear thinning is known. An example is described in J.P. Ibar,

"Smart Processing of Plastics Through Vibration Controlled Shear Thinning and Orientation", 1997 ASME International Mechanical Engineering Congress and Exposition.

5 US Patent 5,885,495 teaches there are three categories of known processes that use vibration to modify the molding process and/or the properties of molded materials:

1. Mechanical shaking/oscillation or ultrasonic vibration devices to homogenize and increase the density of the material molded, either in the liquid stage or in
10 the solidifying stage, either at a macroscopic or microscopic level See US Patent 4,288,398 to Lemelson; US Patent 3,298,065 to Pendleton; and US Patent 4,925,161 to Allen et al. These patents do not directly concern the use of vibration to lower the viscosity of a melt to
15 increase their processability during conversion, nor do they address the use of packing vibration to increase the melt elasticity.

2. Processes based on the fact that material rheology is a function of vibration frequency and amplitude in
20 addition to temperature and pressure: See US Patent 4,469,649; EP Patent 0 273 830; US Patent 5,306,129; US Patent 4,919,870; CA Patent 1,313,840; and EP Patent 0 274 317; all to Ibar. The teachings in these patents, however, do not alter the viscosity of the melts in a way which
25 preserves the viscosity reduction nor do they optimize shear thinning effects by the introduction of ribs on the surface to create oscillating extension flow.

3. Processes using vibration to generate heat locally by internal friction or to decrease surface stresses at the
30 wall interface between the melt and the barrel or the die to increase throughputs: See Casulli et al. "The Oscillating Die: A Useful Concept in Polymer Extrusion" Polym. Eng. Sci.; 30 (23), 1551 (1990) and Wong et al.

"Flow of Thermoplastics in an Annular Die under Parallel Oscillations", Polym. Eng. Sci.; 30 (24), 1574 (1990), for example. These processes do not try to modify the viscosity of the melt per se, in a way which would be similar to a reduction of the molecular weight average of the macromolecules.

The polymer molding industry would greatly welcome an apparatus and/or method which decreases the viscosity of macromolecules without changing their mechanical performance. Such a process is described in US Patent 5,885,495 to Ibar where the macromolecules are pulled apart to produce the disentanglement effect which is responsible for the lowering of viscosity.

In general, in order to use frequencies for the oscillating extensional flow which remain economically feasible to practice industrially, it is necessary to lower the temperature of the melt to a temperature zone well within the rubbery flow region of the polymer.

For amorphous polymers, the lowest possible temperature limit to enable flow to take place is the glass transition temperature of the polymer, T_g . Below that temperature, the polymer is no longer a melt and becomes a solid glass which does not flow. For such polymers, it is generally possible to find a disentanglement processing temperature well above T_g at which the polymer can still flow and be treated mechanically at constant temperature according to US Patent 5,885,495.

Semi-crystalline polymers, such as Nylon 66, are amorphous melts above their temperature of crystallization, T_c , and solids presenting a crystallographic structure below T_c . In general, the glass transition temperature of the remaining amorphous phase present, which co-exists with the crystalline phase, is located much below T_c , hence

in the solid state of the polymer. For such semi-crystalline polymers, the disentanglement processing window is restricted since it can only be located above T_c .

For some semi-crystalline polymers, such as ENGAGE 8180, a trademark for a metallocene Polyethylene polymer sold by Dupont-Dow Elastomers LLP, the amount of entanglement is so high, and the resulting elasticity of the melt is so sensitive to the effect of a shear oscillation, that manageable low mechanical frequencies (below 50 Hz) are able to bring the melt in the optimized elastic region favorable to extensive shear-thinning capable of producing disentanglement [2], even at temperatures well above the crystallization temperature. For instance, the crystallization temperature T_c for ENGAGE 8180 is 60°C, and the most favorable temperature range which optimizes shear-thinning capable of producing disentanglement is between 110°C and 160°C. As a consequence, there is no interference or limitation from the phenomenon of crystallization in obtaining high elastic state for a flowable, extrudable ENGAGE 8180. This is not the case for other semi-crystalline polymers, such as Nylon 66, for which desirable high elastic states cannot be obtained, under the effect of combined shear oscillation (of manageable frequency) and temperature, at temperatures above the crystallization temperature T_c of the polymer. For such semi-crystalline polymers, temperature cannot be lowered enough before crystallization temperature is reached, which interferes with the disentanglement process.

The present invention is a new apparatus and method which applies the disentanglement process taught in US Patent 5,885,495 for use on an industrial scale and which also identifies additional measures involving temperature control and shear thinning techniques that are novel and

unobvious over the prior art.

SUMMARY OF THE INVENTION

The present invention overcomes the prior art problems and shortcomings in a method and apparatus which produces
5 a novel product wherein extensional shear vibration is exercised on a molten plastic as it is continuously extruded to pass through treatment stations at determined temperature, frequency and amplitude of vibration, during a certain time and under specific vibrating conditions for
10 each station, to obtain a controlled degree of shear-thinning (elastic state) susceptible to result in a progressive decrease of the entanglements between the macromolecules, in order to significantly and controllably decrease the viscosity of the extruded melt in a manner
15 which can be beneficial to future molding operations requiring a lower melt viscosity.

A method and apparatus are disclosed for decreasing in a continuous way the viscosity of molten polymers, such as a metallocene polyethylene or polycarbonate, prior to
20 or during a molding operation such as injection molding, extrusion, thermoforming, blow molding or compounding. When a significant reduction of viscosity is desirable, the plastic melt is submitted to the action of a specific mechanical extensional shear vibration, with minimum or no
25 external pressure, within a specific range of amplitude and frequency of melt oscillation to cause the melt to become highly elastic, and simultaneously to be fatigued in extensional flow conditions for a certain time, maintaining this high elastic state which corresponds to a high degree
30 of shear-thinning, until the macromolecules have partially

or totally disentangled, in a controllable manner, at which stage the melt is ready for a molding operation such as a simple quenching operation or an extrusion process followed by quenching to produce pellets or compounds with a better mix or a lower viscosity when remelted, or an injection molding or similar molding operation where the melt viscosity has been greatly reduced allowing a better processability of the injected part, for instance allowing the use of a lower temperature of injection, a lower pressure of injection or both, or even in a value gate or extruder where surfaces of the rod, stem, rotor or screw of these units can be equipped with the rib means of the present invention and move according to the present invention to achieve the shear-thinning and viscosity reduction effect.

Accordingly, one object of this invention is to provide a molding apparatus and/or method which can reduce the viscosity of melt in a given controllable degree by shear-thinning/disentanglement of the macromolecules while the resin is in the melt.

Another object of this invention is to provide a molding apparatus and/or method which can continuously produce reduced viscosity resins which can be either stored and bagged as special low melt viscosity producing pellets or pumped to another molding site for immediate use.

These and other objects are achieved through the advent of a novel apparatus and/or a method of using the same. The novel apparatus includes, among other things, at least one station which defines a cavity, the treatment cavity, wherein a molten moldable material can pass into and/or flow through it to be treated to produce at least partially disentangled melts or even melts that are only subjected to shear thinning, e. g. according to the process

described in US Patent 5,885,495.

The treatment station of the apparatus includes means for exerting a shear drag at given strain rate of the molten moldable material as it flows through from the inlet
5 to the outlet of the treatment station.

The treatment station includes rib means for exerting a variation of the shear strain rate of the molten moldable material as it flows through from the inlet to the outlet of the treatment station such that it produces extensional
10 acceleration/deceleration of the flow.

The apparatus includes means to move the melt, e.g. to continuously push and/or drag and/or pump the melt located in the station treatment cavity from the inlet conduit towards its outlet conduit.

15 Also included in this apparatus are means to vary the gap which the molten, moldable material can pass and/or flow through.

Also included in the apparatus are means to continuously vent the treatment cavity to prevent the formation of bubbles or cavitation during the treatment.
20

The apparatus also includes various known monitoring and control devices for temperature, pressure, torque exerted on the molten moldable material contained within the treatment cavity(s).

25 Another object of the present invention to provide a novel method of processing semi-crystallizable polymers in order to disentangle their melt without the interference of crystallization by varying the temperature during the shear thinning or disentanglement treatment in a
30 controllable way which pushes down to a lower temperature the beginning of crystallization and, therefore, allows the use of lower temperatures for increasing the elasticity of the melt by the shear oscillation methods.

Other objects, aspects and advantages of the present invention will be apparent to those skilled in the art upon reading the specification and the appended claims which follow.

5 The various features of novelty which characterize the invention, are pointed out with particularity in the claims annexed to, and forming a part of this disclosure. For a better understanding of the invention, its operating advantages and specific objects attained by its uses,
10 reference is made to the accompanying drawings and descriptive matter, in which preferred embodiments of the invention are illustrated.

BRIEF DESCRIPTION OF THE DRAWINGS

15 FIGS. 1A, 1B, 1C, 1D, 1E and 1F are schematic illustrations of embodiments of a shear thinning and/or disentanglement apparatus designed in accordance with the present invention having a single feeder and a single treatment cavity, wherein the shear vibration under
20 extensional flow is produced by rotating at constant speed and/or at modulated speed and/or by pure oscillation an incoming melt which flows outwards over ribs disposed on at least one surface.

25 FIG. 2A is a schematic illustration of one embodiment of a disentanglement apparatus designed in accordance with the present invention having a single feeder and a single treatment cavity, wherein the shear vibration under
30 extensional flow is produced by feeding a molten moldable material through a gap composed of at least one rotating and/or oscillating surface presenting on its contour a profile over which the molten material flows and/or can be

dragged and/or is being pushed through and/or pumped through.

FIG. 2B is a sectional view of FIG. 2A taken across the axis of FIG. 2A.

5 FIG. 3 is a schematic illustration of an embodiment of a shear-thinning and/or disentanglement apparatus designed in accordance with the present invention having a single feeder and more than one treatment cavity, separated by a pumping section, wherein the shear vibration
10 under extensional flow in the treatment cavity section is produced by feeding a molten moldable material through a gap composed of at least one rotating and/or oscillating surface presenting on its contour a profile over which the molten material flows and/or can be dragged and/or is being
15 pushed through and/or pumped through. FIG. 3 also illustrates the use of the invention in an extruder.

FIG. 4 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein
20 the shear vibration under extensional flow is created in the gap of concentric conic surfaces in relative motion with respect to one another, at least one of which presenting an array of ribs and bumps and rotating at constant speed and/or at modulated speed and/or in a pure
25 oscillation mode.

FIG. 5 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention.

FIG. 6 is a schematic illustration of one embodiment
30 of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention.

FIG. 7 is a schematic illustration of one embodiment of the disentanglement apparatus designed in accordance

with the present invention wherein the feeder is an extruder and/or a plunger, the disentanglement treatment occurs in a series of stations connected through gear or screw pumps, and the accumulator of the last station is
5 connected to a pelletizer line and/or the feed of an extruder or injection molding equipment or injection molding cavity.

FIG. 8 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus
10 designed in accordance with the present invention wherein the shear vibration under extensional flow is created in the gap between cylindrical or conical rollers in close contact to one another and creating a rolling and/or vibrating wall, and a core surface at the center, at least
15 one surface rotating at constant speed and/or at modulated speed and/or in a pure oscillation mode.

FIG. 9 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein
20 the shear vibration under extensional flow is created by the relative motion in the molten moldable material of a train of submarine-shaped wagons circulating, at constant speed or at a modulated speed, in a torus ring in communication with an inlet and an outlet conduits.

FIG. 10 is a schematic illustration of one embodiment of the disentanglement apparatus designed in accordance with the present invention wherein the feeder is an extruder and/or a plunger, the disentanglement treatment occurs in a series of superposed and interconnected ring
25 stations defined by a tubular track through which trains of submarine-shaped wagons circulate at controlled speed, and the accumulator of the last ring station is connected to a pelletizer line and/or the feed of an extruder or
30

injection molding equipment.

FIG. 11 is a schematic illustration of one embodiment of the power transmission of the shear-thinning/disentanglement apparatus designed in accordance with the present invention wherein the shear flow obtained by pure rotation and the controlled extensional fatigue of the molten moldable material in the treatment cavity created by the oscillation are independently applied on both axes of a differential which combines both rotation and oscillation (also called an epicyclic drive).

FIG. 12A is a schematic end view of one embodiment of the shear vibration transmission of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration in the molten moldable material in the treatment cavity is created by the oscillation of the inner cone of a concentric conical assembly attached to one axis of an epicyclic differential, the rotary oscillation being created through a cam and a piston.

FIG. 12B is a side view of FIG. 12A.

FIG. 13 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow in the molten moldable material is created by the relative motion of two concentric conic surfaces at constant speed or at a modulated speed, or at a combined constant and modulated speed, while the molten moldable material is pushed and/or dragged and/or pumped from the inlet conduit to the outlet conduit of the treatment cavity.

FIG. 14A is a schematic illustration of one embodiment of the disentanglement apparatus designed in accordance with the present invention wherein one or several feeders feed molten moldable material to a series of tubular

cavities, intermeshing or not, which themselves connect, directly or through pumping sections, to other series of tubular cavities, so on and so forth until the last series of treatment stations which connects to a collector and a
5 pellitizer line and/or the feed of an extruder or injection molding equipment.

FIG. 14B is a view similar to FIG. 14A, but showing a different mode of operation.

FIGS. 15A, 15B and 15C are schematic illustrations of
10 one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the rib means are ribs/bumps on at least one of the surfaces in contact with the molten moldable material have a specific shape to create a given
15 profile of strain rate and strain rate variation under extension, and are aligned in a specific manner with respect to the axis of surface rotation in order to create a certain amount of drag flow. FIGS. 15A to 15C define some of the shape parameters and the angle of tilt. The
20 contours of the ribs are calculated to eliminate any possibility of turbulence or other non linear defect in the flow, which should remain laminar. The height of the rib, e , the width of the ribs, w , the radius $R1$, $R2$ and $R3$ which define the curvature to accelerate ($R1$) or decelerate ($R3$)
25 the flow, can be different along cross sections aa and bb (the cross section bb is not shown here). The strain rate vector decomposes into two components, which defines two directions of drag flow. The shape of the ribs along each axis aa or bb is determined by the strain rate profile (acceleration followed by deceleration) desired in these
30 directions. Both profiles can be used to define conditions of shear vibration and/or extensional fatigue which can couple up to optimize elasticity of the melt (shear-

thinning) resulting in disentanglement effects.

FIG. 16 is a graph plotting specific volume (in cc/g) of Zytel 101, a Nylon 66 from the Dupont de Nemours Company (Geneva, Switzerland), versus Temperature ($^{\circ}\text{C}$), at two
5 different cooling rates, one corresponding to a slow cooling rate, the other one corresponding to a quenching operation typical of the cooling speed obtained in injection molding, for instance. These curves correspond to a 2mm thick slab of Nylon 66 cooled from 300°C to room
10 temperature.

FIG. 17 is a graph plotting the relative elasticity in shear mode, G'/G^* , while cooling at $10^{\circ}\text{C}/\text{min}$ for a Nylon 66. G' is the elastic modulus and G^* the complex modulus for a frequency of 10 rad/s.

15 FIG. 18 is a sectional view of an embodiment of the invention that has two concentric treatment chambers.

FIG. 19 is a partial side elevational view of a rotor, rod or stem with honeycomb rib means according to the invention.

20 FIG. 20 is a view similar to FIG. 19 of an embodiment of the invention with rail means uses as the rib means of the invention.

FIG. 21 is an enlarged perspective view of the rail means.

25 FIG. 22 is an axial sectional view, partly in elevation, of a valve gate made according to the present invention.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention pertains to apparatuses for
30 reducing the viscosity of moldable materials (e.g.,

polymers) through disentanglement processes. It also pertains to methods of using such apparatuses.

The apparatus encompassed by the present invention includes at least one slit cavity, the treatment cavity, where molten moldable material flows through under conditions where shear vibration under extensional flow takes place to enable disentanglement to occur. The apparatus is not limited to any specific cavity design nor configuration, provided laminar flow without turbulence or cavitation prevails. For instance, the cavity can have a constant gap or a variable gap. The cavity can have sections of constant gap followed by sections of variable gaps. The gap cross section dimensions at any given point of the flow path determine the strain and strain rate of deformation on the molten moldable material at that point. Examples of calculation of strain and strain rate as a function of gap geometry can be found in J.D. Ferry, "Viscoelastic Properties of Polymers", Appendix C, page 640, Second Edition, John Wiley & Sons, NY. Library of Congress Catalog Card#: 76-93301. For an annulus (circular slit), the gap cross-section is determined by the radius and height of the gap. Pressure drop, shear stress at the wall and flow rate are a strong function of melt viscosity which varies with strain rate, temperature and gap profile. In summary, the gap profile is crucial not only to determine the extent of flow, i.e. the throughput, but also to implement a certain type of flow. For instance, a gap convergence creates an acceleration of flow, and its extension. Conversely, a gap divergence translates into a deceleration and a contraction of flow. Obviously, a succession of restriction and enlargement of the gap, in the pathway of a flowing molten moldable material, results in a periodic variation of the strain rate from

acceleration to deceleration, and of the strain from extension to contraction. If ribs are regularly disposed on at least one surface in contact with a melt flowing in the gap created by two adjacent surfaces, a periodic shear oscillation under extensional flow is created which, according to the present invention, can be set to produce a high elastic state (shear-thinning) favorable to induce disentanglement effects, using operating conditions of strain rate and strain taught in US Patent 5,885,495.

Upon reading this specification, those skilled in the art will know what types of slit geometry and configuration can be used when practicing the invention. For instance, the disentanglement treatment cavity can be an extrusion barrel/screw die or valve gate bushing/rod modified to incorporate the rib means features capable of bringing the melt in the processing window region required by US Patent 5,885,495 to produce disentanglement. This can be done by re-designing the screw profile and the gap between the barrel and the screw and by adding specifically designed ribs and/or grooves on both the inside surface of the barrel and on the screw. The modified screw rotates inside the close fitting barrel, shearing the molten moldable material over the profiled gap. A multiple screw treatment cavity can be built wherein the screw flights intermesh.

When it is the case, the action of the intermeshed screw is, in addition to producing a vibrational flow under extension, to move forward the molten moldable material due to positive displacement, in a way somewhat similar to that of a gear pump. Single screw cavity and nonintermeshing multiple screw disentanglement cavities lack this positive displacement, but the ribs and grooves can be spaced and positioned with respect to the rotation motion such that a longitudinal strain rate is added to that caused by

pressure flow. Upon reading this specification, those skilled in the art will know what types of ribs and grooves can be added to an extrusion barrel and screw dies in order to practice the invention and create a pumping effect at the same time.

Use of the invention in a valve gate is discussed later in this disclosure, in connection with FIG. 22.

In general, the preferred treatment cavity design of the present invention will depend, in part, upon the nature of the resin to be elasticized or disentangled, the throughput desired and the resources available to the practitioner of the invention. For instance, if enough resources are available, an apparatus comprising multi disentangling stations is desirable wherein each individual station is set to optimize throughput, not disentangling efficiency, but viscosity reduction is compounded at each station to yield a final disentangled melt, produced at fast rate, with the proper final viscosity reduction ratio. For instance, a 5 station disentangling apparatus, working at a low efficiency rate of 27.5% per station ($\mu_{out}/\mu_{in} = 0.725$), produces a melt with final viscosity reduction of 5, which is $(0.725)^{-5}$. The throughput of such a multi station disentangling apparatus is many times greater than the throughput of a single station working to produce the same result in terms of viscosity reduction. The operating cost per weight produced is also greatly reduced.

The inlet through which the molten moldable material passes through the treatment cavity is connected to at least one feeder for preparing a molten, moldable material. The feeder is spaced from the treatment cavity and any other feeder which may be present. The feeder includes a means for expelling the molten, moldable material through

its outlet. This expelling action can be accomplished by any suitable means known to those skilled in the art. Some examples of feeders which include such an expelling means include, without limitation, screw pumps as used in
5 extrusion and in injection molding apparatuses, twin screws as used with blends, plungers as used in two-stage molding apparatuses, gear pumps, and the like. This expelling action can include means to pressure pulse the melt to facilitate flow, as described in References [12,13,14-
10 16,19,20], although such a conditioning is not required by the present invention.

It is also within the scope of this invention for the apparatus to include a plurality of feeders. If a plurality of feeders are employed, the molten, moldable material
15 prepared in each need not be the same. For example, each feeder can contain the same material. They can also contain the same material but at a different temperature, and/or different molecular weight characteristics, and/or different degree of disentanglement. Specifically, one
20 material in one feeder may be a highly disentangled fraction coming from a different treatment station (downstream or else), whereas another material may be less disentangled. This changes the concentration of disentangled fractions and allows to engineer specific
25 bimodal blends of grades having different states of disentanglement. The separate feeders can also contain materials which are virgin in one and recycled in another. Moreover, the individual feeders can contain completely different materials or materials which are filled or
30 unfilled with fibers, fillers (such as wood, flour or ESD powder to name a few), different concentration of blends of polymers and/or polymers/liquid crystal polymers, pigments, anti-oxidants, flame retardant compounds, and the

like, and/or any combination thereof. The second or third feed can be introduced into the disentanglement system at any inlet stage of the multi stations which chain up head to tail to continuously produce disentangled melts of given viscosity characteristics.

The apparatus can also include at least one accumulator for collecting the treated molten, moldable material before it is sent to either a pelletizer system or to a pumping station. In one embodiment of the present invention, the accumulator defines a cavity which continues to shear the collected disentangled melt and contains a series of immersed intermeshed rollers with ribs on their surface to preserve and maintain the state of disentanglement until the melt is pumped out of the accumulator by the controlled activation of a screw pump or a gear pump. One non-limiting example of such disentanglement retaining means is the device illustrated by FIG. 8. In another embodiment of the present invention, the disentangled melt is pumped to a pelletizer line which quickly freezes the disentangled melt into pellets which are dried and bagged, ready for shipment. Any type of pelletizer line can be used. Upon reading this specification, those skilled in the art will know what type of pelletizer line can be added to the accumulator to freeze the state of disentanglement obtained through the treatment in order to practice the invention.

The treatment station includes means for exerting a shear vibration coupled with extensional flow on the molten, moldable material contained within the treatment cavity. The vibration is either created through direct oscillating means or indirectly, through the rotation of at least one profiled surface (i.e. with rib means) in contact with the molten moldable material. In other words,

even pure continuous rotation in only one direction can locally oscillate the melt because of the presence of the rib means which periodically and locally accelerate and decelerate the melt as it moves past the ribs, bumps or grooves in the treatment gap. In one embodiment of the invention, a shaft supporting a surface in contact with the molten moldable material is oscillated by means known to a person in the art or any combination of them. A rotary vibration can be induced by hydraulic, pneumatic, electrical, electromagnetic means and the use of cams, connecting rods, and/or crankshafts. Any skilled person in the art will know how to create a rotary oscillation of frequency between 1 Hz and 100 Hz and of amplitude between 0.1 and 20 degrees, depending on the overall dimensions of the device. The periodic motion created does not require to be a pure sine wave and can be a mixture of several sine waves to create such periodic oscillation as square waves, triangular waves and the like.

In another embodiment of the present invention, the oscillation of the shaft connected to at least one surface in contact with the molten moldable material is coupled with a continuous rotation of the shaft, occurring at a certain rotation per minute. The coupling of the two motions can be done by programming the combined motion profile, which corresponds to a modulated rotation, and requesting the PID controller to follow the desired signal. But this solution may require extraneous efforts from the electric motors or hydraulic actuators and, in another embodiment of the present invention, the two motions are implemented from separately independent means, such as two electrical motors, and combined through an epicycle differential to recreate the modulated rotation on the shaft of the treatment cavity. In yet another embodiment

of the present invention, two oscillatory motions of different frequency and amplitude are combined through the epicyclic differential to optimize shear vibration and extensional fatigue of the molten moldable material in order to practice the invention.

Another object of the present invention is to enable a shear vibration under extensional flow to occur with a full control of the strain rate and of the acceleration and deceleration of the shear strain. US Patent 5,885,495 teaches how to set both the frequency and amplitude of a shear vibration at a given temperature to achieve a high elastic state of the melt producing extensive shear-thinning favorable for producing disentanglement of the macromolecules and obtain a reduction of melt viscosity. But strain, strain rate and strain acceleration are the derivative of one another and cannot be set independently. As explained earlier, the use of ribs and/or grooves and/or bumps (collectively and separately here called "rib means") to profile and modulate the gap dimension, in conjunction with the relative motion of the surfaces which carry those ribs/grooves, allows to further control the degree of extensional flow, melt fatigue and melt flow acceleration/deceleration. The number of ribs/grooves, the spacing between the ribs/grooves, the height of the ribs/grooves, the width of the ribs/grooves, the surface area of the ribs versus the surface area of the grooves, are all interrelated parameters of the present invention, which are set to create a high elastic state in the melt to produce a large viscosity reduction and disentanglement. The addition of rib means increases to 3 or 4 degrees of freedom (depending on whether one or two surfaces are structured with ribs/grooves) instead of 2 in US 5,855,495, the ability to apply to the melt a chosen strain rate,

strain amplitude and frequency of shear vibration under extensional fatigue. The advantage of the extra degree(s) of freedom can be used, in a specific embodiment of the present invention, to optimize viscosity-reduction and disentanglement effects, i.e. reduce the energy required to disentangle a certain quantity of entangled resin and/or increase the throughput by accelerating the kinetics. Upon reading this specification, those skilled in the art will know how to calculate the dimensions of the ribs, their number and the speed of rotation of the lateral dragging flow to produce the desired and controlled strain rate and acceleration/deceleration in the treatment cavity in order to practice the invention.

In another embodiment of the present invention, the ribs/bumps top surface is curved to smooth out any sharp angle which could be detrimental to the flow, in particular with regard to non linear flow defects which might arise from unsmoothed angles including the formation of micro-bubbles. Specifically, the shape of the bumps/ribs is to avoid such non linear flow defects.

In yet another embodiment of the present invention, the shape and relative disposition (pitch) of the ribs/bumps is determined by their ability to create local stress fields which can percolate into a cooperative network for specific conditions of rotation speed Ω , frequency ω and amplitude α of shear vibration, and temperature. That is the stress fields of one rib must overlap that of an adjacent rib.

Also included in this apparatus are means for continuously moving the melt, e.g. to drag and/or pump the melt located in the station treatment cavity from the inlet conduit towards its outlet conduit. This is accomplished by means of the disposition of the ribs with respect to the

rotational flow direction. If the ribs are disposed perpendicular to the rotation direction, the molten moldable material is dragged in that direction and any motion in the longitudinal direction is due to pressure flow exerted at the inlet of the treatment cavity. Pressure might have a negative effect on shear-thinning and/or the kinetics of disentanglement and, for this reason, should be maintained in the range which does not adversely affect the treatment. By orienting the ribs by a certain angle with respect to the axis of rotation, it is possible to create a longitudinal drag component with its own strain rate and acceleration. This longitudinal component of the rotational motion creates a pumping effect which reduces the pressure required to produce certain flow rate and contributes to the transport and disentanglement of the molten moldable material. Upon reading this specification, those skilled in the art will know what ribs orientation will produce the desired and controlled pumping effect in the treatment cavity in order to practice the invention.

In one embodiment of the present invention, the surface profile of ribs/grooves is created by machining and grooving the surface of the treatment cavity permanently and in another embodiment of the present invention, foils of profiled strips which are manufactured separately are laid over and firmly mounted, for instance tack-welded on the body of the treating surfaces. In one embodiment of the present invention the strips are made of a metallic material and, in another embodiment, they are made of any other material capable of sustaining the combination of shear forces, vibration and temperature required for the treatment.

The treatment station includes means for varying the

amplitude of the shear vibration applied on the molten moldable material as it flows through from the inlet to the outlet of the treatment station. US Patent 5,885,495 teaches that slippage may occur at high amplitude of melt shear oscillation which disrupts the efficiency of the
5 disentanglement process. When the desired amplitude for disentanglement is applied all at once, the risks of slippage are increased, which suggests to gradually increase the strain amplitude of vibration until the
10 correct combination of frequency-temperature and strain % is obtained. In a discontinuous process, such as described in US Patent 5,885,495, the settings and parameters are programmed to vary gradually, which avoids the slippage problem. Specifically, the strain % of the shear vibration
15 is increased little by little in step-wise fashion, given a certain temperature and frequency of oscillation. Likewise, at a given temperature and strain %, the frequency is increased little by little until the desired combined setting is achieved. For a continuous
20 disentanglement process, the gradual increase of the strain % can be accomplished by a gradual change of the geometry of the gap along the pathway of melt flow. The strain amplitude is function of the gap geometry, as explained before. For example, one can increase the shear strain %
25 by increasing the radius of the cylinders of the annulus which form the gap. Specifically, a preferred embodiment of the present invention uses a pair of concentric conic surfaces for defining the gap in the treatment cavity. This preferred embodiment offers the added advantage of
30 being able to easily change the gap dimension by pulling the inner cone surface with respect to the outside cone surface. Gaps between 0.5mm and 5mm can be obtained that way, which can be adjusted on the fly as the molten

moldable material flows in the treatment cavity. Another preferred embodiment consists of concentric cylindrical surfaces whose radius increases in stages as flow proceeds from the inlet to the outlet of the treatment cavity.

5 In one specific embodiment of the present invention, the gap height is automatically adjustable through an actuator and a controller allowing an increase or decrease of the gap at intermittent intervals, thus modulating the gap dimension in a controllable way.

10 Also included in this apparatus is a means for controlling the temperature of the material within the feeder(s), accumulator(s) and/or treatment cavity. Any suitable temperature controlling means can be employed when practicing this invention.

15 Examples of suitable temperature control means include, without limitation, the implementation of the following: (a) hot and cold oil circulated through passages in the injection machine, the accumulator, the injection nozzle and/or the mold, (b) resistance cartridges
20 positioned within the injection machine, the accumulator, the injection nozzle and/or the mold, (c) hot pipes inserted into the injection machine, the accumulator, the injection nozzle, and the mold, and/or (d) fluid which is embedded in the injection machine, the accumulator, the
25 injection nozzle and/or the mold and whose temperature can be controlled by dielectric means. Those skilled in the art will be able to select the temperature controlling means which best suits their needs after reading this specification.

30 Also included in this apparatus is a means for controlling the pressure of the molten moldable material within the feeder(s), accumulator(s) and/or disentanglement treatment cavities. In particular, the rate of flow

in the screw pump(s), gear pump(s), treatment cavities and extruder can be controlled by melt pressure in the treatment cavity measured by pressure sensors and transducers. Any suitable pressure controlling means can
5 be employed when practicing this invention. Those skilled in the art will be able to select the pressure controlling means which best suits their needs after reading this specification.

Also included in this apparatus is a means for
10 monitoring and/or controlling the torque of the material within the treatment cavity. The torque is directly related to the modulus of the molten moldable material being treated and reflects the state of viscosity. Any suitable torque monitoring and/or controlling means can be
15 employed when practicing this invention.

Examples of suitable torque control means include, without limitation, the implementation of the following:
(a) measure of the current used by the motor(s) to maintain a given rotation speed of the shaft driving the relative
20 motion of at least one surface in contact with the molten moldable material to be disentangled, (b) installation of a shear stress transducer at the end of the treatment cavity, (c) measure of the speed of transmission and attenuation of ultrasonic acoustic waves through the gap
25 in the treatment cavity. Those skilled in the art will be able to select the torque controlling means which best suits their needs after reading this specification.

In addition to the above, the apparatus can include many different optional features. For example, the
30 apparatus can include a purge valve which can be positioned between the accumulator(s) and the mold and/or between the feeder(s) and the accumulator(s).

Another optional feature which can be included in the

present invention is a mixing chamber positioned in the pathway of the inlet to the treatment cavity and/or at the outlet of the treatment cavity. This mixing chamber, which can consist of a static mixer in one preferred embodiment, can homogenize the temperature and the viscosity of the molten moldable material, and optimize the mixing of the different slices of the molten moldable material treated in the gap of the treatment cavity. In the case several feeders are used, the mixing chamber creates a more homogeneous mix of the different ingredients.

Specific embodiments of this invention are illustrated in the figures. These illustrations are merely examples of manners in which to practice the invention. They are, in no way, intended to limit the scope of this invention.

Other objects, aspects and advantages of the present invention will be apparent to those skilled in the art upon reading the specification and the appended claims which follow.

A more complete appreciation of the present invention, and many of the attendant advantages thereof, will be readily ascertained as the invention becomes better understood by reference to the following detailed description, when considered in connection with the accompanying figures briefly described below.

Referring now to FIGS. 1A to 1F, FIG. 1A is a schematic illustration of one embodiment of a disentanglement apparatus designed in accordance with the present invention having a single feeder 4 and a single treatment cavity 3, between an undersurface 2 of a top member 54 and a top surface 56 of a bottom member 58 wherein the shear vibration under extensional flow is produced by rotating at constant speed and/or at modulated speed and/or by pure oscillation an incoming melt 1 which

flows outwards over ribs or rib means 5 disposed on at least one surface 2. The molten moldable material 1 flows circularly from the center of the disk to the rim where a doctor blade collects it and a gear pump positively displaces it away from the treatment cavity. The cavity gap 3 is structured by the ribs 5 which can be shaped and configured in a variety of ways, as shown in FIGS. 1C, 1D, 1E and 1F, specifically designed to exert the maximum control over the strain rate, strain rate periodic variation with given frequency and extensional strain as the melt flows over the rib means from the inlet to the outlet and in order to optimize its disentanglement.

In one embodiment of the present invention, one surface in contact with the melt rotates at constant speed Ω and the other surface is fixed. The strain rate is locally modulated by the presence of the ribs which create the extensional vibration required to produce the high elastic state responsible for shear-thinning and, in time, disentanglement effects. The number of ribs, the spacing between them (pitch) the height of the ribs, the width of the ribs, and the height of the gap in between the ribs are all interrelated parameters of the present invention, which must be specifically designed in order to produce the adequate periodic strain rate with proper frequency, strain amplitude and frequency capable of producing disentanglement effects, as taught in US 5,855,495. For instance, for a gap height between 0.5 and 4mm, rib height can be approximately 25% of gap height and rib width 50% of gap height. The number of ribs, to be spaced regularly, depends on the strain and frequency required to increase elasticity/shear-thinning for the disentanglement process and on the speed of rotation which determines angular, and thus tangential velocity across the ribs. When the space

between the ribs is relatively important, care should be taken to design the ribs corners and shape angles to avoid local turbulence and other non linear flow defects. FIGS. 1A to 1F show several possible geometries for the ribs 5. In one embodiment of the invention (FIG. 1F), the height of the ribs is not constant across the radius and increases slightly from the center of the disk to the edge, or vice versa. In another embodiment of the present invention, the ribs surface is not flat but is, instead, grooved at regular intervals perpendicular to the radial direction of flow (FIGS. 1C and 1D).

In another embodiment of the present invention, the moving surface is not rotating but oscillates back and forth with a certain amplitude α and frequency ω . Yet, in another embodiment of the present invention a combination of oscillation and constant rotation is exerted on the moving surface. The value of the amplitude and frequency of vibration and the value of the melt temperature are determined according to US Patent 5,885,495 in order to increase the amount of elasticity in the melt, and through shear-thinning. The constant speed of rotation Ω is also determined to bring the melt within the proper processing window for disentanglement to occur, and its value is adjusted, along with the number of ribs, the geometry of the ribs and their disposition with respect to melt flow direction, in order to optimize disentanglement, that is to say favor the kinetics of viscosity reduction to make it as efficient as possible in the shortest time as possible.

In another preferred embodiment of the present invention, both surfaces in contact with the molten moldable material have ribs and/or grooves. Yet, in another important embodiment of the present invention, both

surfaces in contact with the molten moldable material are in controlled independent motion. In this case, although the apparatus is more complicated to design and operate, disentanglement is more efficient and occurs deeper. 5 throughout the cavity gap, allowing the use of wider gaps which results in increased productivity.

FIGS. 2A and 2B are schematic illustrations of one embodiment of a disentanglement apparatus designed in accordance with the present invention having a single feeder and a single treatment cavity, wherein the shear vibration under extensional flow is produced by feeding a 10 molten moldable material 3 through a gap (also identified by numeral 3) composed of at least one rotating and/or oscillating surface 6 or 7 presenting on its contour a profile over which the molten material flows and/or can be dragged and/or is being pushed through and/or pumped through. In FIG. 2B the outside surface 6 may be 15 rotated/oscillated at constant speed Ω and constant frequency and amplitude of oscillation (when oscillation is added) and the other e.g. cylindrical surface 7 may be fixed, or, conversely, surface 7 may be rotated/oscillated (FIG. 2A) at constant speed and surface 6 may be fixed, or, 20 both surfaces may be rotated/oscillated independently. The strain rate is locally modulated by the presence of ribs and/or grooves on either surface 6 and 7, or on both surfaces, which create the extensional and fatigue flow with vibration required to produce disentanglement effects. 25 As already emphasized, the number of ribs/grooves, the height of the ribs/grooves, the width of the ribs/grooves, 30 are all inter-related parameters of the present invention, which must be specifically designed in order to produce the adequate strain rate, strain amplitude and frequency capable of producing disentangle-ment effects, as taught

in US Patent 5,855,495. For instance, the rib on one surface might be a continuous bump configured as a right helix, whereas its counterpart on the facing surface across the gap is another helix with same pitch but turning left. The two helices can be grooved at an angle with the axis of the helix to produce a periodic variation of the gap height along the helix rib top surface. In such a case, when the two surfaces are put in relative motion, by the rotation of at least one surface, the melt is not only sheared and oscillated under extensional flow but also dragged through towards the exit end. The molten moldable material 3 flows helicoidally from the inlet of the treatment cavity, at the left of FIG. 2A, toward the outlet, at the right, where it is collected and a gear pump positively displaces it away from the treatment cavity, or it is used to fill a mold cavity which happens when the invention is applied in conjunction with a valve gate system in hot runner applications, for example. The cavity gap 3 is profiled by the ribs which can be shaped and configured in a variety of ways, as already shown in FIGS. 1A to 1F, specifically designed to exert the maximum control over the strain rate, strain rate variation and extensional strain as the melt flows from the inlet to the outlet and in order to optimize viscosity reduction and disentanglement. Band heaters 8 are located around the metallic surfaces defining the cavity and are controlled by thermocouples (not shown in FIG. 2A) in PID control loops 32.

The apparatus of the invention as illustrated in FIGS. 2A and 2B thus is for continuously reducing the viscosity of molten moldable polymeric material 3 by shear vibration under extensional flow to cause shear-thinning conditions ideal for disentanglement, the apparatus including at least

one station treatment cavity defined by a gap composed of two closely separated surfaces 6 and 7 in relative motion with each other at given speed and/or submitted to relative oscillations, with given frequency and amplitude, as to
5 produce a shear deformation on the molten moldable material 3 and a controlled variation of the gap dimension by axial, circumferential, radial or other undulations of either or both surfaces 6 and 7, presenting on their contour a profile of ribs and/or bumps and/or grooves over which the
10 molten moldable material 3 can flow and/or can be dragged and/or is being pushed through and/or pumped through. The treatment cavity has an inlet 30 through which the molten moldable material 3 can pass into and an outlet 31 through which it can exit the treatment cavity. The treatment
15 cavity geometry allows the shear strain and the shear strain rate imposed on the passing molten moldable material to vary controllably. At least one feeder 33 is for preparing the molten moldable material 3, the feeder including a means for expelling the molten, moldable
20 material through its outlet into the station treatment cavity inlet 30. In this embodiment at least one accumulator 34 is provided for collecting the treated molten, moldable material before it is sent to either a pelletizer system or to a pumping station or to a mold
25 cavity. Means such as a driver 35 are provided for exerting the shear vibration of given frequency and amplitude coupled with fatigue extensional flow on the molten moldable material 3 contained within the treatment cavity. An example of means for controllably varying the
30 gap dimension which the molten moldable material can pass and/or flow through are shown in the embodiment of FIG. 6 there a central member of the apparatus is generally conical and can be moved axially (laterally) to vary the

gap. An example of means for continuously venting the treatment cavity to prevent the formation of bubbles or cavitation during the treatment is schematically shown at 36 in FIG. 2A. An example of means for monitoring and controlling the temperature of the molten moldable material contained within the feeder 33 is schematically shown at 37 in FIG. 2A, for the treatment cavity is schematically shown at 38 and for the pumping station is schematically shown at 39. An example of means for monitoring and controlling the pressure of the molten moldable material contained within the feeder, the treatment cavity and the pumping station are shown schematically at 47, 48 and 49 respectively. An example of means for monitoring and controlling the torque exerted on the molten moldable material contained within the treatment cavity is shown at 50 in FIG. 2A and 2B. With several stations connected to one another through gear pumps or screw pumps such as those at 11 in FIG. 3 or 26 in FIG. 7, the chain of treatment stations have a first station connected directly or through a gear pump and/or a static mixer (e.g. 52 in FIG. 7) to an extruder and a last station connected to a pelletizer line or a pumping station, or a mold cavity for immediate use of the disentangled melt.

In one embodiment of the present invention, one surface in contact with the melt, which has a pattern of ribs as explained in FIG. 1A, is moved longitudinally with speed V , as shown for surface 7 in FIG. 2A, and the other surface is rotated/oscillated. The melt undergoes the effect of a combined (tensorial) periodic strain rate with its two components, in the longitudinal and rotational directions, adjusted to impose an optimized disentanglement effect. Yet, in another embodiment of the present invention, the longitudinal motion of 7 occurs with

modulated periodic speed in order to incorporate an oscillation component susceptible to facilitate the raise of the elasticity of the molten moldable material in the gap while the lateral rotation of 6, with or without its own oscillation, contributes, among other things, to the extensional fatigue in order to disentangle the melt according to US Patent 5,885,495.

In another embodiment of the present invention, the moving surface is not rotating but oscillating back and forth with a certain amplitude α and frequency ω . Yet, in another embodiment of the present invention a combination of oscillation and constant rotation is exerted on the moving surface(s). The value of the amplitude and frequency of vibration, the melt temperature, and the constant speed of rotation Ω , along with the number of ribs, are determined according to US Patent 5,885,495 in order to increase the amount of elasticity in the melt, and cause the melt to undergo extensional flow and fatigue, at given value of elasticity (shear-thinning) of the melt, two critical conditions for disentanglement to occur. Specifically, the number of ribs, the geometry of the ribs and their disposition with respect to melt flow direction, are designed not only to create specific acceleration and deceleration patterns in the cavity gap susceptible to favor the kinetics of disentanglement, but, at the same time, to allow a certain pumping of the melt towards the outlet of the treatment cavity by a dragging effect from the component of the strain rate tensor which aligns with the longitudinal direction of flow.

FIG. 3 is a schematic illustration of one embodiment of a disentanglement apparatus designed in accordance with the present invention having a single feeder and more than one treatment cavity, separated by a pumping section 11,

wherein the shear vibration under extensional flow in the treatment cavity section is produced by feeding a molten moldable material 3 through a gap composed of at least one rotating and/or oscillating surface 10 presenting on its contour a profile 12 over which the molten material flows and/or can be dragged and/or is being pushed through and/or pumped through, and a barrel 9 which can also be rotated/oscillated or can be stationary and which is heated/cooled through outside means (not represented). The surface profile(s) or rib means 12 consist of different patterns depending on the location of the treatment station along the extrusion axis and adapted to the progressive change of the viscosity. FIG. 3 displays a few patterns possible such as small square/round bumps on the surface, or tilted and elongated ribs creating a split of the strain rate along the rotational and longitudinal axes, but many other designs, all referred to here as rib means, are conceivable, which a person skilled in the art will be able to adapt after reading this disclosure. The pumping station 11 is made of the few flights of a screw pump with given pitch and helix angle to pump the melt at given rate from one treatment station to the next by rotating the core surface 10. The apparatus described in FIG. 3 can be adapted to interface with the basic hardware of an extruder which already provides a barrel 9, heating and cooling means, and a motor to produce the rotation of the moving surface 10, which would replace the screw of the extruder. In one embodiment of the present invention, the feeder which provides a liquid molten moldable material to the treatment cavity is itself another extruder or an extruder connected to a gear pump. For example, the molten moldable material is produced by the first extruder and is distributed through a breaker plate and a cross-head die

to the inlet section of the second extruder, perpendicular to the first extruder, which attaches to the left end side of FIG. 3, and as illustrated in FIG. 7 by elements 23 and 24. The moving surface 10 is rotated by the second extruder and its barrel 9 is stationary. The total length of the barrel is adjustable and is function of the numbers of stations required to achieve a certain level of viscosity reduction and the throughput desired.

Yet, in another embodiment of the present invention, only one extruder is needed, but the screw, which is replaced by the treatment apparatus described in FIG. 3 in order to practice the present invention, is substantially extended in length to have a first section performing like a normal screw with its own melting, metering and pressurizing zones, followed by the disentanglement sections described in FIG. 3.

FIG. 4 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow is created in the gap 3 of concentric conic surfaces 13 and 14 in relative motion with respect to one another, at least one of which presenting an array of ribs and bumps 12 and rotating at constant speed Ω , and/or at modulated speed

$[\Omega + \sum \alpha_i \sin(\omega_i t + \theta_i)]$ and/or in a pure oscillation mode $\sum \alpha_i \sin(\omega_i t + \theta_i)$, where $\alpha_i, \omega_i, \theta_i$, are, respectively, the modulated amplitude, the frequency and the phase of the oscillation (the subindex "i" refers to the decomposition of the periodic motion into its Fourier's components). As explained before, the speed of rotation, the oscillation parameters, the pitch between the ribs, the shape and dimension of the ribs/bumps and grooves (between the ribs),

and the angle of tilt with respect to the cone axis of the ribs, are all inter-related and depend on the strain rate and acceleration required to produce disentanglement, as taught in US Patent 5,885,495.

5 In FIG. 4, the gap height or width can be increased or decreased by the relative axial or lateral translation of the concentric cones, which can be done through the implementation of linear bearings on the shaft in the direction of the straight double arrow. In a particular and specific embodiment of the present invention, the gap width is automatically adjustable through an actuator and a controller opening or closing the gap, allowing an increase of the gap at intermittent intervals, to facilitate the extrusion of the treated molten moldable material out of the treatment cavity. The reason for such a feature can easily be understood: the disentanglement process requires a relatively narrow gap to implement the conditions of strain and strain rate susceptible to disentangle the melt; but these conditions are not favorable to the throughput. By opening and closing the gap intermittently between a value which is favorable to the treatment and a value which is favorable to the throughput, a compromise can be found which optimizes the disentanglement process. The period between two successive openings of the gap can be programmed between 1 second and 10 minutes, without exclusion of other possible values. The variation of the gap width or height between its minimum and maximum value can be programmed between 5% and 200% of the treatment value.

30 It is another object of the present invention to have a central computer controlling the value of all the parameters of the disentanglement process. Specifically, in FIG. 4, items 8 and 15 are shown to respectively

represent band heaters and cooling circuits to regulate precisely the temperature of the body of the surfaces in contact with the molten moldable material. Temperature probes (not shown) are inserted near the molten moldable material to send electrical signals back to the central processing unit for activating the heating or cooling means in order to maintain the temperature in the gap constant. The operator can interface with a user friendly computer screen to enter all the parameters required to operate the disentanglement apparatus according to the present invention. For example, the operator enters the value of the temperature in the treatment cavity, the speed of rotation of the inner cone 14, the amplitude and frequency of the oscillation of the cone (which can be zero when there is only constant rotation and the rib means alone create the vibration effect) and the period and amplitude of the gap intermittent opening (whose variation can also be zero).

FIG. 5 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow is created in the gap of a series of concentric cylindrical surfaces 13 and 14 of increasing larger diameter, in relative motion with respect to one another, at least one of which presenting an array of ribs and bumps and rotating at constant speed and/or at modulated speed and/or in a pure oscillation mode.

FIG. 6 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow is created in successive sections of the treatment cavity having a

variable gap arising from the relative separation of concentric surfaces in relative motion with respect to one another, at least one of which presenting specifically configured and disposed ribs and bumps 12 and rotating at constant speed and/or at modulated speed and/or in a pure oscillation mode. As shown in Fig. 6, the shape of the ribs/bumps and their relative position is varied along the pathway of the molten moldable material in the treatment cavity depending on the amount of local melt pressure, the magnitude of the longitudinal drag strain rate desired and the speed and mode of rotation of the moving surface.

FIG. 7 is a schematic illustration of one embodiment of the disentanglement apparatus designed in accordance with the present invention wherein the feeder 23 is an extruder and/or a plunger, and the disentanglement treatment occurs in a series of stations 24 connected through gear or screw pumps 26. In each of the stations 24, means 25 are provided to rotate and/or oscillate at least one surface in contact with the molten moldable material passing through it in order to create flow conditions favorable to disentanglement, as described in US Patent 5,885,495 and in this disclosure. The temperature, the rotation speed, the oscillation amplitude and frequency are independently set for each station, but are controlled from a central processing unit which overlooks all aspects of this multi-stations disentanglement process. For practicing particular embodiments described earlier in this Patent application, stations 24 and the means 25 can be provided for by traditional extruder drives, for which barrel and screw can be specifically adapted and modified to conform to disclosures specified in FIGS. 3, 4, 5 or 6. Mixing chambers can be added in 26, as well as in 25 means to

modulate the gap dimension intermittently. At the end of the multi-stations, the disentangled melt is either pumped into an accumulator, not shown in FIG. 7, but described in this disclosure to prevent the melt to re-entangle, and this accumulator is connected to a pelletizer line and/or to the feed of an extruder or injection molding equipment, or the exiting melt is directly sent into a cavity, such as a mold cavity of a hot runner system using a valve gate modified to encompass the present invention.

FIG. 8 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow on a molten moldable material 3 is created in the gap between cylindrical or conical rollers 27 in close contact to one another and a core surface at the center 28 with ribs, bumps or grooves 12 and which is either cylindrical or conical. FIG. 8 can be considered a cut through the system of cylinders which one could imagine vertical, with their axis perpendicular to the Figure, and the flow of molten moldable material occurring from the inlet at the top, along the axis of the cylinders or cones, towards the outlet at the bottom. More generally, at least one surface 27 or 28 is rotating at constant speed and/or at modulated speed and/or in a pure oscillation mode, as explained in several previous figures. For instance, both surfaces are turning at constant speed, respectively Ω and Ω' . The rotation of rollers 27 can be from individual power sources or through a common drive, shown as 29 in FIG. 8, which can be a belt or any other appropriate driving mechanism known to the art. At least one surface has a set of ribs/bumps 12 or grooves on it, shaped, disposed and configured to practice the invention, as already disclosed several times for the

other figures. It is easy to conceive the presence of ribs/bumps or grooves on the core cylinder 28. Some are shown in FIG. 8. Rollers 27 are in close contact but do not touch each other directly, so they can turn in the same direction (counterclockwise in FIG. 8). In one embodiment of the present invention, the distance which separates them is very small, of the order of 0.03 to 0.05mm in order to prevent the molten moldable material 3 to come through their separation. A set of spring loaded ball bearings can be used to keep the rolling cylinders at very close proximity while rotating in the same direction through means 29. In another embodiment of the present invention, rollers 27 can have ribs and bumps disposed on their surface so that they intermesh in the region of closest proximity. An example of this embodiment is shown at the bottom of FIG. 14. Starting at a certain speed of rotation Ω c, the layers of molten moldable material in contact with 27 have enough elasticity built up from the speed of rotation of rollers 27 to "stretch over", i.e. ignore the contours of the gap formed where the rollers intermesh or come into close contact, and create an homogeneous round circularly rotating layer which encapsulates and drives in its rotation the rest of the molten moldable material enclosed in the gap between the rollers 27 and roller 28. The stretch-over moving wall, created by cooperative motion across the rotating rollers, can be used, in one preferred embodiment of the present invention, to minimize or avoid slippage at the wall while practicing the disentanglement process through rotation/oscillation of 28. Furthermore, this embodiment allows far greater speed of extrusion through the treatment cavity than is normally available at low pressure drop, because wall slippage is known to prevent fast extrusion throughput. Finally, depending on

the speed of rotation of 28, processing the molten moldable material under conditions where a stretch-over layer is created can present positive advantages on the kinetics of disentanglement.

5 FIG. 9 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow is created by the relative motion in the molten moldable material 17 of
10 a train of oblong members or ovally-shaped wagons 16 circulating, at constant speed or at a modulated speed, in a torus ring in communication with an inlet 21 and an outlet 22 conduits. The motion of 16 is driven by a cylinder 19 through links 18. Cylinder 19 is driven by an actuator
15 connected to a motor. Links 18 are a spider ring connecting rigidly 19 and 16, and letting the flow pass through without resistance or interference. The torus is jacketed by a thermal fluid and/or band heaters and water passages allow control of the temperature inside the ring.
20 As motion of 16 in the molten moldable material 17 is driven by 19, the gap at any specific location of the ring periodically increases and decreases as the ovally shaped wagons pass through it, and thus periodic shear strain conditions are created with intensive extensional flow
25 around the wagons. When the conditions of strain rate and strain rate variation (acceleration/deceleration) are set to practice disentanglement in the molten moldable material according to US Patent 5,885,495, the treated melt is being pushed ahead towards the outlet hole 22 communicating with
30 the inlet 21 of the next torus ring one station down. An assembly of torus rings 67 piling up on top of one another and communicating through their respective inlet and outlet holes is shown in FIG. 10.

FIG. 10 is a schematic illustration of one embodiment of the disentanglement apparatus designed in accordance with the present invention wherein the feeder is an extruder and/or a plunger feeding molten moldable material in 21 of the first torus ring, the disentanglement treatment occurs in each of a series of superposed and interconnected ring stations 17 defined by a tubular track through which a train of ovally-shaped wagons 16 circulate at controlled speed, driven by a common core cylinder 19, attached to the wagons. The molten moldable material is treated according to the present invention, as described in FIG. 9, and exits a ring station in 22 which directly connects with the inlet of the ring station located under it. At each station, the temperature is adapted to account for the change in viscosity produced by disentanglement. At the bottom of the column in FIG. 10, the outlet of the last ring is connected to an accumulator 20 which is itself connected to a pelletizer line and/or the feed of an extruder or injection molding equipment (not shown).

FIG. 11 is a schematic illustration of one embodiment of the power transmission of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration and the controlled extensional fatigue of the molten moldable material in the treatment cavity are applied on both axes 310 and 320 of an epicyclic differential. An epicyclic differential is composed of 3 principle elements: (a) the case planet carrier 310; (b) the torque arm sleeve sun gear 320; and c) the central bore sun gear 330. These 3 elements are mechanically connected by the sun and planet gears which make up the epicyclic differential gear train.

The shaft attached to the surface to rotate and/or oscillate in the treatment station of the present

invention, goes into 330. The Willis formula establishes the relationship between the rotational speeds of the 3 elements 310, 320 and 330, allowing to combine at 330 the motions which are imposed, independently, at 310 and 320:

5

$$N_3 = p N_2 + N_1 / K$$

where N1 is the rotational speed of the case 310, N2 is the instantaneous rotational speed of the torque arm sleeve motion 320, and N3 is the rotational speed found for the motion of the central bore 330. p is the internal ratio
10 between the torque arm sleeve 320 and the central bore 330, i.e. when there is no rotation on the 310 axis (the case is maintained fixed). Likewise, K in Willis equation is the reduction ratio between the case 310 and the central bore 330, i.e. when there is no rotary oscillation on N2.
15 Thus a differential also operates as a gear box with reduction ratio K.

Specifically, a rotary vibration motion can be set at 320 and a pure rotation with given controllable speed of rotation at 310. The motions on axes 310 and 320 are
20 totally independent and can be programmed separately. The combination of motions 310 and 320 arises naturally at 330 with no or minimum backlash. According to one embodiment of the present invention, an epicyclic differential can replace the gear box which connects the motor of an
25 extruder and the shaft driving the screw. This replacement allows to simultaneously rotate and oscillate such a shaft as disclosed in FIGS. 3 or 4 which is introduced in the barrel, provided the other axis of the epicyclic differential is driven by a rotary vibrating device. This
30 device can also be used to drive the screw of an injection molding machine, combining rotation and oscillation of the

screw, a combination which produces easier flow through extensive shear-thinning.

FIG. 12 is a schematic illustration of one embodiment of the shear vibration transmission of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration in the molten moldable material in the treatment cavity is created by the oscillation of the inner cone of a concentric conical assembly 114 attached to one axis of an epicyclic differential, the rotary oscillation being created through a cam 110 and a piston 112. In this embodiment, the treatment cavity is a pair of concentric cones, such as disclosed in Figure 4, with a constant gap clearance 114 where the molten moldable material (which fills 114) is sheared and oscillated back and forth through handle 110 activated by a piston 112. Frame 116 includes the heating/cooling jacket filled with thermal fluid and the thermocouple to control the temperature. The clearance gap between the two cones can be adjusted at the beginning of the operation between 0.5mm and 5mm. Once adjusted it remains constant throughout the treatment. The piston arm can be attached to the handle 110 at different level positions 113, which allows to modify the amplitude of the shear rotational vibration along with the torque available. The piston displacement can also be varied, which allows another degree of freedom to modify the amplitude of rotation of the rotary oscillation. The molten moldable material is entered in the apparatus through an extruder and gear pump shown as 111, and is pulled out of the cone assembly through another gear pump 115. In this particular embodiment, the gear pump at the outlet recirculates the throughput back to inlet 111 by a separate connection, allowing several passes before the melt is pumped out of the disentanglement

apparatus. In this embodiment, the disentangled melt is produced intermittently but in a continuous way. The extruder inlet valve and the outlet valve 117 to the pelletizer unit are both closed while the several passes of molten material through the treatment cavity occur in close circle. When a desired viscosity reduction has been achieved, after several passes, the extruder is signaled to let in a new batch of untreated molten moldable material in the treatment cavity. The gap clearance can be opened wider during this purge to prevent the disentangled melt to be pressurized. The outlet valve 117 to the pelletizer line is kept open and the recirculation outlet valve (not shown) is kept closed during the purge of the treatment cavity. Conversely, those valves are respectively closed and open during the multi-passes treatment.

FIG. 13 is a schematic illustration of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the shear vibration under extensional flow in the molten moldable material is created by the relative motion of two concentric conic surfaces at constant speed or at a modulated speed, i.e. combined constant and vibrated speed, or under pure rotary oscillation while the molten moldable material is pushed and/or dragged and/or pumped from the inlet conduit 220 to the outlet conduit 270 of the treatment cavity (FIG. 13). In this embodiment, unlike for the embodiment disclosed in FIG. 12 which applies to an intermittent release of treated molten material, the treatment cavity and its attachments clearly define a station, such as item 24 in FIG. 7, suitable for a continuous disentanglement process. It is clear in FIG. 13 that flanges 220 and 270 are identical, allowing a repeat to other stations of what can be considered standard

features in the description of this station. Molten moldable material 250 enters the treatment station through inlet 220, goes through a spider and fills the cavity created by two concentric cones defining a gap. Inlet 220 can be connected to a gear or screw pump. For the first station, as disclosed in FIG. 7, an extruder is used ahead of the gear pump to prepare the molten moldable material and pressurize it to a given value. The inside cone is continued beyond the treatment section 260 to become 280, which is connected to the epicyclic differential with its two independent motion axis, as described in Figure 11. The molten moldable material 250 is flowing from the inlet 220 to the outlet 270 due to the combination of pressure flow, and the rotation of the inside cone and the presence of tilted ribs on the surface of the inside and outside cones, as explained in FIGS. 4 or 6. While the molten moldable material 250 passes through the gap created by the cones, it disentangles due to the combined action of the rotation and/or oscillation of the inside cone 260 through the shaft 280. The temperature and pressure of the molten moldable material is measured at several places 210 along the pathway from 220 to 270. Band heaters 230 are wrapped around the frame and water passages 240 and 242 are made to cool 250 from both the outside (240) and the inside (242) in order to maintain its temperature precisely controlled. The outlet 270 can be connected to a gear or screw pump. In the case of a gear pump, the rotation speed of the pump is controlled to build the melt pressure back to the value required for the next treatment station. FIG. 7 discloses how to build a full chain of treatment stations, such as the one disclosed in FIG. 13, in order to continuously and economically disentangle a polymer resin. The cones can be replaced by cylindrical cavities.

FIGS. 14A and 14B are schematic illustrations of one embodiment of the disentanglement apparatus designed in accordance with the present invention wherein one or several feeders 420 feed molten moldable material to a series of tubular cavities 450, intermeshing or not, which themselves connect, directly (FIG. 14B) or through pumping sections 400, to other series of tubular cavities, until the last array of treatment stations, which connect to a collector 410 and a pelletizer line and/or the feed of an extruder or injection molding equipment (not shown). The surfaces of the tubular cavities are covered with ribs/bumps and/or grooves 450, designed to synthesize the proper strain rate profile to achieve disentanglement and to drag the molten moldable material forward to the next tubular station and along the axis of the tubular cavities. At least one surface 430 is moving in order to shear the melt under vibration and accomplish the extensional fatigue which are the two requirements to produce disentanglement efficiently. This can be exerted through the rotation of rotor 430 at given rotation speed, or by rotary vibration of rotor 430 at given frequency and amplitude, or by combination of the two previous motions. At the FIG. 14B, the two tubular cavities are positioned to intermesh in their center, along their axis, which is an embodiment useful to apply in the accumulator at the final outlet before the pelletizer line to prevent recovery of the disentanglement.

FIGS. 15A to 15C are schematic illustrations of one embodiment of the treatment cavity of the disentanglement apparatus designed in accordance with the present invention wherein the ribs/bumps on at least one of the surfaces in contact with the molten moldable material have a specific shape to create a given profile of strain rate and strain

rate variation under extension, and are aligned in a specific manner with respect to the axis of surface rotation in order to create a certain amount of drag flow. FIGS. 15A to 15C define some of the shape parameters and the angle of tilt. The contours of the ribs are calculated to eliminate any possibility of turbulence or other non linear defect in the flow, which should remain laminar. The height of the rib, e , the radius $R1$, $R2$ and $R3$ which define the curvature to accelerate ($R1$) or decelerate ($R3$) the flow, can be different along cross sections aa and bb (the cross section bb is not shown here). The strain rate vector decomposes into two components, which defines two directions of drag flow. The shape of the ribs along each axis aa or bb is determined by the strain rate profile desired in these directions. Both profiles can be used to define conditions of shear vibration and/or extensional fatigue which can couple up to optimize shear-thinning and disentanglement effects.

According to another object of this invention the temperature controls of the molding apparatus are varied to enable cooling of the melt at a given rate until a given temperature is reached, while simultaneously adjusting the frequency of shear oscillation as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement efficiency. According to another technique of the invention, the melt temperature is controlled to change between two temperature values by alternately cooling and heating the melt while disentanglement or simply shear thing takes places, and simultaneously adjusting the frequency of shear oscillation as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement

efficiency.

In one embodiment of the present invention, the melt temperature in each of the treatment stations is made to vary along the pathway of the extrudate passing through it, by gradually varying the velocity of a thermal cooling fluid flowing in the coolant runners embedded in the barrel walls, to produce a cooling effect with determined rate from the inlet of the station to the outlet, provided the coolest temperature is greater than the T_c whose value is determined by the given cooling rate.

In one embodiment of the invention, cooling fluids are circulating within runners located in short sections of the treatment station, each section with its own fluid velocity controlled by the amount of opening/closing of a needle gate located at the inlet or outlet of the cooling section.

In another embodiment of the present invention, cooling effect within a treatment station can be realized by increasing or decreasing the cross-section area of the runners and/or changing their location with respect to the moving melt under treatment and/or by interposing materials, heat sinks or gaps of different thermal conductivity (for instance air gaps) between the cooling medium and the melt passageway.

In yet another preferred embodiment of the present invention, the temperature of the thermal fluid passing through the embedded runners varies, for instance by controlled mixing of two fluids at two different temperatures, or the endlessly repeated sequence of the successive passage of two fluids maintained at different temperatures.

In another embodiment of the present invention, the melt temperature in the treatment station is locally varied by means of dielectric heaters and/or microwave heaters in

combination with conductive means.

Any suitable temperature controlling means can be employed when practicing this invention. Those skilled in the art will be able to select the temperature controlling means which best suit their needs after reading this specification.

In another embodiment of the present invention, the apparatus which produce shear oscillation under extension of the melt is constantly adjusted as a function of the temperature of the melt, while the disentanglement treatment takes place in the station, in order to keep the relative elasticity, (G'/G^*) equal to a predetermined value. Any suitable controlling means of the oscillation frequency and amplitude as a function of temperature can be employed when practicing this invention, including, without limitation, closed loop controllers, PID controllers, fuzzy logic controllers and the like. Those skilled in the art will be able to select the controlling means which best suit their needs after reading this specification.

In another embodiment of the present invention the means that produce the continuous drag at constant speed of the melt by relative motion of the surfaces confining the melt are constantly adjusted as a function of the temperature of the melt, while the disentanglement treatment takes place in the station, in order to keep the relative elasticity, (G'/G^*) equal to a predetermined value. Any suitable controlling means of the drag speed as a function of temperature can be employed when practicing this invention, including, without limitation, closed loop controllers, PID controllers, fuzzy logic controllers and the like.

FIG. 16 demonstrates the effect of cooling rate on the

value of the onset of crystallization temperature, T_c for a semi-crystalline polymer melt. The slowly cooled melt, cooled at approximately $10^\circ\text{C}/\text{min}$, starts to crystallize at 240°C . This corresponds to the sharp drop of the specific volume in the bottom trace of FIG. 16, associated with the formation of a more dense phase, the crystallites. FIG. 17 shows that the melt elasticity of the amorphous state is disrupted and interfered with by the presence of the crystallites which act as cross-linking tie points between the macromolecules. The sharp increase of the relative elasticity is not due to an increased cooperative interaction between the macromolecules, which is a criterion to induce disentanglement, but rather to the increase of the tie points.

The upper trace in FIG. 16 corresponds to a melt cooled at approximately $200^\circ\text{C}/\text{min}$. The onset of crystallization is now at $T_c = 180^\circ\text{C}$, which is 60°C below the temperature of crystallization for the slowly cooled melt.

As a consequence of the effect of cooling rate on the value of T_c , the amorphous nature of the melt is preserved to lower temperatures, here an extra 60°C . Temperature is decreasing as one submits the melt to a fast cooling rate, and, eventually, it reaches the new T_c where crystallization takes place. For instance, it takes 18 seconds to cool from 240°C to 180°C at $200^\circ\text{C}/\text{min}$. If we control temperature in such a way that it starts to raise just before it reaches the new T_c , then the melt does not crystallize and remains amorphous over an extended 60°C , which can be made available for disentanglement purpose by increasing its elasticity through shear oscillation under extensional fatigue. For instance, if one chooses to disentangle the amorphous melt when its relative elasticity

(G'/G^*) = 0.83, FIG. 17 shows that for a frequency of 10 rad/s, it is not possible to reach that elasticity level without the interference of an intense crystallization which takes place starting at $T_c = 242^\circ\text{C}$. The value of (G'/G^*) is only equal to 0.3 at the onset of crystallization, for this frequency. This value can be raised by using higher frequency of shear oscillation. Using the time-temperature superposition principle which applies to the rheological behavior of viscoelastic melts, one can calculate the frequency which should be applied to raise the elasticity of the melt at $T = 242^\circ\text{C}$ from (G'/G^*) = 0.3 to (G'/G^*) = 0.83. The suggested frequency is 5,382 rad/s (857 Hz). While achieving such frequency of oscillation is certainly doable, it would not be economically practical and/or would cause serious problems of noise in a production environment. A preferred solution is precisely the purpose of the present invention. If the temperature could be lowered to 220°C , which is below the T_c at slow cooling rate (and therefore not doable without the means of the present invention), but above the T_c of the fast cooling rate, the frequency of oscillation to raise (G'/G^*) to the proper disentanglement range becomes 166 rad/s (27 Hz), which is within the practical range of acceptable industrial solutions, for economic reasons. Reducing temperature further down to 200°C would lower oscillation frequency even lower, in order to maintain the same value for (G'/G^*).

According to the present invention, as applied to Nylon 66 for example, the melt temperature is decreased during the disentanglement treatment in such a way as to lower T_c by e.g. 60°C , by the effect of a fast cooling rate on T_c , and subsequently increased to avoid cooling past 180°C , which is the T_c of the fastly cooled melt, until

- another temperature is reached, at which stage the cooling cycle starts over again. More specifically, the upper temperature can be $T = 245^{\circ}\text{C}$, and the lower temperature 185°C . The cycle time to reverse the change of temperature
5. is 18 seconds. This can be achieved in many different ways by those skilled in the art, who will be able to select the temperature controlling means which best suit their needs.

One advantageous and economical means consists in alternatively pumping at fast flow rate a thermal fluid accumulated in isothermally maintained tanks at two

10 temperatures, one temperature higher by approximatively 15°C than the upper temperature of the cycle range, the other lower by also 15°C than the lower temperature of the range. The individual fluids are only pumped for a portion

15 of the cycle time to allow for inertia effect to take place at the switch of pumping fluids. The temperature of the fluids in the tank can be adjusted, along with the proportional timing for alternating the pumping of the two fluids, in order to achieve the correct temperature profile

20 which fits the disentanglement treatment. Those skilled in the art would know how to design and control such a system, built with temperature controllers, thermocouples, heating and cooling means, and a servo-controlled switching valve, all in a servo-loop conveniently programmable from

25 a master computer.

In another embodiment of the present invention, the temperature is profiled along the pathway of the flowing melt by controlling the flow rate of the cooling thermal fluid circulating in the coolant runners or the cooling

30 jackets inside the die and/or barrel of the treatment cavity. For example, the coolant thermal fluid can circulate, from the exit end of the treatment station towards the inlet, in a spirally shaped runner of

decreasing cross-section, located further away from the melt cavity as it gets closer to the inlet end of the station, resulting in a gradient of cooling capacity along the melt flow axis. Other means known to those in the art of mold temperature control, such as band heaters, heat pipes, heat sinks, air gaps etc, can be used to create the correct temperature difference between the inlet and the outlet of the treatment station to generate the desired cooling rate profile as the melt passes through. The same temperature profile can be repeated from station to station, maintaining the melt in an amorphous state as disentanglement proceeds.

For a given temperature, elasticity increases with frequency. For a given frequency, elasticity increases as temperature decreases. In order to control precisely the amount of elasticity of the melt at each temperature, a preferred embodiment of the present invention is to adjust the value of the frequency of shear oscillation to optimize disentanglement at each melt temperature. Accordingly, in a preferred embodiment of the present invention, the number of ribs, bumps or grooves per turn on the rotating surfaces of the treatment station varies with the temperature of the melt at that location. The density of rib means decreases as temperature decreases, and conversely, increases as temperature increases along the pathway.

When the disentanglement system is made up of a series of small sections which can be independently controlled mechanically and thermally, each section temperature can be coupled with the optimum disentanglement variables at that temperature: the best oscillation frequency, the best rotation speed, and the best strain amplitude for the extensional fatigue. The temperature profile is determined by the temperature difference between adjacent sections.

When the temperature of the thermal fluid is cycled in time, the frequency of oscillation is synchronized to vary according to a program to maintain the desired elasticity profile that optimizes disentanglement at all times. For most practical applications, frequency can be varied between 1 and 100 Hz.

FIG. 18 illustrates an embodiment of the invention 500 that includes a pair of polymer melt treatment chambers or annular (cylindrical or conical or both) gaps 506 and 510, defined between the inner and out surfaces of a continuously rotatable (Ω) and/or rotatably oscillatable (ω , α) rotor or cylinder 508, and the outer surface of inner fixed cylinder or cone 504, and the inner surface of outer fixed cylinder or cone 512. The surfaces of rotor 506 are provided with the rib means of the present invention and/or the surfaces of fixed members 504 and 512 that bound gaps 506 and 510 have the rib means thereon, as long as there are at least one set of rib means in each chamber. A fixed shaft or support 502 carries the fixed member 504.

Examples of possible diameters for the surfaces in the embodiment of FIG. 18 are: outside of 502 is 7.62cm; outside of 504 is 15.24cm; inside of 508 is 15.88cm; outside of 508 is 23.50cm; inside of 512 24.36cm; and outside of 512 is 32.98cm. The gaps 506 and 510 are typically 1.6mm to 6mm but can be thicker or thinner depending on the type of melt and the element diameters. This is for minimum through-put of 65 kg/hour for polycarbonate.

FIG. 19 illustrated an embodiment of the invention that has a rotor, stem, screw, tube or shaft 602 for bounding the inside of a melt treatment gap and rotatable and or oscillatable about its axis. Rib means 604 in the surface of rotor 602 are in the form of honeycomb rib

means. Although hexagonal cells of the honeycomb means 604 are illustrated, other polygonal shapes can be used. Each cell has a pair or side wall 606 that extend at an angle θ to the flow direction F which is also parallel to the rotor axis. Each cell also has upstream and downstream transverse walls 612, 608, and upstream and downstream angles walls 614, 610. The transverse walls extend at an oblique angle to the flow direction F while the angles walls extend at an acute angle. These walls also intersect the upstream and downstream ends of the next side wall 606 in the honeycomb means. This arrangement has been found by the inventor to be very useful in that it causes the repeated and periodic dividing of the melt as it passes each intersection as the rotor rotates, thus advancing the local acceleration/ deceleration of the melt at each intersection and creating the viscosity reducing effect. Typical wall thicknesses for the walls of the honeycomb means is from about 0.15mm to about 1.0mm, depending on the diameter D of the rotor 602 and the rotation speed (continuous and/or oscillating), frequency and amplitude, and melt type. Wall height is typically about 0.1mm to 1.0mm. The distance d between side walls 606 also depends rotor diameter, speed and melt type. Distance d is typically about 1.5 to 5.0mm. The angle θ may be 2 to 85° or preferably about 30 to 60°.

The embodiment of FIGS. 20 and 21 comprises a rotor, stem, screw, tube or shaft 702 for bounding the inside of a melt treatment gap and rotatable and or oscillatable about its axis. Rib means 704 in the surface of rotor 702 are in the form of rail means that are reminiscent of rail road tracks. Rail means 704 are formed of pairs of parallel rail walls 708 spaced by distance l (about 0.1 to about 1.2mm) that is comparable to the distance d for the honeycomb means of FIG. 19, and extending at angle θ to the

flow direction F as was the case in the FIG. 19 embodiment. A plurality of cross or tie walls 708 extend between the rail walls. The wall height h_r of rail walls 706 is about 0.5 to 1.5mm, and the width w_r is about 0.4 to 1.2mm. The bases of the cross walls 708 are spaced by w_b from each other by about 0.5 to a few centimeters depending on the strain rate acceleration requirements, along the rail walls and are shorter than the rail walls 706, with a height e of about 0.1 to 1.5mm. The top width w_t of each cross wall is about 0.1 to 1.5mm. The sides of the cross walls 708 are inclined at an angle δ to the surface of rotor 702 by about 25 to 89°.

FIG. 22 is an embodiment of the invention which is applied to a valve gate in the hot runner system of an injection molding machine. The valve gate has a gate portion 802 with a passage 804 for receiving a polymer melt, and a stem or rod 808 having a valve face or end 818 that can be moved axially by known drive means schematically shown by arrow 812. To open and close the passage and admit or stop the flow of melt into a valve gate bushing 806, the stem 808 is moved in the direction of arrow 812. As with the other embodiments of the invention, stem 808 and/or the inner surface of bushing 806, carry rib means 810 and is continuously and/or oscillatingly rotated (Ω ; ω , α) about its axis. This produces a significant reduction in the viscosity and injection pressure of the melt. The viscosity is reduced with or without an increase in temperature, resulting in a shortened cycle time.

The rotation of the stem and even axial movement can be imparted to the stem 808 to advance the melt in bushing 806. The continuous rotational speed of stem 808 is a function of the diameter of the stem (for example about 6mm to 15mm) and is much higher (about 300 to 1700 rpm) than

for extrusion machines which use much larger diameter tubes(1 to 125 rpm).

Similar arrangements are possible for the extruder section, the injection section and various other possible parts of known equipment for processing polymer melts.

The complex rib means of the invention can be made using: electro-forming; electro-etching; laser soldering; laser forming; electro-discharge machining (EDM); investment casting or mechanical machining, to name a few. Suitable materials for the rotor and barrel are stainless steel, 4140 steel, high Rockwell bronze and the like. For the rotors the outer surface of a rod, e.g. 9mm in diameter and 23cm long can be formed with the rib means. For the barrel, an 18mm diameter tube, for example, can be cut along a plane parallel to the tube axis, the inner surfaces formed with the rib means and the semicylindrical or semiconical halves welded back together.

As general considerations, the shape, geometry, dimensions and spacing of the ribs, bumps and/or grooves of the rib means are selected and tailored depending on the type of polymer melt to be processed, following the criterion of my US Patent 5,885,495. These design criterion can be optimized for the specific melt, with the rheological parameters as taught by this patent especially with regard to G'/G^* , frequency and amplitude of strain created by the squeezing and relaxing of the melt by the rib means.

While specific embodiments of the invention have been shown and described in detail to illustrate the application of the principles of the invention, it will be understood that the invention may be embodied otherwise without departing from such principles.

WHAT IS CLAIMED IS:

1. An apparatus for reducing viscosity of a melt of molten moldable polymeric material by shear vibration under extensional flow to cause at least shear thinning, said apparatus comprising:
- 5 gap means defining first and second surfaces which are spaced from each other to form a treatment cavity gap between the surfaces, at least one of the surfaces being movable relative to the other surface to produce a shear deformation on a polymer melt in the gap;
- 10 drive means for moving at least one of the surfaces with respect to the other surface;
- rib means on at least one of said first and second surfaces, said rib means having a contour over which the melt moves, the contour being selected so that the rib means exerts a periodic variation of shear strain rate on the melt as the melt flows through the gap and as the at least one surface moves to produce local extensional acceleration and deceleration of the melt;
- 15 inlet means defining a gap inlet through which the melt passes into the gap;
- 20 outlet means defining a gap outlet through which the melt exits the gap; and
- a feeder connected to the gap inlet for preparing the melt and for feeding the melt to the gap inlet.
- 25
2. An apparatus according to claim 1, including drive means cooperating with the rib means for exerting a shear vibration of selected frequency and amplitude coupled with fatigue extensional flow on the melt in the gap.
- 30
3. An apparatus according to claim 1, including an



accumulator connected to the gap outlet for collecting the melt from the gap outlet, means for varying the amplitude of the shear vibration applied on the melt, means for continuously moving the melt through the gap from the inlet toward the outlet, means for continuously venting the treatment cavity gap to prevent the formation of bubbles or cavitation during the treatment, means for monitoring and controlling the temperature of the melt contained within the treatment cavity gap, means for monitoring and controlling the pressure of the melt within the gap and means for monitoring and controlling a torque exerted on the melt contained within the treatment cavity gap.

4. An apparatus according to claim 1, including a plurality of gap means connected to one another through gear pumps or screw pumps, said plural gap means including a first treatment station connected directly or through a gear pump and/or a static mixer to an extruder, an accumulator connected to the gap outlet for collecting the melt from the gap outlet and a last station connected to said at least one accumulator.

5. An apparatus according to claim 1, wherein the rib means comprise at least one of ribs and bumps and grooves.

6. An apparatus according to claim 3, wherein the means for continuously moving the melt comprises at least one of means for pushing and dragging and pumping the melt, the apparatus further including means for controllably varying a width of the gap.

7. An apparatus according to claim 1, wherein said gap is substantially flat, said first and second surfaces



* C L H S *

being flat.

8. An apparatus according to claim 1, wherein said gap is annular.

5 9. An apparatus according to claim 8, wherein at least one of said first and second surfaces is cylindrical.

10. An apparatus according to claim 8, wherein at least one of said first and second surfaces is conical.

10 11. An apparatus according to claim 10, including axial drive means for moving at least one of the surfaces axially to vary the width of the treatment cavity gap.

15 12. An apparatus according to claim 1, wherein said rib means comprises spaced apart rib walls which have a pitch selected so that a stress field caused by one rib wall in the melt overlaps a stress field caused by an adjacent rib wall in the melt.

13. An apparatus according to claim 12, wherein said rib walls extend one of radially, circumferentially and helically.

20 14. An apparatus according to claim 13, wherein said rib walls are continuous.

15. An apparatus according to claim 13, wherein said rib walls are interrupted.

16. An apparatus according to claim 13, wherein said rib walls vary in height along the at least one of said



first and second surfaces.

17. An apparatus according to claim 13, wherein said rib walls are V-shaped.

5 18. An apparatus according to claim 1, wherein the gap is annular and has a radius that changes at least once between the gap inlet and the gap outlet of the treatment cavity gap.

19. An apparatus according to claim 1, wherein said rib means comprises a plurality of spaced apart bumps.

10 20. An apparatus according to claim 19, wherein said bumps are polygonal.

21. An apparatus according to claim 19, wherein said bumps have a rounded contour selected to eliminate turbulence of the melt moving past said bumps.

15 22. An apparatus according to claim 1, wherein said gap is formed between a plurality of outer rotors which are rotatable against each other to form an inner space, and an inner rotor rotatable in the inner space, the inner rotor carrying said rib means.

20 23. An apparatus according to claim 1, wherein said gap is formed by an outer toroidal housing, an inner rotor and a series of oblong members connected to each other and to the rotor and rotatable in the toroidal housing.

25 24. An apparatus according to claim 1, wherein said gap comprise a pair of housings connected to each other by



a pump and a rotor rotatable in each housing defining the gap between each rotor and its respective housing.

25. An apparatus according to claim 1, wherein said gap is annular, said first surface being an outer surface of a rotor and said second surface being an inner surface of a barrel for receiving the rotor.

26. An apparatus according to claim 25, wherein the drive means comprises dual drive means for applying a continuous rotation to the rotor and for applying an oscillating rotation to the rotor for exerting the shear vibration of selected frequency and amplitude.

27. An apparatus according to claim 26, wherein the dual drive means comprises a differential drive means which is capable of independently controlling the continuous rotation and the frequency and amplitude of the oscillating rotation.

28. An apparatus according to claim 27, wherein the differential drive means comprises an epicyclic drive means.

29. An apparatus according to claim 28, including an extruder connected to the gap, the epicyclic drive being connected to the extruder for driving the extruder.

30. An apparatus according to claim 1, wherein the drive means cooperates with the rib means for exerting a shear vibration of selected frequency and amplitude coupled with fatigue extensional flow on the melt in the gap at least to the extent that the melt is subjected to



disentanglement.

31. An apparatus according to claim 30, including an accumulator connected to the gap outlet for collecting the melt from the gap outlet and disentanglement retaining means in the accumulator to moving the melt in the accumulator to maintain disentanglement in the melt.

32. An apparatus according to claim 1, wherein the first surface is the outer surface of an extruder rotor and the second surface is the inner surface of an extruder barrel for receiving the extruder rotor, the apparatus including temperature controlling means for heating and cooling melt in the gap and said drive means comprising motor means for rotating the rotor.

33. An apparatus according to claim 1, wherein the first surface is the outer surface of a valve gate rod and the second surface is the inner surface of a valve gate bushing for receiving the rod.

34. An apparatus according to claim 1, including an extruder having an outlet and forming at least part of said feeder, and a cross head die connected between the extruder outlet and the gap inlet for feeding melt from the extruder outlet to the gap inlet.

35. An apparatus according to claim 1, wherein said first surface is the outer surface of a rotor and carries said rib means, said second surface being an inner surface of a barrel for receiving the rotor, gap passing through a plurality of stations in said barrel for extruding the melt and for disentangling the melt.



36. An apparatus according to claim 35, wherein at least one of the stations contains a screw on said rotor for moving the melt along the gap, at least one of the stations contains means for pressurizing the melt in the gap, and at least one of the stations included the rib means for disentangling the melt.

37. An apparatus according to claim 1, including an injection molding machine connected to the gap for at least one of receiving treated melt from the gap outlet or supplying untreated melt to the gap inlet.

38. An apparatus according to claim 1, including recirculation means connected between a downstream part of the gap and an upstream part of the gap for recirculating at least some of the melt for additional viscosity reduction treatment in the gap.

39. An apparatus according to claim 1, including temperature control means for controlling the temperature along the gap.

40. An apparatus according to claim 39, wherein the temperature control means controls the temperature along the gap to maintain a selected elasticity of the melt.

41. An apparatus according to claim 39, wherein the temperature control means controls the temperature along the gap to maintain a selected temperature profile of the melt along the gap.

42. An apparatus according to claim 1, wherein the



rib means have a density distribution of ribs along at least one of the first and second surfaces, the apparatus including temperature control means for controlling the temperature along the gap, the temperature being controlled to change along the gap as a function of the rib density with higher temperature corresponding with higher rib density.

43. An apparatus for producing a disentangled polymer comprising:

means for raising the temperature of a polymer until it melts into a melt;

shear means for subjecting the melt to a disentanglement treatment by shear oscillation under extensional fatigue deformation until a state of entanglement between macromolecules of the polymer has been altered to a desired level as measured by a change in viscosity of the melt;

melt temperature control means for varying the temperature of the melt during the disentanglement treatment in a manner which induces lowering of the temperature of crystallization of the polymer by a dynamic effect of cooling rate on the onset of crystallization while avoiding crossing of such a transition

conveying means for conveying the melt to a station; and

processing means for subjecting the melt to a further operation for solidifying the polymer.

44. An apparatus as recited in claim 43, wherein said melt temperature control means enable cooling of the melt at a selected rate until a selected temperature is reached, while simultaneously adjusting the frequency of the shear



oscillation as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement efficiency.

45. An apparatus as recited in claim 43, wherein said
5 melt temperature control means enable cooling of the melt at a selected rate until a selected temperature is reached, while simultaneously adjusting the amplitude of the shear oscillation as a function of melt temperature in order to maintain a specific state of elasticity of the melt which
10 favors disentanglement efficiency and avoid melt fracture.

46. An apparatus as recited in claim 43, wherein said melt temperature control means enable cooling of the melt at a selected rate until a selected temperature is reached, while simultaneously adjusting the rate of shear flow to
15 which the melt is subjected as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement efficiency.

47. An apparatus as recited in claim 46, wherein the
20 shear means comprise rotating surfaces engaged with the melt, the rotation speed of the rotating surfaces in contact with the melt being programmed as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement
25 efficiency.

48. An apparatus as recited in claim 47, wherein at least one of the rotating surfaces has ribs, the number of ribs per turn on the rotating surface being selected to create a local extensional shear periodic deformation,



which varies with the temperature of the melt at that location.

5 49. An apparatus as recited in claim 48, wherein a density of said ribs on the surface at any given location of the disentanglement treatment decreases as temperature decreases, and, conversely, increases as temperature increases, according to the requirement to maintain at each location a specific state of elasticity of the melt which favors disentanglement.

10 50. An apparatus as recited in claim 43, wherein said melt temperature control means enable the melt temperature to vary controllably between two temperatures by alternatively cooling and heating the melt while disentanglement takes places and simultaneously adjusting
15 the frequency, amplitude and of shear oscillation as a function of melt temperature in order to maintain a specific state of elasticity of the melt which favors disentanglement efficiency.

20 51. An apparatus as recited in claim 43, wherein the melt temperature is profiled along the pathway of the flowing melt by controlling the flow rate of the cooling thermal fluid circulating in coolant runners or cooling jackets inside the die and/or barrel of the treatment cavity.

25 52. An apparatus as recited in claim 51, wherein the coolant thermal fluid can circulate, from the exit end of the treatment station towards the inlet, in a spirally shaped runner of decreasing cross-section, located further away from the melt cavity as it gets closer to the inlet



end of the station, resulting in a gradient of cooling capacity along the melt flow axis.

53. An apparatus as recited in claim 48, wherein melt temperature control means are those known in the art of mold temperature control, such as band heaters, heat pipes, heat sinks, air gaps, designed and activated to create the correct temperature difference between the inlet and the outlet of the treatment station to generate the desired cooling rate profile as the melt passes through.

54. An apparatus as recited in claim 43, wherein the same temperature profile can be repeated from station to station, maintaining the melt in an amorphous state as disentanglement proceeds.

55. An apparatus as recited in claim 43, wherein a different temperature profile is imposed within the treatment cavity of each station, to account for the changing properties of the disentangled melt as the treatment proceeds from station to the next maintaining the melt in an amorphous state as disentanglement proceeds.

