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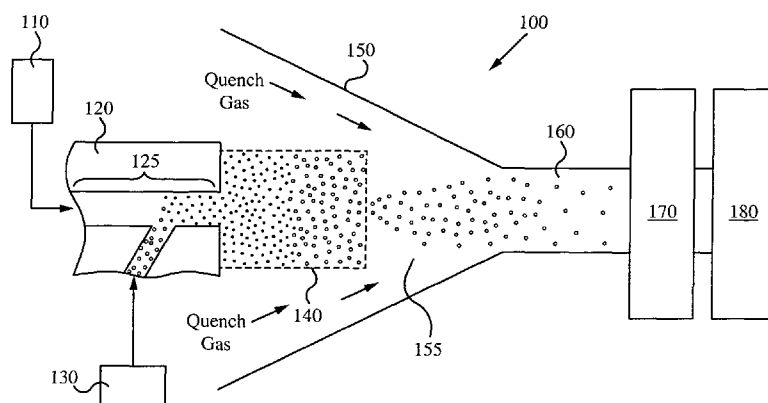


Fig. 1

(57) Abstract: A system comprising: a plasma production chamber configured to produce a plasma; a reaction chamber vaporize a precursor material with the plasma to form a reactive mixture; a quench chamber having a frusto-conical surface and a quench region formed within the quench chamber between an ejection port of the reaction chamber and a cooled mixture outlet, wherein the quench region configured to receive the reactive mixture from the ejection port, to cool the reactive mixture to form a cooled mixture, and to supply the cooled mixture to the cooled mixture outlet; and a conditioning fluid injection ring disposed at the ejection port and configured to flow a conditioning fluid directly into the reactive mixture as the reactive mixture flows through the ejection port, thereby disturbing the flow of the reactive mixture, creating turbulence within the quench region and cooling the reactive mixture to form a cooled mixture comprising condensed nanoparticles.

METHOD AND APPARATUS FOR MAKING UNIFORM AND ULTRASMALL NANOPARTICLES

CROSS-REFERENCE TO RELATED APPLICATIONS

The present application claims priority to co-pending U.S. Patent Application Serial No. 11/110,341, filed on April 19, 2005, entitled, "HIGH THROUGHPUT DISCOVERY OF MATERIALS THROUGH VAPOR PHASE SYNTHESIS" and to co-pending U.S. Provisional Application Serial Number 60/928,946, filed May 11, 2007, entitled "MATERIAL PRODUCTION SYSTEM AND METHOD," both of which are hereby incorporated by reference as if set forth herein.

FIELD OF THE INVENTION

The present invention relates to methods of and apparatus for quenching a reactive medium containing gas or vapor phase in order to produce uniform and ultrasmall nanoparticles.

BACKGROUND OF THE INVENTION

Gas or vapor phase particle production is an important technique for producing engineered nanoparticles. In a particle-producing reactor, basic product species are formed within extremely short time spans following ejection of a hot, reactive medium from an energy delivery zone. Following ejection from the delivery zone, further formation mechanisms determine the ultimate characteristics of the final product.

Although chemical reactions such as nucleation and surface growth within precursor materials occur largely during energy delivery, these formation mechanisms continue to be active in the first short moments following ejection. More prevalent in the post-ejection time period are bulk formation mechanisms such as coagulation and coalescence, which operate on already formed particles. Any proper conditioning of the hot, reactive medium following ejection from the energy delivery zone must account for these and other formation mechanisms to form a final product having desired characteristics. In some instances, maintaining a mixture at too high a temperature can lead to overly agglomerated particles in the final product.

In addition to particle formation, proper conditioning must account for post-formation processing of the product. Although particles, once formed, cool rapidly through radiative

heat loss, the residual gas in which they are entrained after formation cools much more slowly, and especially so when confined. Confinement is necessary to some degree in any controlled-environment processing system, and economic concerns usually dictate relatively small, confining controlled environments. Therefore, such systems must provide efficient mechanisms for cooling of the entire gas-particle product, yet also provide for efficient transport of the product to collection points within the system.

Transport of particles within a gas stream relies on the entrainment of the particles, which is largely a function of particle properties, e.g., mass, temperature, density, and interparticle reactivity, as well as gas properties, e.g., density, velocity, temperature, density, viscosity, and composite properties, such as particle-gas reactivity. Cooling of a gas by definition affects gas temperature, but also may easily lead to changes in other properties listed above, exclusive of mass.

What is needed in the art is a method of and an apparatus for balancing efficient cooling and transport of a gas-particle product, which requires careful optimization of process parameters.

SUMMARY OF THE INVENTION

In the embodiments of the present invention, features and methods are included to ensure extremely rapid quenching of reactive mixtures from vapor phase to solid phase, thereby producing uniform nanoparticles.

In one aspect of the present invention, a particle production system is provided. The system comprises a plasma production chamber configured to produce a plasma stream. A reaction chamber is fluidly coupled to the plasma production chamber and has an ejection port. The reaction chamber is configured to receive the plasma stream from the plasma production chamber, vaporize a precursor material with the plasma stream to form a reactive mixture stream comprising the vaporized precursor material entrained within plasma stream, and supply the reactive mixture stream to the ejection port. The system also comprises a quench chamber having a wide end, a narrow end, a frusto-conical surface that narrows as it extends from the wide end to the narrow end away from the ejection port of the reaction chamber, a cooled mixture outlet formed at the narrow end, and a quench region formed within the quench chamber between the ejection port and the cooled mixture outlet. The quench region is fluidly coupled to the ejection port of the reaction chamber and is configured to receive the reactive mixture stream from the ejection port of the reaction chamber, to cool

the reactive mixture stream to form a cooled mixture stream, and to supply the cooled mixture stream to the cooled mixture outlet. A conditioning fluid injection ring is disposed at the ejection port of the reaction chamber and configured to flow a conditioning fluid directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber, thereby disturbing the flow of the reactive mixture stream, creating turbulence within the quench region and cooling the reactive mixture stream to form a cooled mixture stream comprising condensed nanoparticles.

In another aspect of the present invention, a method of producing uniform particles is provided. The method comprises producing a plasma stream within a plasma production chamber, applying the plasma stream to a precursor material, and vaporizing the precursor material with the plasma stream within a reaction chamber, thereby forming a reactive mixture stream comprising the vaporized precursor material entrained within the plasma stream. The reaction chamber is fluidly coupled to the plasma production chamber and has an ejection port. The reactive mixture stream flows through the ejection port and into a quench region of a quench chamber. The quench chamber has a wide end, a narrow end, a frusto-conical surface that narrows as it extends from the wide end to the narrow end away from the ejection port of the reaction chamber, a cooled mixture outlet formed at the narrow end, and the quench region formed within the quench chamber between the ejection port and the cooled mixture outlet. A conditioning fluid flows through an injection ring disposed at the ejection port of the reaction chamber. The conditioning fluid flows directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber, thereby disturbing the flow of the reactive mixture stream and creating turbulence within the quench region. The reactive mixture stream is quenched within the quench region to form a cooled mixture stream comprising condensed nanoparticles. The cooled mixture stream flows through the cooled mixture outlet of the quench chamber.

In preferred embodiments, the quench chamber further comprises an annular supply portion disposed between the perimeter of the reaction chamber and the frusto-conical surface. The annular supply portion supplies a conditioning fluid into the quench region in an annular formation along a path different from the flow of the conditioning fluid through the conditioning fluid injection ring. In some embodiments, the annular supply portion comprises a plurality of supply ports disposed in an annular formation around the reaction chamber. In other embodiments, the annular supply portion comprises one continuous supply port disposed in an annular formation around the reaction chamber.

In preferred embodiments, the conditioning fluid injection ring flows the conditioning fluid directly into the reactive mixture stream at an angle substantially perpendicular to the flow of the reactive mixture stream.

In some embodiments the conditioning fluid is a gas. In some embodiments, the conditioning fluid is super-cooled gas or liquid gas, including, but not limited to, liquid nitrogen and liquid helium. The type and form of the conditioning fluid flowing through the injection ring can be the same or different from the conditioning fluid flowing through the annular supply portion.

It is contemplated that the plasma stream can be produced in a variety of ways. However, in a preferred embodiment, the plasma production chamber produces the plasma stream by energizing a working gas.

In some embodiments, the precursor material flows directly into the plasma production chamber via a precursor supply port on the plasma production chamber prior to its vaporization. Additionally or alternatively, the precursor material can flow directly into the reaction chamber via a precursor supply port on the reaction chamber prior to its vaporization.

In preferred embodiments, the reaction chamber comprises an insulating material, thereby forming an enthalpy maintenance region within the reaction chamber. In this respect, the enthalpy of the reactive mixture stream can be maintained at a predetermined threshold level for a period of time within the enthalpy maintenance region of the reaction chamber. Preferably, the reaction chamber comprises a ceramic material.

In preferred embodiments, a collection device is fluidly coupled to the cooled mixture outlet of the quench chamber via a conduit. The conduit preferably has substantially the same diameter as the cooled mixture outlet. The collection device receives the cooled mixture stream from the quench region and separates condensed particles from the cooled mixture stream. Ideally, these condensed particles are nanoparticles.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a schematic illustration of one embodiment of a system for producing uniform nanoparticles using a quench chamber in accordance with the principles of the present invention.

FIG. 2 is a graph illustrating average mixture temperature in relation to time and distance as material is introduced, vaporized and cooled within embodiments of the present invention.

FIG. 3 is a schematic illustration of one embodiment of a system for producing uniform nanoparticles using a quench gas in a turbulent quench chamber in accordance with principles of the present invention.

FIG. 4 is a schematic illustration of one embodiment of a system for producing uniform nanoparticles using a liquid conditioning fluid in a turbulent quench chamber in accordance with the principles of the present invention.

FIG. 5 is a flowchart illustrating one embodiment of a method of producing uniform nanoparticles in accordance with the principles of the present invention.

DETAILED DESCRIPTION OF THE INVENTION

The description below concerns several embodiments of the invention. The discussion references the illustrated preferred embodiment. However, the scope of the present invention is not limited to either the illustrated embodiment, nor is it limited to those discussed. To the contrary, the scope should be interpreted as broadly as possible based on the language of the Claims section of this document.

In the following description, numerous details and alternatives are set forth for purpose of explanation. However, one of ordinary skill in the art will realize that the invention can be practiced without the use of these specific details. In other instances, well-known structures and devices are shown in block diagram form in order not to obscure the description of the invention with unnecessary detail.

This disclosure refers to both particles and powders. These two terms are equivalent, except for the caveat that a singular “powder” refers to a collection of particles. The present invention may apply to a wide variety of powders and particles. Powders that fall within the scope of the present invention may include, but are not limited to, any of the following: (a) nano-structured powders(nano-powders), having an average grain size less than 250 nanometers and an aspect ratio between one and one million; (b) submicron powders, having an average grain size less than 1 micron and an aspect ratio between one and one million; (c) ultra-fine powders, having an average grain size less than 100 microns and an aspect ratio between one and one million; and (d) fine powders, having an average grain size less than 500 microns and an aspect ratio between one and one million.

The present invention will be readily understood by the following detailed description in conjunction with the accompanying drawings. To facilitate this description, like reference numerals designate like elements.

Preferably, the dimensions of the quench chamber of the present invention have the following general relationships: the diameter of the wide end is substantially greater than that of the ejection port of the reaction chamber, the diameters of the one or more conditioning fluid supply ports are substantially smaller than that of the injection port, the diameter of the narrow end of the constricting chamber is substantially smaller than the diameter of the wide end and substantially equal to the diameter of the ejection port. Additionally, because of the inclusion of the annulus of smaller ports, the diameter of the ejection port is necessarily smaller than that of the wide end of the quench chamber. More specifically, the constricting chamber preferably has a first dimension of approximately 12 inches, constricting to a second dimension of approximately two inches over a distance of 24 inches. Preferred aspect ratios, i.e., ratios of the first diameter to the distance between the first and second ends, range between one to three and one to two.

Flow of the conditioning fluid into the one or more supply ports is preferably caused by formation of a negative pressure differential with the cooled mixture outlet, which also aids in maintaining flow of the mixture through the chamber. This negative pressure differential is preferably formed by fluidly coupling a suction generator or vacuum formation system with the cooled mixture outlet. In alternative embodiments, active injection of conditioning fluid is contemplated, but this scheme has many disadvantages when compared to passively drawing conditioning fluid into the system by vacuum.

Because the present invention preferably uses a pressure differential to motivate flow of the conditioning fluid through the one or more supply ports, variation of the combined surface area of the one or more supply ports allows variation of the flow rate of conditioning fluid. As described below, in some configurations a difference in flow rates between the conditioning fluid sheath and the reactive gas vapor contributes to a conditioning effect of the fluid. Therefore, adjustment of the flow rate of the conditioning fluid permits optimization of the conditioning effect that a flow rate differential provides.

Within the present invention, many configurations of the smoothly varying constrictions of the chamber are contemplated. In the preferred embodiments, these constrictions will smoothly vary in such a way so as to accelerate fluid flow and provide a Venturi-effect pressure differential within the quench chamber. In general, the constriction shape is determined while accounting for several factors affecting the conditioning of the reactive gas-vapor. Two factors are of major concern. First, adequate space must be provided within the region proximal to the first end of the constricting chamber to

accommodate rapid expansion of the hot gas-vapor following its flowing into the chamber. Second, constriction of the chamber within the region proximal to the narrow end of the constricting chamber must not occur so rapidly that undue turbulence is introduced into the gas-vapor as it flows to the cooled mixture outlet. For any chamber having fixed length between the wide and narrow ends, these requirements present contradictory design concerns. However, the several embodiments of the present invention include designs that accommodate both concerns.

In a preferred configuration for constrictions within the quench chamber, a cone like (frusto-conical) surface constricts, at a constant rate or otherwise, from the wide end to the cooled mixture outlet at the narrow end. The quench chamber of the present invention preferably comprises a thin shell. The exterior of the quench chamber is can be cooled by a fluid cooling system, to dissipate heat absorbed into the body of the quench chamber from the gas particle mixture. As mentioned above, this heat will primarily be supplied to the quench chamber body in the form of radiation from the newly formed particles as the rapidly cool within the quench chamber. In order to avoid overheating of the chamber body, the fluid cooling system is preferably included.

The present invention considers a wide variety of gas phase particle production systems including combustion-based systems, plasma based systems, laser ablation systems and vapor deposition systems. The preferred systems take material inputs in a broad range of forms, including solid phase inputs, and provide product in high surface area forms, including nanopowders. In addition, the process controls preferably provide a fine degree over a plurality of reaction parameters, permitting fine gradients of product composition ratios to be produced.

A wide variety of material types and forms can be processed in preferable particle production reactors used in the present invention. Without prejudice, the present invention specifically considers the provision of materials in the following forms: solid, liquid and gas. An exemplary particle production system is a plasma powder production reactor, which is included within several of the exemplary embodiments discussed below. The plasma reactors considered within the present invention can have many means of energy delivery, including the following: DC coupling, capacitive coupling, inductive coupling, and resonant coupling. In general, vapor phase nanopowder production means are preferred. The embodiments of the present invention can use elements of nano-powder production systems similar to those disclosed in U.S. Pat. Application Serial No. 11/110,341, filed on April 19, 2005 and entitled,

“HIGH THROUGHPUT DISCOVERY OF MATERIALS THROUGH VAPOR PHASE SYNTHESIS”, which is currently published as U.S. Publication No. 2005-0233380-A. In such a nano-powder production system, working gas is supplied from a gas source to a plasma reactor. Within the plasma reactor, energy is delivered to the working gas, thereby creating a plasma. A variety of different means can be employed to deliver this energy, including, but not limited to, DC coupling, capacitive coupling, inductive coupling, and resonant coupling. One or more material dispensing devices introduce at least one material, preferably in powder form, into the plasma reactor. The combination within the plasma reactor of the plasma and the material(s) introduced by the material dispensing device(s) forms a highly reactive and energetic mixture, wherein the powder can be vaporized. This mixture of vaporized powder moves through the plasma reactor in the flow direction of the working gas.

Referring now to FIG. 1, the particle production system 100 comprises a plasma production unit 120 having plasma production chamber 125. The plasma production chamber 125 is preferably disposed within the plasma production unit 120, which can include several types of inputs for power, gas, and precursor materials. Inputs are provided to the plasma production chamber 125 by a variety of supply systems.

The plasma production unit 120 is configured to produce a plasma stream within the plasma production chamber 125. It is contemplated that the plasma stream can be produced in a variety of ways. However, in a preferred embodiment, a working gas flows from a working gas supply device 110 into the plasma production chamber 125, where energy is delivered to it, thereby forming the plasma stream. Preferably, an overall system controller provides control signals to the working gas supply device 110. Additionally, a power supply system (not shown) can also be coupled to the plasma production chamber 125. Preferably, the overall system controller provides control signals to the power supply system as well.

A reaction chamber 140 is fluidly coupled to the plasma production chamber 125 and configured to receive the plasma stream from the plasma production chamber 125. In a preferred embodiment, the reaction chamber 140 has a larger diameter than the plasma production chamber 125.

In some embodiments, the plasma production chamber 125 is fluidly coupled with a material supply device 130, thereby allowing precursor material, such as powder, from the material supply device 130 to be delivered directly into the plasma production chamber 125. Precursor material is stored within the material supply 130. A material supply conduit

preferably passes from the material supply device 130 to the plasma production chamber 125. The material supply device 130 preferably includes a controllable delivery system that provides material to the conduit. Preferably, the conduit enters through airtight seals and terminates within the plasma production unit 120 at a selected location. Furthermore, an overall system controller is preferably configured to supply control signals to the material supply device 130. Additionally or alternatively, the material supply device 130 can be fluidly coupled directly to the reaction chamber 140, thereby allowing precursor material from the material supply device 130 to be delivered directly into the reaction chamber 140.

In embodiments where the precursor material is delivered into the plasma production chamber 125, the plasma stream mixes with the precursor material, forming a reactive mixture stream. The coupling between the plasma production chamber 125 and the reaction chamber 140 permits delivery of the mixture stream from the plasma production unit 120 into the reaction chamber 140.

In some embodiments, portions of the reaction chamber 140 are constructed of an insulating material configured to maintain the enthalpy of a plasma stream within a portion thereof above a predetermined threshold. Preferably, maintaining the enthalpy of the plasma extends a resonance time of the plasma within the reaction chamber 140. In some embodiments, portions of the reaction chamber 140 are constructed of a material with high thermal durability. In these embodiments, the portions so constructed are configured to attain a high temperature during operation of the reaction chamber 140. In some embodiments, portions of the reaction chamber 140 are constructed of a ceramic material. Preferably, the material used is boron nitride.

Within the reaction chamber 140, the plasma stream preferably vaporizes the precursor material, thereby forming a reactive mixture stream comprising the vaporized precursor material entrained within the plasma stream. In some embodiments, this vaporization of precursor material can begin in the plasma production chamber 125 if the precursor material is introduced into the plasma production chamber 125.

Preferably, reaction chamber 140 is shaped and the operational parameters of the system 100 are controlled so that as the mixture stream enters the reaction chamber 140, it is maintained above an enthalpy threshold. This maintenance takes place within an enthalpy maintenance region somewhere within the reaction chamber 140. Preferably, the average enthalpy of the mixture stream falls as it moves away from the enthalpy maintenance region with its minimum enthalpy (within the reaction chamber 140) coming at the ejection port to

the quench region 155. In some embodiments, the resonance time of the mixture within the chamber 140 is above a threshold length of time controlled by an overall system controller.

Furthermore, in some embodiments of the present invention, the reaction chamber 140 is constructed and the operational parameters of the apparatus 100 are chosen so that the vaporized precursor material begins to condense within the mixture stream while it is within the reaction chamber 140. In some other embodiments, construction and operational parameters of the system 100 are chosen so that the vaporized precursor material begins to condense within the mixture stream while it is within the quench region 155.

A quench chamber 150 is fluidly coupled to the reaction chamber 140 through an ejection port at the end of the reaction chamber 140. The ejection port is configured to supply the reactive mixture stream from the reaction chamber 140 into a quench region 155 of the quench chamber 150. In a preferred embodiment, the reaction chamber 150 has a frusto-conical shape, narrowing as it extends away from the ejection port and towards a cooled mixture outlet. Although the figures show quench chamber 150 substantially open at the wide end, preferably the wide end of the quench chamber 150 is substantially closed except for fluid ports through which a fluid can be received. The cooled mixture outlet is disposed at the narrow end of the quench chamber 150 that preferably leads into a cooling conduit 160. The quench region 155 is formed within the quench chamber 150 between the ejection port of the reaction chamber 140 and the cooled mixture outlet. In a preferred embodiment, an annular supply portion is formed between the perimeter of the reaction chamber 140 and the quench chamber 150. This annular supply portion can comprise a plurality of supply inlets or one continuous supply inlet disposed in an annular formation around the ejection port of the reaction chamber 140. The annular supply portion is configured to supply a conditioning fluid, preferably from a conditioning fluid supply device, to the quench region 155. In a preferred embodiment, these supply inlets are channels of adjustable size that directly couple the conditioning fluid supply to the quench region 155, yet permit for controlled flow of the conditioning fluid to the quench region 155. In FIG. 1, conditioning gas is supplied roughly along the vectors marked "Quench Gas." In a preferred embodiment, the conditioning fluid is supplied through airtight inlets and outlets to the quench region 155 (preferably from a dedicated supply of conditioning fluid), where it preferably mixes with and cools the reactive mixture stream from the reaction chamber 140.

Motive fluid flow within the system can be motivated by a suction generator 180, such as a motive vacuum pump, pulling a negative pressure on a cooling conduit 160 that is fluidly

coupled to the cooled mixture outlet of the quench chamber 150, thereby motivating mass flow through the outlet of the quench region 155. However, the flow rate of the motive fluid into the quench region 155 is preferably controlled by the overall control system.

The cooling conduit 160 receives the particle and gas mixture from the quench region 155. Preferably, the mixture is pulled into the cooling conduit 160 by the suction generator 180. However, in some embodiments, a motive pump or other system within a sampling or collection device 170 provides some motive force to pull the mixture. Of course, to some extent, pressure provided by the plasma production chamber 125 and the conditioning fluid supply can motivate the flow of the mixture into the cooling conduit 160.

In some embodiments, the cooling conduit 160 is equipped with an active cooling system. In some embodiments, a conditioning fluid, such as argon, is supplied into the gas input couplings of the cooling conduit 160. In some of these embodiments, the conditioning fluid is a cooling and entraining gas. In some of these embodiments, the conditioning fluid is a passivating gas.

The cooling conduit 160 fluidly connects the quench region 155 with a sampling or collection device 170. The conduit 160 is preferably coupled to the quench region 155 through airtight means. The collection device 170 is preferably positioned between the cooling conduit 160 and the suction generator 180. The collection device 170 is configured to receive the cooled mixture via the cooling conduit 160, sample or collect material, such as condensed particles, having appropriate characteristics from the mixture, and permit remains of the mixture to flow to the suction generator 180, which is fluidly coupled through a conduit. Furthermore, the collection device 170 can take multiple samples, at selected times, and can sample discontinuously, which allows for sampling from a gas-particle streams whose composition may vary from time to time without contamination from previous product.

It is contemplated that the collection device 170 can be configured in a variety of ways. In one embodiment, the collection device 170 comprises a sampling structure, at least one filled aperture formed in the sampling structure, and at least one unfilled aperture formed in the sampling structure. Each filled aperture is configured to collect particles from the mixture stream, such as by using a filter. The sampling structure is configured to be adjusted between a pass-through configuration and a collection configuration. The pass-through configuration comprises an unfilled aperture being fluidly aligned with a conduit, such as the cooling conduit 280, thereby allowing the unfilled aperture to receive the mixture stream

from the conduit and the mixture stream to flow through the sampling structure without substantially altering the particle content of the mixture stream. The collection configuration comprises a filled aperture being fluidly aligned with the conduit, thereby allowing the filled aperture to receive the mixture stream and collect particles while the mixture stream is being flown through the filled aperture.

It is contemplated that the sampling structure can be adjusted between the pass-through configuration and the collection configuration in a variety of ways. In one embodiment, the sampling structure is a disk-shaped structure including an annular array of apertures, wherein the annular array comprises a plurality of the filled apertures and a plurality of the unfilled apertures. The sampling structure is rotatably mounted to a base, wherein rotational movement of the sampling structure results in the adjustment of the sampling structure between the pass-through configuration and the collection configuration. In another embodiment, the sampling structure is a rectangular-shaped structure including a linear array of apertures, wherein the linear array comprises a plurality of the filled apertures and a plurality of the unfilled apertures. The sampling structure is slideably mounted to a base, wherein sliding of the sampling structure results in the adjustment of the sampling structure between the pass-through configuration and the collection configuration.

As mentioned above, the collection device 170 preferably permits the suction generator 180 to provide a motive force therethrough. However, in some embodiments, the collection device 170 provides additional motive force. In some embodiments, a collection device 170 supplants the motive force provided by the suction generator 180 and provides a substitute motive force to the cooling conduit 160.

The overall control system (not shown) sends signals to the working gas supply 110 and power supply to set operational parameters. Parameters for the working gas supply 110 determine the rate at which the working gas feeds into the plasma production chamber 125. Power supply parameters determine the voltage and amperage at which power is supplied to the plasma production chamber 125. In combination, these parameters determine the characteristics of the plasma produced within the plasma production chamber 125. Furthermore, the material supply device 130 provides a metered stream of material through the material supply conduit to the conduit's terminus location within the plasma production chamber 125. This exposes the material to plasma within the chamber. The rate at which material is provided into the chamber 125 preferably is determined by the overall control system. This rate, and other system parameters, determines characteristics of the mixture

stream formed within the plasma production chamber 125. Furthermore, although the material supply device 130 is described as providing only a single material into the plasma production chamber 125 at a single location, in some embodiments of the present invention, the material supply device 130 supplies a plurality of materials into the plasma production chamber 125 and/or the reaction chamber 140 at one or more locations.

In the system 100, the mass flow rate of material through the system is controlled to permit effective quenching at achievable conditioning fluid flow rates. Preferably, this rate is controlled via the mass flow rate of material into the plasma production chamber 125. Specifically, the material supply device 130 and the control system are configured to control a mass flow rate delivery of the precursor material into the plasma stream to achieve a rate that permits cooling of the mixture stream to one quarter of the melting point of the material extremely rapidly. Preferably, this flow rate is selected with reference to achievable conditioning fluid flow rates within the quench region 155 and with reference to achievable turbulence within the quench region 155.

While the configuration of system 100 provides an improvement in terms of quench rate over the prior art, the quench rate can be further improved. FIGS. 3 and 4 illustrate systems similar to system 100, but with additional features that result in an improved quench rate.

FIG. 3 illustrates one embodiment of a particle production system 300. In the system 300, the reactive mixture flows from the reactor chamber 140 into the quench region 155, such as in FIG. 1. While the mixture flows into the quench region 155, conditioning fluid, labeled "Quench Gas" is supplied into the quench region 155 via an annular supply portion similar to the annular supply portion discussed above. A portion of the conditioning fluid is diverted or otherwise supplied to an injection ring disposed at the end of the reaction chamber 140. This injection ring is configured to flow the quench gas directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber 140, thereby disturbing the flow of the reactive mixture stream, creating turbulence within the quench region and cooling the reactive mixture stream at the earliest point possible as the mixture leaves the reaction chamber.

It is contemplated that the injection ring can be configured in a variety of ways. In preferred embodiments, the injection ring flows the conditioning fluid directly into the reactive mixture stream at an angle substantially perpendicular to the flow of the reactive mixture stream. However, it is contemplated that other injection angles are within the scope

of the present invention as well. Furthermore, in a preferred embodiment, the injection ring comprises a plurality of injection ports, such as nozzle structures 352 and 354, disposed in an annular configuration around flow of the reactive mixture. The injection ring is configured to induce a high degree of turbulence within the conditioning fluid and the reactive mixture, and ultimately the quench region 155.

As the reactive mixture moves out of the reaction chamber 140, it expands and mixes with the conditioning fluid. Parameters related to conditioning fluid supply are controlled to permit the nozzles 352 and 354 to produce a high degree of turbulence and promote mixing with the reactive mixture. This turbulence can depend on many parameters. Preferably, one or more of these parameters are adjustable to control the level of turbulence. These factors include, but are not limited to, the flow rates of the conditioning fluid and any modification to the flow path of the fluid.

After entering the quench region 155, particle formation mechanisms are active. The degree to which the particles agglomerate depends on the rate of cooling. The cooling rate depends on the turbulence of the flow within the quench region. Preferably, the system is adjusted to form a highly turbulent flow, and to form very dispersed particles. For example, in preferred embodiments, the turbidity of the flow within the quench region is such that the flow has a Reynolds Number of at least 1000. Preferably, the turbulence is controlled to achieve a rate of cooling of the mixture stream that moves the mixture stream temperature to one quarter of the melting point of the material within a very short time after the reactive mixture exits the reaction chamber 140.

Following injection into the quench region, cooling, and particle formation, the mixture flows from the quench chamber 150 into the cooling conduit 160. Suction generated by an external device, such as the suction generator previously discussed, preferably moves the cooled mixture from the quench region 155 into the conduit 160. The cooled mixture can flow to a collection or sampling device, such as described above with respect to FIG. 1.

In FIG. 3, the conditioning fluid supplied to the quench region through the injection ring disposed at the ejection port of the reaction chamber 140 and through the annular supply portion disposed between the perimeter of the reaction chamber 140 and the quench chamber 150 is a gas. In one embodiment, the gas supplied is argon. However, other gases can be used as well.

It is contemplated that in addition to a gas being used as the conditioning fluid, a super-cooled gas, or liquid gas, can be used as the conditioning fluid. Such conditioning

fluids include, but are not limited to, liquid nitrogen and liquid helium. In FIG. 4, system 400 provides a conditioning fluid to the quench region 155 in the form of a liquid gas, which is labeled as "Quench Liquid." In a preferred embodiment, the quench liquid flows through an injection ring directly into the reactive mixture as the reactive mixture leaves the reaction chamber 140, similar to the quench gas described above with respect to FIG. 3. However, it is contemplated that any combination or configuration of the features illustrated in FIGS. 2-4 are within the scope of the present invention. For example, a quench gas can be supplied through the annular supply portion, while a quench liquid is supplied through the injection ring. Alternatively, the quench liquid can be supplied through the annular supply portion and into the quench region 155 without the use of the injection ring.

Preferably the temperature of the liquid and the flow rate thereof are configured to achieve a rate of cooling of the mixture stream that moves the mixture stream temperature to one quarter of the melting point of the material within an extremely short time after the reactive mixture leaves the reaction chamber 140. The hot reactive mixture absorbs the moisture from the quench liquid, thus resulting in an increased quench rate above that achieved by only using a quench gas. Preferably the temperature and conditioning liquid flow rate are selected with reference to desired mass flow rates within the system.

FIG. 5 is a flowchart illustrating one embodiment of a method 500 of producing uniform nanoparticles in accordance with the principles of the present invention. As would be appreciated by those of ordinary skill in the art, the protocols, processes, and procedures described herein may be repeated continuously or as often as necessary to satisfy the needs described herein. Additionally, although the steps of method 500 are shown in a specific order, certain steps may occur simultaneously or in a different order than is illustrated. Accordingly, the method steps of the present invention should not be limited to any particular order unless either explicitly or implicitly stated in the claims.

At step 510, a plasma stream is produced within a plasma production chamber. It is contemplated that the plasma stream can be produced in a variety of ways. However, in a preferred embodiment, the plasma production chamber produces the plasma stream by energizing a working gas that flows through the chamber.

At step 520, the plasma stream is applied to a precursor material, thereby vaporizing the precursor material. This application of the plasma stream to the precursor material can take place in the plasma production chamber and/or in a reaction chamber fluidly coupled to the plasma production chamber. Either way, the plasma stream flows into the reaction

chamber and a reactive mixture stream is formed within the reaction chamber. The reactive mixture stream preferably comprises the vaporized material entrained within the plasma stream.

At step 530, the reactive mixture stream flows through the ejection port of the reaction chamber and into a quench region of a quench chamber. The quench chamber has a wide end, a narrow end, a frusto-conical surface that narrows as it extends from the wide end to the narrow end away from the ejection port of the reaction chamber, a cooled mixture outlet formed at the narrow end, and the quench region formed within the quench chamber between the ejection port and the cooled mixture outlet.

At step 540, a conditioning fluid flows through an injection ring disposed at the ejection port of the reaction chamber. The conditioning fluid flows directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber, thereby disturbing the flow of the reactive mixture stream and creating turbulence within the quench region. As discussed above, the conditioning fluid is preferably a gas or a liquid gas. Additionally, or alternatively, the conditioning fluid can be supplied through the annular supply portion disposed between the perimeter of the reaction chamber and the quench chamber.

At step 550, the reactive mixture stream is rapidly quenched within the quench region to form a cooled mixture stream. The cooled mixture stream preferably comprises condensed nanoparticles.

At step 560, the cooled mixture stream flows through the cooled mixture outlet of the quench chamber to a collection device., preferably via a conduit. In a preferred embodiment, the conduit has substantially the same diameter as the cooled mixture outlet.

At step 570, the collection device separates condensed particles from the cooled mixture stream as the cooled mixture stream flows through the collection device. In a preferred embodiment, the collection device separates the condensed particles from the cooled mixture stream by using one or more filters.

FIG. 2 illustrates a graph 200 showing the intended effect of features of embodiments of the present invention on quench rates in particle production systems. The graph 200 charts exemplary variations in gas/material or mixture temperature with time (position in the apparatus) as they enter a plasma production chamber, such as chamber 125, vaporize or become plasma, mix, travel into a reaction chamber, such as reaction chamber 140, begin to form particles, and enter the quench region, such as quench region 155. Relationships

between position within the apparatus and position on the graph are roughly illustrated by vertical correlation between FIGS. 1 and 2.

The vertical axis of the graph charts mixture temperature. The oval legends denote phases of matter for the material being operated on by the apparatus. Vapor is on top, below that liquid, and below that solid. The dashed lines indicate critical temperatures relative to the material concerned. The 'bp' line denotes boiling point, the 'mp' line denoted melting point. The 'mp/4' line denotes one quarter of the material melting point.

Within the plasma production chamber 125, the temperature rapidly increases, causing sublimation of the material from solid phase to vapor phase. As the mixture moves into the reactor chamber, 140 the temperature and enthalpy of the mixture remain substantially constant, staying within the vapor phase of the material. However, as the mixture moves to the end of the reaction chamber 140, the temperature decreases, reaching a minimum (within the reaction chamber) at the downstream edge of the reaction chamber 140.

Once ejected, the mixture enters the quench region 155, where it expands and cools rapidly. One or more of highly turbulent flow, sufficiently low mass flow rate, and liquid quenching with super cooled gas forms sufficient mixing of the mixture with conditioning fluid to cool the mixture smoothly and rapidly through 'bop', liquid phase, 'mp' and part of solid phase to 'mp/4' within a short enough time to avoid undesired agglomeration and promote the production of uniform nanoparticles. The supply of conditioning fluid through the annular supply portion results in the quench rate illustrated as line 210. While this quench rate is a substantial improvement over the prior art, the use of the injection ring and/or the liquid gas results in an even faster quench rate illustrated as line 210'.

During resonance within the reaction chamber and quench region, particles form. Because the mixture is cooled rapidly, there is a short time period during which agglomeration occurs. As the mixture of particles and hot gas continues to mix with the conditioning fluid, the mixture of gas and particles moves out of the narrow end into the conduit. Overall, the quench period within a highly turbulent quench region and/or a quench region fed by a super-cooled gas conditioning liquid as in some embodiments of the present invention is much shorter than with standard quenching. Eventually the conditioning fluid and the mixture reach thermal equilibrium, preferably at room temperature.

Thus, features of the embodiments of the present invention decrease the period during which particles formed can agglomerate with one another. Ultimately, this decrease in potential agglomeration produces particles of more uniform size, and in some instances

produces smaller-sized particles. Both of these features lead to particles with increased dispersibility and increased ratio of surface area to volume.

The present invention has been described in terms of specific embodiments incorporating details to facilitate the understanding of the principles of construction and operation of the invention. As such, references herein to specific embodiments and details thereof are not intended to limit the scope of the claims appended hereto. It will be apparent to those skilled in the art that modifications can be made to the embodiments chosen for illustration without departing from the spirit and scope of the invention.

CLAIMS:

What is claimed is:

1. A particle production system comprising:
 - a plasma production chamber configured to produce a plasma stream;
 - a reaction chamber fluidly coupled to the plasma production chamber and having an ejection port, wherein the reaction chamber is configured to receive the plasma stream from the plasma production chamber, vaporize a precursor material with the plasma stream to form a reactive mixture stream comprising the vaporized precursor material entrained within plasma stream, and supply the reactive mixture stream to the ejection port;
 - a quench chamber having a wide end, a narrow end, a frusto-conical surface that narrows as it extends from the wide end to the narrow end away from the ejection port of the reaction chamber, a cooled mixture outlet formed at the narrow end, and a quench region formed within the quench chamber between the ejection port and the cooled mixture outlet, wherein the quench region is fluidly coupled to the ejection port of the reaction chamber and is configured to receive the reactive mixture stream from the ejection port of the reaction chamber, to cool the reactive mixture stream to form a cooled mixture stream, and to supply the cooled mixture stream to the cooled mixture outlet; and
 - a conditioning fluid injection ring disposed at the ejection port of the reaction chamber and configured to flow a conditioning fluid directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber, thereby disturbing the flow of the reactive mixture stream, creating turbulence within the quench region and cooling the reactive mixture stream to form a cooled mixture stream comprising condensed nanoparticles.
2. The system of Claim 1, further comprising an annular supply portion disposed between the perimeter of the reaction chamber and the frusto-conical surface, the annular supply portion fluidly coupled to the quench region and configured to supply a conditioning fluid into the quench region in an annular formation along a path different from the flow of the conditioning fluid through the conditioning fluid injection ring.

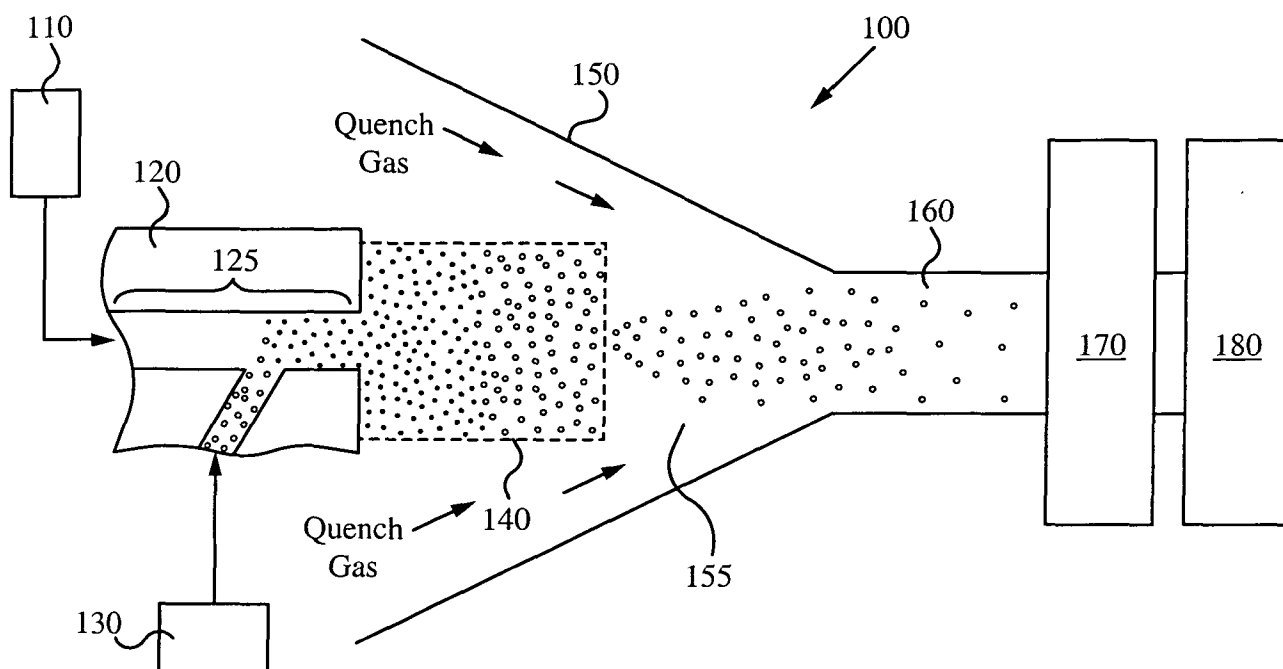
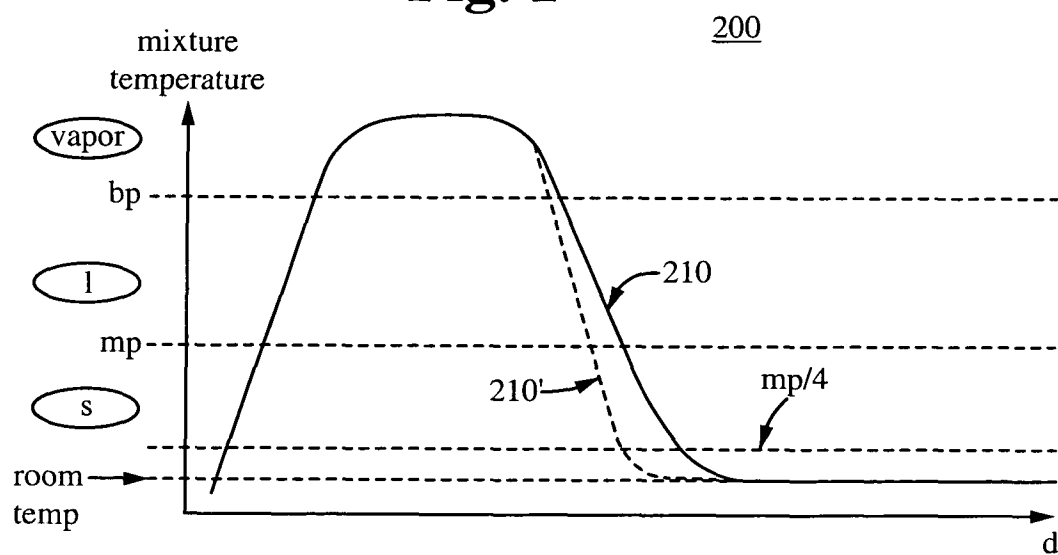
3. The system of Claim 2, wherein the annular supply portion comprises a plurality of supply ports disposed in an annular formation around the reaction chamber.
4. The system of Claim 2, wherein the annular supply portion comprises one continuous supply port disposed in an annular formation around the reaction chamber.
5. The system of Claim 1, wherein the conditioning fluid injection ring is configured to flow the conditioning fluid directly into the reactive mixture stream at an angle substantially perpendicular to the flow of the reactive mixture stream.
6. The system of Claim 1, wherein the plasma production chamber is configured to form the plasma stream by energizing a working gas.
7. The system of Claim 1, wherein the plasma production chamber comprises a precursor supply port configured to supply the precursor material directly into the plasma production chamber prior to its vaporization.
8. The system of Claim 1, wherein the reaction chamber comprises a precursor supply port configured to supply the precursor material directly into the reaction chamber prior to its vaporization.
9. The system of Claim 1, wherein the reaction chamber is formed from insulating material.
10. The system of Claim 9, wherein the reaction chamber comprises a ceramic material.
11. The system of Claim 1, further comprising a collection device fluidly coupled to the cooled mixture outlet of the quench chamber via a conduit, the conduit having substantially the same diameter as the cooled mixture outlet, wherein the collection device is configured to receive the cooled mixture stream from the quench region and separate condensed particles from the cooled mixture stream.

12. A method of producing uniform particles, the method comprising:
- producing a plasma stream within a plasma production chamber;
 - applying the plasma stream to a precursor material;
 - vaporizing the precursor material with the plasma stream within a reaction chamber, thereby forming a reactive mixture stream comprising the vaporized precursor material entrained within the plasma stream, the reaction chamber being fluidly coupled to the plasma production chamber and having an ejection port;
 - flowing the reactive mixture stream through the ejection port and into a quench region of a quench chamber, the quench chamber having a wide end, a narrow end, a frusto-conical surface that narrows as it extends from the wide end to the narrow end away from the ejection port of the reaction chamber, a cooled mixture outlet formed at the narrow end, and the quench region formed within the quench chamber between the ejection port and the cooled mixture outlet;
 - flowing a conditioning fluid through an injection ring disposed at the ejection port of the reaction chamber, wherein the conditioning fluid flows directly into the reactive mixture stream as the reactive mixture stream flows through the ejection port of the reaction chamber, thereby disturbing the flow of the reactive mixture stream and creating turbulence within the quench region; and
 - quenching the reactive mixture stream within the quench region to form a cooled mixture stream comprising condensed nanoparticles; and
 - flowing the cooled mixture stream through the cooled mixture outlet of the quench chamber.
13. The method of Claim 12, wherein the quench chamber further comprises an annular supply portion disposed between the perimeter of the reaction chamber and the frusto-conical surface, and wherein the method further comprises the step of the annular supply portion supplying a conditioning fluid into the quench region in an annular formation along a path different from the flow of the conditioning fluid through the conditioning fluid injection ring.
14. The method of Claim 13, wherein the annular supply portion comprises a plurality of supply ports disposed in an annular formation around the reaction chamber.

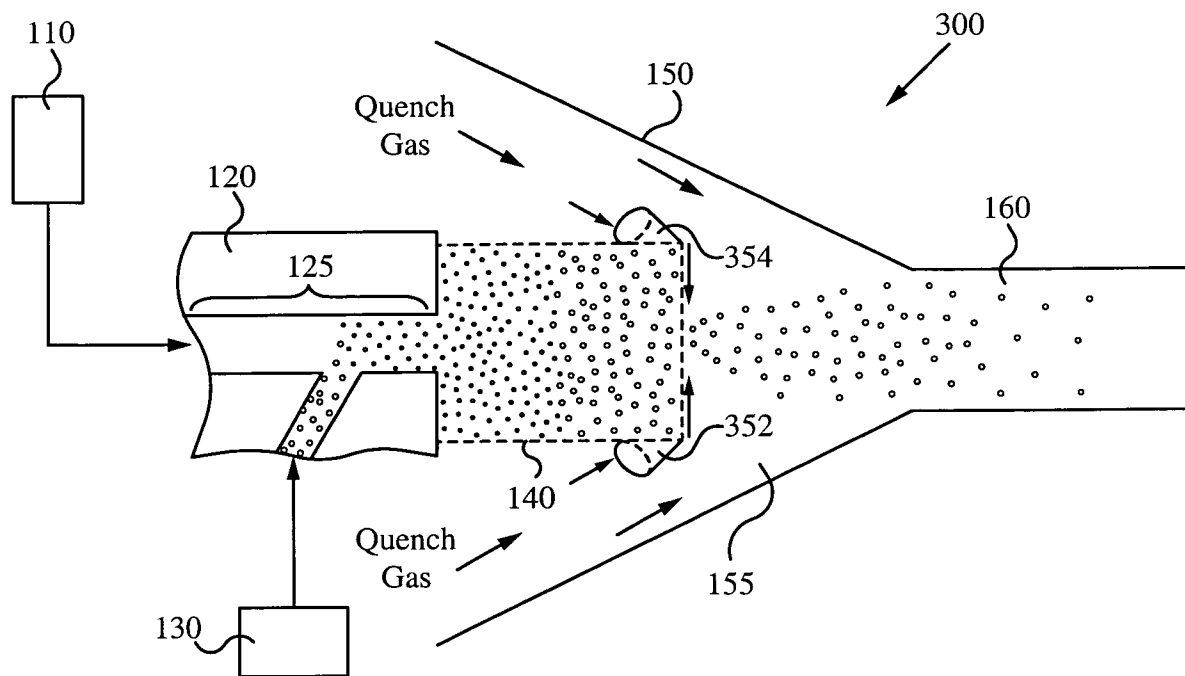
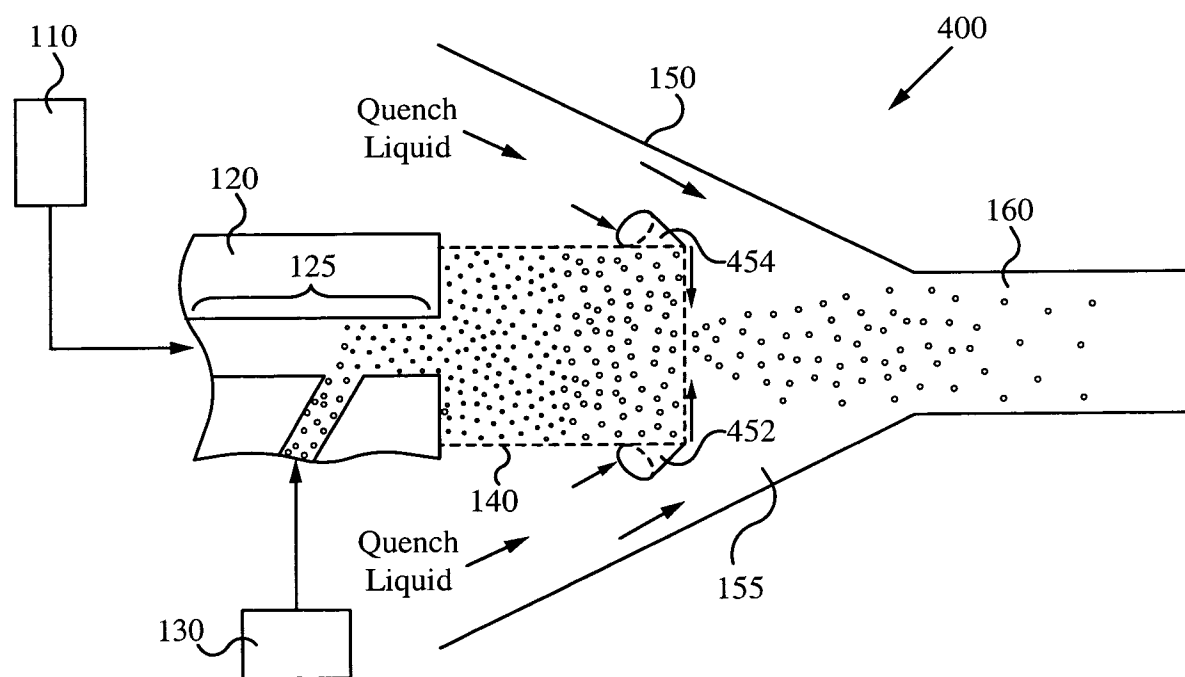
15. The method of Claim 13, wherein the annular supply portion comprises one continuous supply port disposed in an annular formation around the reaction chamber.
16. The method of Claim 12, wherein the conditioning fluid injection ring flows the conditioning fluid directly into the reactive mixture stream at an angle substantially perpendicular to the flow of the reactive mixture stream.
17. The method of Claim 12, wherein the conditioning fluid is a super-cooled gas.
18. The method of Claim 12, wherein the conditioning fluid is liquid nitrogen.
19. The method of Claim 12, wherein the conditioning fluid is liquid helium.
20. The method of Claim 12, wherein the plasma production chamber produces the plasma stream by energizing a working gas.
21. The method of Claim 12, further comprising the step of flowing the precursor material directly into the plasma production chamber via a precursor supply port on the plasma production chamber prior to its vaporization.
22. The method of Claim 12, further comprising the step of flowing the precursor material directly into the reaction chamber via a precursor supply port on the reaction chamber prior to its vaporization.
23. The method of Claim 12, wherein the reaction chamber comprises an insulating material, thereby forming an enthalpy maintenance region within the reaction chamber, and wherein the method further comprises the step of maintaining the enthalpy of the reactive mixture stream for a period of time within the enthalpy maintenance region of the reaction chamber.
24. The method of Claim 23, wherein the reaction chamber comprises a ceramic material.

25. The method of Claim 12, wherein a collection device is fluidly coupled to the cooled mixture outlet of the quench chamber via a conduit, the conduit having substantially the same diameter as the cooled mixture outlet, and wherein the method further comprises the steps of:
- the collection device receiving the cooled mixture stream from the quench region; and
 - the collection device separating condensed particles from the cooled mixture stream.

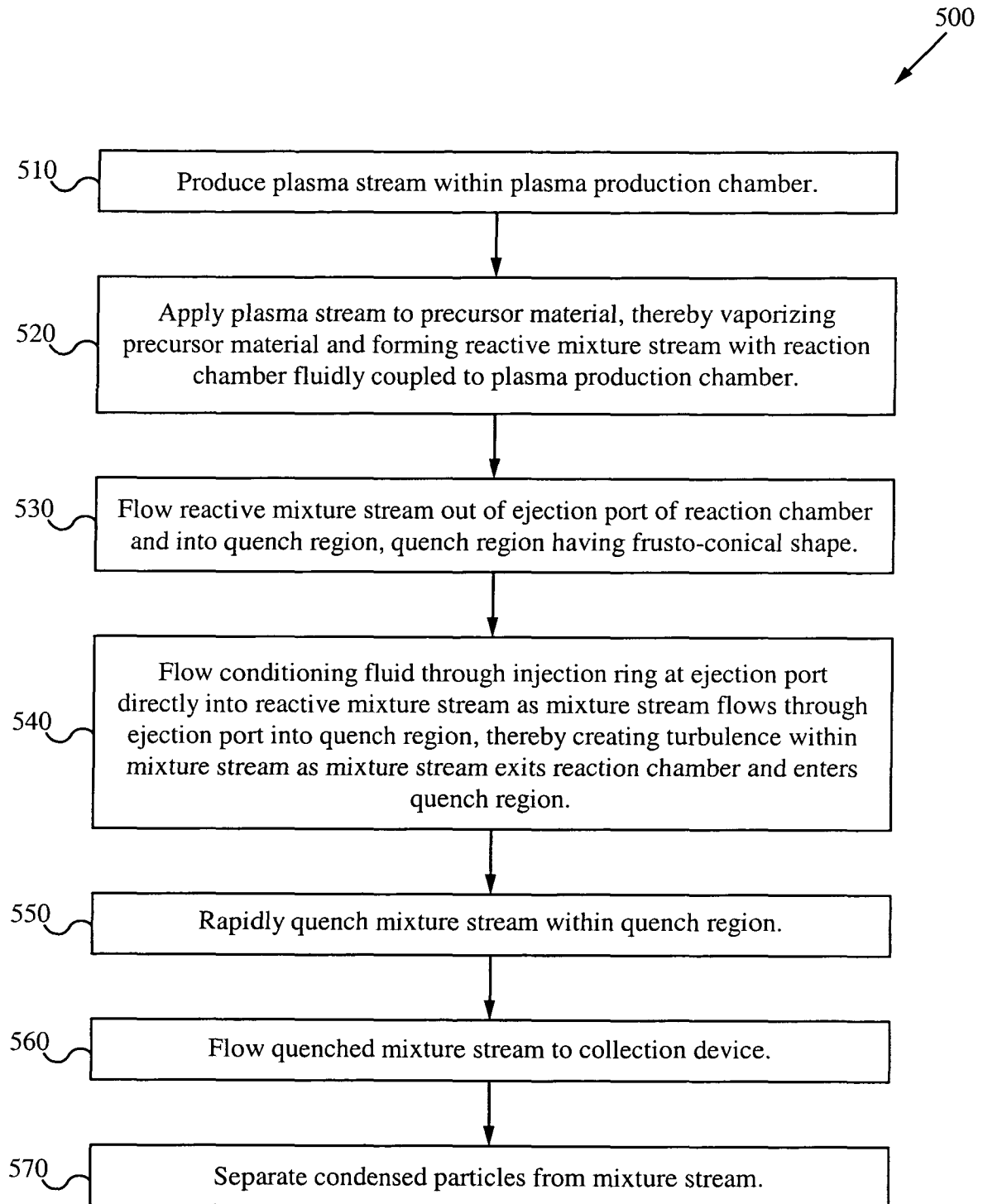
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**Fig. 1****Fig. 2**

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**Fig. 3****Fig. 4**

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**Fig. 5**

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US 08/06003

A. CLASSIFICATION OF SUBJECT MATTER

IPC(8) - H01L 21/20 (2008.04)

USPC - 438/584

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC(8) - H01L 21/20 (2008.04)

USPC - 438/584

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

IPC(8) - H01L 21/20; C23C 16/10 (2008.04)

USPC - 438/584; 118/723R; 977/844,880

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

PubWEST(USPT,PGPB,EPAB,JPAB); Google

Search Terms Used:

uniform nanoparticles, plasma chamber, frusto-conical, supercool nitrogen, quench plasma, ceramic plasma reactor

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 2003/0223546 A1 (McGREGOR et al.) 04 December 2003 (04.12.2003), para [0018], fig. 1,2	1-25
Y	WO 2006/079213 A1 (BOULOS et al.) 03 August 2006 (03.08.2006), para [0034-0038], fig. 11	1-25
Y	US 2006/0051505 A1 (KORTSHAGEN et al.) 09 March 2006 (09.03.2006), para [0025, 0904]	9, 10, 23 and 24
Y	US 2007/0063364 A1 (HSIAO et al.) 22 March 2007 (22.03.2007), para [0028]	11 and 25
Y	US 2005/0000950 A1 (SCHRODER et al.) 06 January 2005 (06.01.2005), para [0005]	17-19
Y	US 7,166,198 B2 (VAN DER WALT et al.) 23 January 2007 (23.01.2007), col 7, ln 51-52	23 and 24

☐ Further documents are listed in the continuation of Box C.

* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search

07 August 2008 (07.08.2008)

Date of mailing of the international search report

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