Hani Henein · Volker Uhlenwinkel
Udo Fritsching *Editors*

Metal Sprays and Spray Deposition

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Preface

This compendium of knowledge in metal spray and spray deposition processes summarizes the technical and scientific state-of-the-art metal treatment and manufacturing via droplet and spray processes to form near-net-shaped components. It is hoped that established production areas and fields such as spray forming (SF), and also emerging fields like additive manufacturing (AM), may be inspired in an economical and ecological sense in developments of new technical approaches.

We are grateful for the contributions of all authors of the chapters in this book. The sharing of their knowledge and experiences in this field is acknowledged. Without them, this book would not have been possible. This book would not have seen the light of day without the assistance of some colleagues and students. In particular, the assistance of Paul Gronau, Evan Chow, and Sining Li was invaluable.

We hope you will enjoy reading and find this book of value for years to come.

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Bremen, Germany — Volker Uhlenwinkel
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Contents

Chapter 1
Introduction

Diran Apelian, Hani Henein, and Udo Fritsching

Near net shape processing or net shape processing has been and continues to be a pursuit of the Materials Science and Engineering community. Net shape processing is a type of manufacturing that produces a product that does not require any further treatment. Near net shape processing is similar except that minor treatment of the product is considered necessary. There are many motivations for developing such routes. Processing metallic and metallic based composite products are capital intensive operations; thus any process that generates a product closer to its final form using less processing steps will require less capital equipment and result in reduced capital investments. Concomitant with the reduction in process steps is the requirement that superior product performance and properties be achieved while reducing the waste generated in processing the part. It is desired to process complex shaped parts with significant throughput and the ability to apply automation in processing. This increases the reliability of products while achieving high volume production. An additional advantage of these processing routes is that they are considered to be green processes.

In the last decade we have seen much interest in green processing or in processes that we term as being sustainable. It may be useful to lay out some basic principles for green processing as it relates to spray forming or droplet consolidation processes. In the most simplistic sense, processes that reduce waste are sustainable processes. Metaphorically speaking, the most sustainable organism we have to

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learn from is nature. Nature has been around for a long time and much can be learned from nature. Nature is cyclic and there is no waste; furthermore, nature uses a few elements (unlike the scenario we are witnessing in the twenty-first century where most of the elements of the periodic table are being utilized). Waste can be further classified as *production waste* or *post-consumer waste*. The former has much to do with green processing, whereas the latter has to do with manufacturing products that can be disassembled (and repaired and reused) as well as creating value out of scrap. A good example of production waste is red mud during Al production; for every kg of Al produced, 3 kg of red mud is also produced. An example of post-consumer waste is the fact that only ~50% of beverage cans in North America are recycled. On another front, and one that has huge promise is the work that is being carried out by Melt Cognition LLC in developing a mini mill for Al production where the starting material is 100% mixed scrap Al, which is intelligently sorted into the various Al alloys (based on chemistry—XRF and LIBS technologies), and subsequently melted and cast [1]. The process is called AIM (Al Integrated Mini-mill) and creates value out of scrap. In other words, one upcycles rather than downcycles in an effort to create value and attain sustainable processing or green processing. The processing of AIM is in principle similar to that of spray forming. Eliminating processing steps such as homogenization and hot working while reducing machining scrap. For these attributes, spray forming is a green process.

To add to the economic and green processing benefits derived from near or near net shape processing, there are microstructural benefits one can obtain by near net shape processing. From a scalar perspective, processes that reduce the diffusion distance between heterogeneities in the final structure are most desirable, as they enable one to attain better properties and performance in the final component. Microstructural refinement of the end product has been and continues to be a goal in metal processing. For centuries mankind has been making components via casting where the solidification rates are small, and the diffusion distances between heterogeneities are large. Castings are heat treated (call it post-solidification processing) specifically to attain microstructural homogeneity and refinement. This is a good example of how spray forming through a droplet consolidation mechanism circumvents all of the post processing as each droplet has a starting refined microstructure. Spray forming can be thought of assembling these individual droplets into a whole. There are numerous near net shape and net shape processing routes developed and continue to be proposed and tested. These include processes such as strip casting, high pressure die casting, powder metallurgy routes, plasma deposition, cold spray, and melt infiltration.

To establish some context, one can describe metal processing through the phase changes that accompany the process. Figure 1.1 illustrates conceptually a classification based on the phases that are being processed. For example, in sand casting, the cavity is filled with a liquid which undergoes solidification. Depending on the size of the casting, local solidification times vary, but in general the solidification rates are quite slow in the order of several degrees per minute. Whereas in deformation processing, there is no phase change, and all of the processing takes

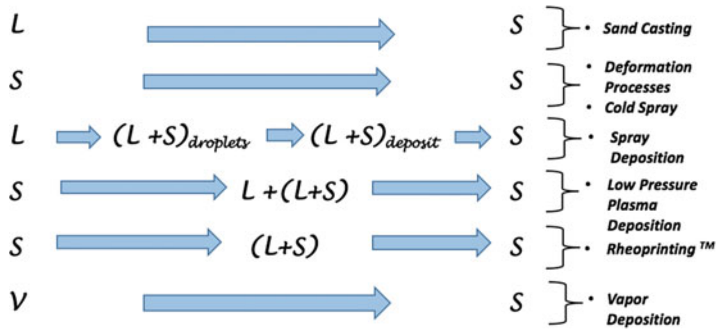

Fig. 1.1 Classification of metal processing based on phase changes in the process

place in the solid state. Cold spray processing and forging are quite dissimilar processes, but what they have in common is that the processing occurs in the solid state. In forging it is the kinetic energy imparted on the workpiece, whereas in cold spray it is the velocity of the powder particles that aid the impact of the powders onto the substrate to form an integral bond. In low pressure plasma deposition, powders are injected into the nozzle and upon exiting one will have a mixture of liquid droplets as well as semi-solid (L + S) droplets impacting the substrate. More recently, with the advent of Additive Manufacturing (AM), we have seen much interest in powder production as most of the AM processes use powder as the starting material. The Rheoprinting™ technology, developed at the Metal Processing Institute, circumvents the use of powders in AM, as the starting material is an ingot and what emerges from the nozzle is a thixotropic metal in the mushy zone [2]. It is AM via control and manipulation of the viscosity of the thixotropic alloy through the nozzle of the printer. Lastly, processing can be done in the vapour phase such as in CVD, PVD, and other related processes.

An important commercial near net shape process is the spray deposition/forming process. It has been nearly half a century since the first publication by Singer [3, 4] described a new method by which atomised droplets are deposited onto a substrate before they are fully solidified. One practice of this process is described here for illustration. The principle behind this process is that molten metal is poured through a nozzle of controlled diameter into a chamber containing inert gas with gas jets directed at the stream of molten metal. There are numerous approaches to the atomization process for molten metals. The liquid melt stream is broken up into droplets. These droplets flow with the atomizing gas exchanging and losing heat while partly or completely solidifying. In spray forming, most droplets trajectories while semi-solid are interrupted by falling onto a substrate. The remaining liquid in the droplets together with some larger liquid droplets aid in filling the pores between deposited droplets. Most droplets in the deposit likely remain separated even by a tiny oxide layer. This mechanism has been described by analogy to a series of balloons filled with ice and water landing onto the substrate by researchers at the University of Alberta. This model would explain why precipitates in a spray formed part are fine in size and homogeneously distributed throughout the deposit despite the very low solidification rate of the deposited ingot and the coarse grain size. Thus, a part in spray forming is built layer by layer as more droplets land on the substrate and subsequently the deposit. Process description and modelling, material evolution models and theories, as well as the current state of the art with various alloy systems are clearly discussed in this book.

There have been great efforts in academia, government and industry to develop the spray forming process and generate unique cost effective products with it. In 1985 the very first Osprey unit was installed in North America at Drexel University (Apelian, Lawley, Doherty); many doctoral theses were published and much of the fundamentals of spray deposition were established [5–16]. In the nearly 50 years of practice of this process, there have been numerous efforts to present article reviews on the status of research and development on the process [17–23]. In addition, in the mid 1990s Lavernia and Wu [21] published a book describing the then state of the art in spray forming. The fundamentals of Spray Forming have also been collected in the Chemical Engineering basic Ullmanns Encyclopedia [22]. As the potential of numerical modelling and simulation has further and further increased in these days, a summarize on Spray Simulation: Modeling and Numerical Simulation of Spray forming Metals has been published in 2004 [23]. Research papers and publications on Spray Forming continue to grow as new knowledge and approaches to practice the process continue to be invented, developed and practiced [24, 25]. Figure 1.2 provides a view of the number of publications that are published as a function of the year of publication. The search was carried out on Web of Science covering the years since 1970, Singer's first publication on the process. Figure 1.3 shows the citations on Spray Forming as a function of year also since 1970. It is clear from both of these plots that activity in this field remains strong. In fact the area of Spray Forming has an H index of 37 indicating that it remains an important area of activity in the field of materials science and engineering. A review of the papers that

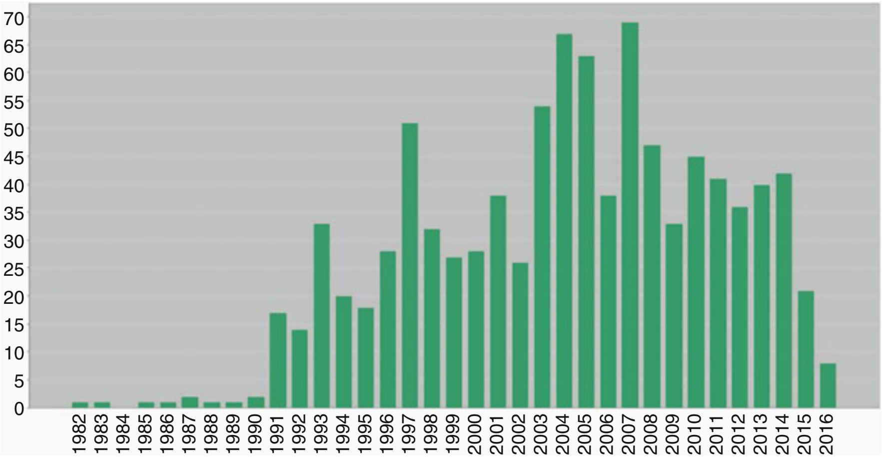

Fig. 1.2 Number of publications on Spray Forming as a function of publication year. Source: Web of Science, June 29, 2016

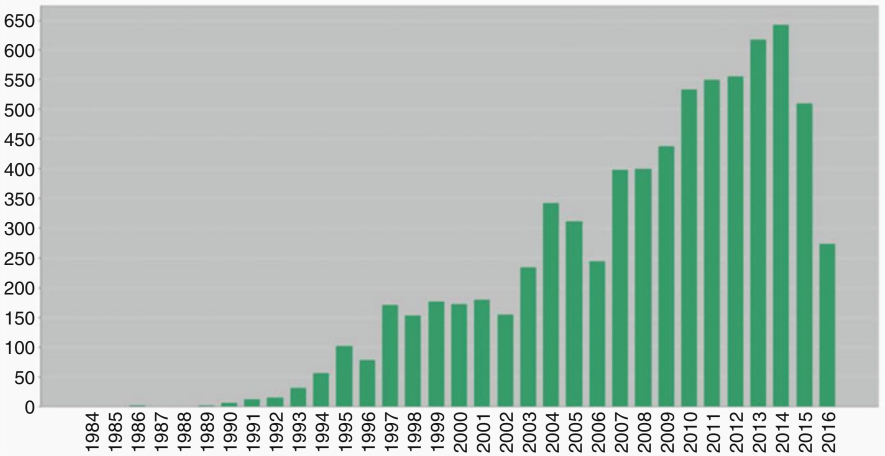

Fig. 1.3 Citations on Spray Forming as a function of year. Source: Web of Science, June 29, 2016

Table 1.1 Frequency of publications on Spray Forming by country

Peoples Republic of China	27%
United States of America	18%
Germany	13%
England	8%
India	7%
Brazil	7%
South Korea	4%
Taiwan	3%
France	2%
Spain	2%
Japan	1%
Wales	1%
Canada	1%
Denmark	1%
Others	6%

Source: Web of Science, June 29, 2016

continue to be published in this field clearly shows that interest in Spray Forming is indeed a worldwide activity. Table 1.1 lists the countries reported to have published on Spray Forming as well as the frequency of such publications.

There have been intense research activities over the past two decades by Bauckhage and the research group of the Collaborative Research Center on Spray Forming at the University of Bremen in Germany. A series of conferences as “International Conference on Spray Deposition and Melt Atomization—SDMA” took place five times between 2000 and 2013 at the University of Bremen [26], sometimes in cooperation with the “International Conference on Spray Forming”.

The meetings always involved more than 100 international participants from academia and industry that intensively presented and discussed actual developments and achievements in the atomization of melts and spray forming of metals. Contributions included papers investigating spray diagnostics as well as numerical modelling and simulation of these processes, process analysis and control, materials properties with special emphasis on new lightweight materials and superalloys and also within conventional copper, steel and aluminium alloys, as well as contributions on processing and downstream treatment of spray formed or spray coated products.

With the emergence of 3D printing with metals or Additive Manufacturing, those that have been active in the field of Spray Forming recognize many fundamental and practical issues that are common to those for Spray Forming. It was felt that an overview on the topic would be timely in order to provide for the community one reference point on the latest developments in Spray Forming. Hence this book is organized into areas of fundamentals in the early chapters. These start with a description of the fundamentals of single fluid atomization. Several techniques are described though not all of them may be easily conducive to Spray Forming. In these techniques, the use of mechanical or electrical energy is used to break up a melt stream. This provides more controlled melt stream break-up conditions, reduced gas consumption, narrower droplet size distribution, and lower overspray powders, while yielding rapidly solidified structures in the spray formed parts. In Chaps. 3, 4, 5 and 7 two fluid atomization is described in terms of its fundamentals along with the fundamentals of Spray Forming with respect to impingement of droplets onto a substrate or deposit as well as transport phenomena governing the process. Diagnostics measurements taken in-situ during atomization is presented in Chap. 6. Chapter 9 described the techniques developed and used to characterize spray formed products. Chapters 10–16 address the state of the art for different alloy systems including aluminium, titanium, and copper and their alloys, steels and superalloys are also addressed. Finally some of the applications of Spray Forming to novel materials such a bulk metallic glasses are outlined.

It is hoped that this compendium of knowledge will spur further activity in this area as well as inspire practical and high throughput approaches to new developments in Additive Manufacturing.

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Chapter 2
Single Fluid Atomization Fundamentals

Abdoul-Aziz Bogno, Hani Henein, Volker Uhlenwinkel, and Eric Gärtner

2.1 Introduction

Atomization is simply defined as the breakup of a liquid stream into droplets. It can be achieved in many ways including spraying through a nozzle, pouring on to a rotating disc, etc. Atomization practice and research usually involve materials processing in their liquid state either at or near room temperature (oil-based liquids, paint spraying, aerosol sprays, etc.) or at high temperature (metal melts). Most of the literature describing atomization mechanisms pertains to two fluid atomization in which a second fluid is applied to break up a melt stream into droplets. Two fluid atomization techniques for molten metals are described in Chap. 3.

In view of the requirement of high liquid/solidification cooling rate, high undercooling but also controllable droplets size, shape and solidification microstructures, single fluid atomization (SFA) has established itself as the atomization technique of choice. It is a containerless solidification technique [1] which consists in the transformation of a bulk liquid into a spray of droplets that generally fall and solidify rapidly by losing heat to a surrounding gas of choice (N_2, Ar or He are commonly used). The bulk liquid is produced by heating a material above its melting point and the droplets, generally of narrow size distribution, are either collected after complete solidification as powders or are deposited in a semi-solid

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state onto a substrate to form a strip or as a spray coating. Controllable droplet shape and size and narrow size spectra are important for many technical applications including spray coating and printing. It has been reported that spherical solar cells produced by dropping method for photovoltaic power generation give better performance due to an improved overall microstructures as compared to the bulk Si solar cells made from Si ingots [2, 3].

In recent years, SFA has become the subject of attention of many researchers through the development of several techniques such as the drop-on-demand [4–6], the pulsated orifice ejection method (POEM) [2], the jet-splitting method [7], the flat-fan and pressure swirl methods [8], the centrifugal atomization [9], the ultrasonic atomization [10–12] and the Impulse Atomization (IA) technique [13].

In this chapter, SFA fundamentals will be discussed based on metals atomization. The melt stream break-up mechanism will be emphasized and the controlling parameters of the mass/heat flux and size distribution will be analyzed based on IA, a typical SFA technique developed at the Advanced Materials and Processes Laboratory (AMPL) of the University of Alberta. Finally, microstructures characterization of droplets obtained by IA will be discussed and a new quantitative method to estimate the primary and secondary nucleation undercoolings during rapid solidification of droplets will be presented.

2.2 Droplet Formation

2.2.1 Mechanism of Stream Breakup

Atomization is defined as the break-up of a liquid stream into droplets. Understanding this breakup mechanism is therefore very important in order to optimize the design and improve the performance of SFA systems. As described by Henein [13] and Yuan [14] the breakup mechanism is controlled by two forces: the potential force induced by the head of liquid above the orifice and an external force (disturbance) applied to the melt. Indeed, a liquid sitting over an orifice will flow through it to form a stream when the gravity force is significantly greater than the surface tension and drag force. While the melt head induces an inertial force to drain the melt through the orifices, effective melt flow requires (1) viscous dissipations through the orifices and (2) overcoming of the force induced by the surface tension of the melt which acts opposite to the direction of flow, as the stream exits the orifice [15]. Thus, the melt requires an external force not only to push it through the orifices but also to act as a disturbance that triggers the stream break-up especially for a small orifice size.

Figure 2.1 shows a schematic of a liquid ligament emanating from an orifice. When the liquid ligament emerges from the orifice as a continuous body of cylindrical form (as shown by the schematic) there occurs a competition on the surface of the ligament between the cohesive and disruptive forces. This competition gives rise to

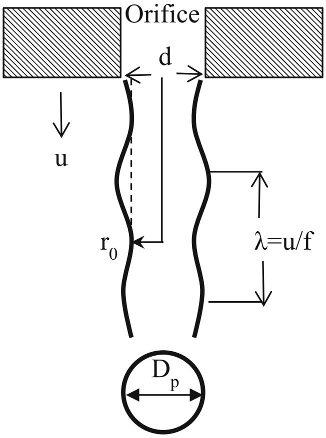

Fig. 2.1 Liquid ligament with periodic perturbations falling with a velocity u from a nozzle orifice of radius r_0 upon an applied impulse of frequency f

oscillations and perturbations of a wavelength λ, which under induced effects of an external disturbance (e.g. impulses) are amplified and the liquid body breaks up into spherical droplets.

According to Rayleigh instability [16], the minimum theoretical wavelength of a perturbation required to break up a liquid ligament of length L is:

$$\lambda = 2\pi r_0 \tag{2.1}$$

where r_0 is the radius of the ligament, which is assumed to be equal to the orifice radius. Therefore, in terms of frequency and velocity, the maximum frequency f_{max} that must be applied for the applied perturbation to induce break-up of the liquid ligament is:

$$f_{max} = \frac{u}{2\pi r_0} \tag{2.2}$$

where u is the velocity of the liquid ligament and f the frequency of the applied perturbation. Thus, if for a given ligament of length L and circumference C, the condition $^{L}/_{C} < 1$ is fulfilled then the ligament is expected to form only one droplet, otherwise if $^{L}/_{C} > 1$, the ligament will break up into several droplets depending on its L and C which are determined by the force applied to push the liquid through the nozzle orifice and the nozzle orifice size [14]. The force used to generate the melt stream varies by process type. For example, in the jet-splitting method, the flat-fan and pressure swirl method and the ultrasonic atomization, the pressure is applied by a gas overpressure being applied to the surface of a melt in a crucible. For the POEM and the drop-on-demand methods, this force is being applied using a piezo-electric crystal. For centrifugal atomization the orifice at the bottom of a crucible holding the melt is sufficiently large as to allow the melt to flow out freely under gravity. Finally, for IA the force that is used can be a combination of both gas overpressure and mechanical pressure.

The resulting droplet diameter D_p can be calculated using Eq. (2.3) where it is assumed that droplets generated from the applied perturbation have the same volume as that within one wavelength of the liquid stream [17].

$$\frac{1}{6}\pi D_p^3 = \pi r_0^2 \lambda \tag{2.3}$$

Expressing Eq. (2.3) in terms of f and u:

$$\frac{1}{6}\pi D_p^3 = \pi r_0^2 \left(\frac{u}{f}\right) \tag{2.4}$$

And, rearranging in terms of D_p:

$$D_p = \left(\frac{6 r_0^2 u}{f}\right)^{1/3} \tag{2.5}$$

While the frequency f is operator dependent, u is determined by dividing the liquid flowrate Q by the cross sectional area of the orifice πr_0^2 [17] yielding Eq. (2.6).

$$u = \frac{Q}{\pi r_0^2} \tag{2.6}$$

2.2.2 *Boundary Between Stream and Dripping Formation*

The quantification of the transition from free stream flow to dripping is important in modeling melt atomization. Based on a model initially used to determine physical properties of liquids, the liquid flow from the bottom of a crucible is given by Eq. (2.7) [17]. The flowrate Q is related to the surface tension (σ), the potential force induced by the liquid head (h), the gravity acceleration constant (g), the liquid density (ρ) as well as the discharge coefficient, C_D across the orifice of cross-sectional radius r_o [15].

$$Q = C_D \pi r_0^2 \sqrt{2g\left(h - \frac{\sigma}{\rho g r_0}\right)} \tag{2.7}$$

Combining Eqs. (2.6) and (2.7), the liquid velocity can be expressed in terms of potential and surface forces [Eq. (2.8)].

$$u^2 = C_D^2 2g\left(h - \frac{\sigma}{\rho g r_0}\right) \tag{2.8}$$

Equation (2.8) may be written in dimensionless form by introducing the Froude number, *Fr,* and Bond number, B_o, as follows.

$$\frac{F_r}{C_D^2} + \frac{1}{B_0} = 1 \tag{2.9}$$

Where

$$F_r = \frac{u^2}{2gh} \tag{2.10}$$

And

$$B_0 = \frac{\rho g r_0 h}{\sigma} \tag{2.11}$$

When h decreases i.e. the level of the melt becomes low (degree of vorticity negligible) the velocity of the stream decreases and the stream will approach laminar flow conditions characterized by a low Reynolds number:

$$Re = \frac{2\rho u r_0}{\mu} \tag{2.12}$$

At low Re it has been shown that there is a linear relationship between C_D and Re as seen in Eq. (2.13), so that a decrease of u (therefore Re) consequent to a decrease in h will lead to a decrease of C_D [15].

$$C_D = a + bRe \tag{2.13}$$

where a and b are respectively the intercept and the slope of the C_D vs Re regression line for low Re values. The melt will stop flowing as a stream when its velocity tends to zero.

When $u \rightarrow 0$, $B_0 \rightarrow 1$ so that the lower critical head height h_{min} for stream formation is expressed by Eq. (2.14) as follows:

$$h_{min} \geq \frac{\sigma}{\rho g r_0} \tag{2.14}$$

When $h < h_{min}$ flow from the orifice would continue by dripping, forming droplets, until h further decreases and the fluid surface tension keeps it at the orifice. Atomization under these conditions typically produces large droplets at low production rates. In fact, under dripping conditions, the formulations for droplet formation following the stream breakup mechanism described above do not apply. The lower critical velocity for stream formation can be expressed in terms of the Weber number, We as follows [18]:

$$We = \frac{2\rho r_0 u^2}{\sigma} > 4 \tag{2.15}$$

2.2.3 Stream Breakup Regimes

Figure 2.2 shows a schematic description of droplet formation by different break-up mechanisms depending on the velocity of the liquid stream.

When u is large enough consequent to an increase of Q and therefore h, the kinetic energy overcomes the surface tension and a continuous liquid stream forms. Droplet formation in this case occurs by Rayleigh instability [16] in the so called "Rayleigh regime" as described earlier. At larger liquid velocity consequent to a larger head height, the relative velocity between the stream and the atomization atmosphere becomes remarkable inducing aerodynamic effects that accelerate the break-up process and shortening of the ligaments lengths is observed. This regime is referred to as "the 1st Wind Break-up" [16]. At a sufficiently high velocity, the static pressure induced on the surface may result in the "whiplash mode" [16, 19]. This mechanism is referred to as "2nd Wind Break-up" [16]. If the velocity of the stream is higher still, "atomization" is observed [16, 19]. Indeed, while viscosity has a damping effect on the growth of disturbances on the liquid surface and surface tension tends to pull the liquid together, aerodynamic forces tend to promote the growth of disturbances. The overall contributions of these forces can be characterized by the Ohnesorge (Oh) non-dimensional number, ratio of viscous forces over inertia and surface tension forces [Eq. (2.16)].

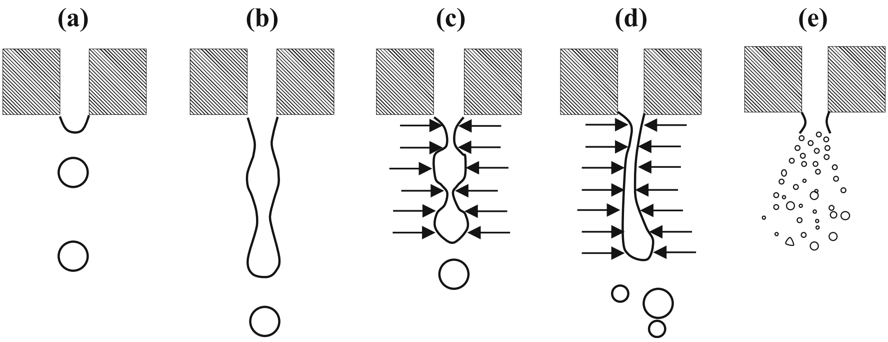

Fig. 2.2 Schematic description of different droplet formation mechanisms (**a**) drip off, (**b**) Rayleigh regime, (**c**) 1st Wind Breakup, (**d**) 2nd Wind Breakup and and (**e**) Atomization

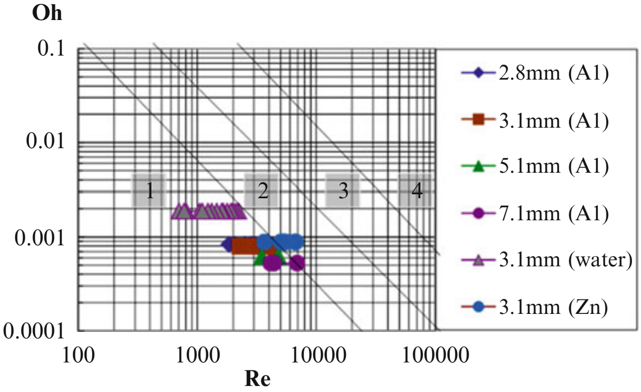

Fig. 2.3 Breakup regimes including examples of stream of Al, water and zinc at various sizes (**1**) Rayleigh regime, (**2**) first wind-induced, (**3**) second wind induced and (**4**) atomization breakup (from [21] with permission)

$$Oh = \frac{\mu}{\sqrt{2\rho\sigma r_0}} \tag{2.16}$$

Or,

$$Oh = \frac{\sqrt{We}}{Re} \tag{2.17}$$

Reitz [20] developed the original work of Ohnesorge [21] to propose a chart of break-up regimes by plotting *Oh* vs *Re* as shown in Fig. 2.3. This was obtained using oils and water. A large *Oh* indicates that viscous effects are more dominant in the liquid. Example data for aluminum, water and zinc processed with various orifice sizes are also shown in the figure.

The data show that water as well as metals melts (Al, Zn) processed at the same orifice sizes remain within the Rayleigh and first wind induced breakup regimes. With a higher flowrate Q (higher *Re*) as one moves from left to right on Fig. 2.3. and the droplet size becomes smaller.

2.2.4 Spheroidization

Spheroidization which is the last stage of droplet formation depends on τ the ratio of times taken to dissipate internal mechanical energy or simply the spherodization time (t_{SP}) to the time taken to dissipate thermal energies t_{th} [Eq. (2.18)].

$$\tau = \frac{t_{SP}}{t_{th}} \tag{2.18}$$

If $\tau < 1$, under the influence of surface tension, the droplets have the time to spheroidize before solidification is completed. And if $\tau > 1$, i.e. solidification completes before spheroidization, the droplets will have the shape of ligaments [22].

Indeed, t_{SP} is the time for any oscillations in a spherical shape to be damped by internal viscous stresses characterized by the viscosity coefficient μ. It can be shown that this time should be proportional to $\left(\rho D_p^2/\mu\right)$ [23] reported that a 200 μm kerosene droplets moving at 10 ms^{-1} in an air-atomized spray retained sphericity after approximately 5 ms, which leads to the following expression of t_{SP}:

$$t_{SP} = \frac{0.1\rho D_p^2}{\mu} \tag{2.19}$$

The selection of gas atmosphere can affect the resultant shape of a droplet. When using inert gas, Eqs. (2.18) and (2.19) would result in droplets spheroidizing. However, in the presence of oxygen in the gas atmosphere, an additional force is introduced. The presence of oxygen will result in the oxidation of the surface of the melt ligament. The time of oxidation of molten metals is generally faster than t_{SP}. If this oxide is strong and adherent to the melt, then the droplets will retain and solidify with the shape of the ligament. The required oxygen partial pressure in the gas atmosphere to accomplish this non-spheroidization will vary with melt composition and temperature of atomization.

2.3 Theoretical Energy Requirement

One of the main advantages of SFA is the low energy requirement as compared to two fluid atomization. For instance, there is no need to compress the gas used for atomizing a liquid as it is the case in a gas atomizer. However, like in all melt atomization processes, there is a compulsory energy requirement for superheating the material above its melting point. And, as there is generally no heat recovery during a SFA process, this heat energy goes to waste as it is removed from the atomization chamber to produce powder.

Besides heating the material to melt it, additional energy input is required to break up the melt into droplets. That energy would be the one needed to create the surface area of atomized droplets. To create a surface area S of droplets atomized from a melt of mass M per unit time, the energy E_σ required is the product of S and the surface tension σ [23].

$$E_\sigma = \sigma S = \sigma \sum_{i=1}^{i=N} \pi D_{pi}^2 \tag{2.20}$$

Where N is the total number of droplets produced from the mass M per unit time (1 s).

Thus, the theoretical energy required to atomize unit mass (E_σ/M) is given by:

$$E_\sigma/M = 6\sigma\rho^{-1}D_P^{-1} \tag{2.21}$$

Where M is the product of density by volume of the liquid droplets:

$$M = \rho\pi \sum_{i=1}^{i=N} D_{Pi}^3/6 \tag{2.22}$$

From Eq. (2.21) it is clear that less energy is required for denser liquid and/or larger orifice size (or droplets size). The theoretical power requirement for atomization of different droplets size of a variety of materials is found to vary from 100 to 1000 times lower than the power required for melting [23]. However, the atomization efficiency (theoretical minimum input power/actual input power) which is generally low is found to be less than 1% so that the true power requirement for melting and atomization are in the same range for all materials [23].

Atomization efficiency is affected by different physical properties of liquid material and cooling gas. Indeed, during atomization process, liquid kinetic energy is converted into droplets surface energy and droplets movement inside the cooling gas. For a fixed liquid flow rate, an increase in the gas density can improve atomization, however an increase of the drag force due to the increase of density should be avoided so that the relative velocity between the liquid ligament and the cooling gas is not reduced.

The atomization of a liquid with higher surface tension (viscosity and density being constant) requires more energy but yield a better efficiency. It is possible that the high surface tension of a liquid acts againts coalescence of droplets after break up.

Viscosity is another liquid physical property that resists break up into droplets and consequently requires more energy. The effect of viscosity is generally minimized by superheating the liquid.

2.4 Single Fluid Atomization Techniques

2.4.1 Overview of Existing Techniques

Single fluid atomization techniques have been developed based on the limitations of two fluid atomization techniques such as gas atomization (GA) and water atomization (WA). Formation and break-up of sheet-ligament during these atomization techniques lead somewhat to irregular droplets formation [24] except for when GA is carried out in inert gas atmospheres. If consistent ligaments size and spherical droplets are required, single fluid atomization techniques are more attractive. In addition performance to requirements characterized by energy consumption, spray quality, mass flux across the spray and size distribution have become a real concern in the production of powder for many applications including additive manufacturing. Two parameters are usually considered in describing size distribution (usually log-normal), the mass median droplet size D_{50} and log-normal or geometric standard deviation σ_{LN}. Almost all applications of powders require a specific droplet size or distribution to be supplied. Although two fluid atomization processes have the advantage of yielding very fine droplets with higher throughput (but usually at low yield), their energy consumption and size distribution present considerable limitations. Thus, based on the design and performance of two fluid atomizers, technology transfer has given rise to a variety of single fluid atomization processes including Centrifugal atomization (CA), Drop on Demand Techniques (POEM, PDOD, StarJet), Continuous uniform droplet generation (UDG) and IA.

2.4.2 Centrifugal Atomization

Centrifugal atomization (CA) has been used for several decades in the metal powder industry. Centriguation of molten metal streams is an efficient method for producing high quality powders. It offers many advantages in terms of a relatively small particle size range (50–250 μm), narrow particle size distribution, spherical shape, processing strategy and flexibility, operating costs, high production rate, and high yield [25]. One of the greatest benefits of centrifugal atomization is the narrow particle size distribution. A narrow distribution is increasing the yield of the final product in the desired particle size range. Hence it may be favored over other common techniques such as gas atomization. These advantages allow for the application of centrifugal atomization in the production of common materials such as: Sn, Pb, Al, Mg, Zn, Ti, Co, and corresponding alloys [26]. Despite these advantages and the flexibility of the process, the application on the industrial scale remains relatively limited. The overall production quantities are estimated to be 100,000 t/year [25]. Hence, it is reported of playing a minor role, in comparison to other well-known technologies, such as gas and water atomization. This is attributed to the lack of scientific knowledge in this field [27]. From a scientific point of view, more fundamental research regarding process design and the atomization mechanism need to be conducted in order to improve the process and increase profitability and practicality. It is a question as to whether industry will improve its applications of centrifugal atomization and thus increase the use of the process for powder production. However, it is not yet able to produce particle sizes below 50 μm. Major problems arise concerning the spray chamber dimensions and cleaning of the device. The startup process in centrifugal atomization is

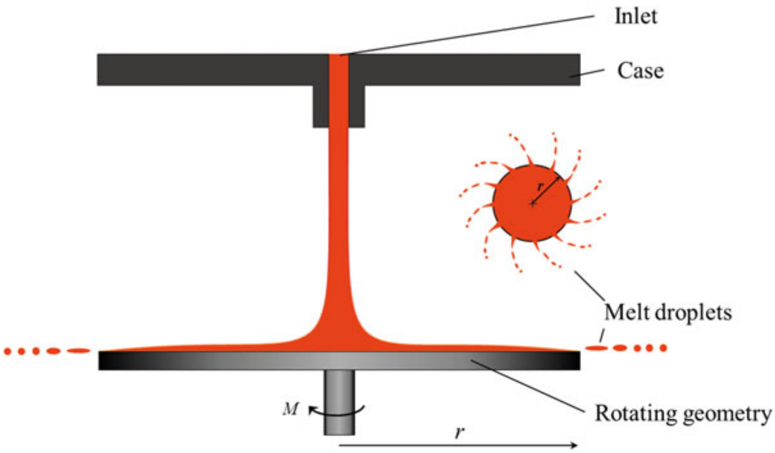

Fig. 2.4 Schematic drawing of a centrifugal atomizer

critical. Pre-solidification on the spinning disc needs to be prevented, and special care must be taken in order to guarantee the stability of the actual atomization process.

2.4.2.1 Melt Flow Disintegration

Centrifugal atomization (also known as spinning disc, rotary, or spinning cup atomization) has received increasingly more attention over the last few decades [25]. In the most standard procedure, a molten metal stream is fed centrally at the top of a rapidly spinning geometry (disc, plate, or cup) forming a liquid film, which is sheared off from the rotating body tangentially (see Fig. 2.4). The underlying mechanism of atomization occurs at the threshold, where the centrifugal force exceeds the viscous force and the surface tension of the melt film at the edge of the disc.

Depending on the melt flow rate on the spinning geometry and the rotational speed, several distinct droplet formation modes can be observed [28]. Figure 2.5 shows the predominant modes when observing a rotating flat disk process: (a) Direct Droplet Formation, (b) Direct Droplet and Ligament Formation, (c) Ligament Formation and (d) Sheet/Film Formation. These regimes may occur on top of the geometry, around, or beyond the edge of the geometry in the horizontal axis. The transition between the different states is due to an increasing liquid flow rate [29]. At relatively small liquid feed rates and rotating speed, the Direct Droplet Formation mode occurs. In this mode, the outer rim of the liquid film begins to alter into a non-uniform shape under the applied centrifugal force. Once the liquid film is unable to maintain its natural shape, the surface tension is overcome by the centrifugal force and a distinct part of the liquid volume detaches from the original body tracing a fine ligament behind it. If the droplet separates

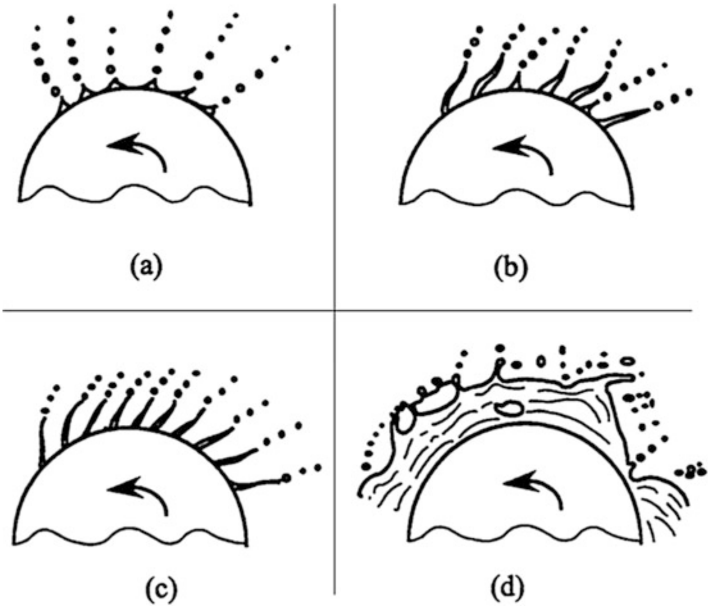

Fig. 2.5 Regimes rotary atomization liquids (**a**) Droplet Formation, (**b**) Droplet and Ligament Formation, (**c**) Ligament Formation and (**d**) Film or Sheet Formation (from [28] with permission)

from the ligament, the ligament will disperse in a series of fine droplets of near uniform size.

Further increasing the flow rate progresses the disintegration regime into the Ligament Formation mode. The volume separation step moves towards the periphery of the rotation disc or cup generating larger ligaments compared to the Direct Droplet mode. Similar to the Direct Droplet mode, ligaments disperse in small volume units-droplets. An even greater melt flow rate will create a continuous film or sheet beyond the geometry rim, forcing the flow disintegration zone off the geometry. Consequently, this regime is addressed as the Film or Sheet Formation mode. The sheet break-up mechanism, regarding the equilibrium state of contraction energy and surface tension at the sheet edge, was thoroughly investigated by Fraser et al. [30].

Figure 2.6a, b shows two in-process pictures of centrifugal melt atomization, using a cup as the rotational geometry to disintegrate the liquid stream into small droplets. Figure 2.6a shows the overall process concept: Metal flow is fed at the top of a rotating geometry. In this case, water is used to increase the cooling rate of the individual metal particles.

Having a closer look at the rotational unit (Fig. 2.6b), one can see the different volume separation modes described earlier. The melt flow is injected from the top of the process chamber onto the cup. The cup is completely filled and ligaments extend from the rim of the cup, disintegrating into small particles.

Fig. 2.6 (**a**) 30 t/h centrifugal atomization of steel with water quenching and (**b**) top view on large cup with metal flow (from [25] with permission)

2.4.2.2 Droplet Generation and Parameters

Droplet separation or atomization will only take place if the centrifugal force exceeds the restoring surface tension of the liquid metal. With the help of a simple force balance, the mean particle size of the produced metal particles is available [29]. The equilibrium condition when interpreting the droplet generation is the following:

$$F_C = F_S \tag{2.23}$$

The centrifugal force F_C and the surface force F_S are in balance. The centrifugal force of a particle on a rotating path is defined by its mass m, the radius of the geometry r_0, and the angular velocity ω.

$$F_C = m r_0 \omega^2 \tag{2.24}$$

The mass of a single liquid droplet can be calculated according to Eq. (2.25) by its density ρ and diameter d.

$$m = \frac{\rho \pi d^3}{6} \tag{2.25}$$

Using the surface force F_S, which is calculated from the surface tension σ and the particle diameter d,

$$F_S = \sigma \pi d \tag{2.26}$$

and integrating both forces in the aforementioned balance, one yields the following expression defining the mean particle diameter produced by centrifugal atomization.

$$d = \sqrt{\frac{6\sigma}{\rho r_0 \omega^2}} \tag{2.27}$$

This simple expression is only applicable for Direct Drop Formation and fails for high metal feed rate where Ligament and Sheet Formation become predominant disintegration modes. Its failure arises from not taking into account the complexity of the bulging film, or aerodynamic forces, or slippage on the atomizer itself [25]. The mean particle size currently attainable by centrifugal atomization varies between 50–250 μm depending on process parameters, especially rotation speed but also material parameters (melt density and surface tension) [31].

2.4.2.3 Rotating Speed and Material Properties

Plookphol et al. [27] used the above expression [Eq. (2.27)] for a series of experiments proving the dependence of the mean particle size on the atomizer size and shape, oxygen level, and feed rate of an SnAgCu alloy. A significant discrepancy has been found when comparing the actual particle size from the process and theoretically calculated. The greatest deviations from the equation were examined in the low rotation regime below $<$15,000 rpm. However, the force balance in centrifugal atomization is able to predict the process outcome from the particle point of view.

Figure 2.7 shows theoretically possible particle sizes depending on the rotation speed for various pure metals at their liquidus temperatures. Generally, it can be seen that with increasing rotation speed a smaller particle size results due to greater centrifugal forces applied to the melt. Aside from the mechanics, there is a clear influence of the material parameters: density and surface tension. The inner diagram compares the square rooted ratio of surface tension to density of all chosen pure metals. Generally, smaller particles result from lower restoring mechanisms of density and surface tension.

2.4.2.4 Atomizer Size and Shape

A number of authors [27, 32, 33] experimentally confirmed the influence of size and shape of the rotating unit. Generally, it was shown that changing the geometry from disc to cup will create significantly smaller particles. The cup geometry favors the disintegration mechanism of the melt flow compared to a simple disc. A variation of the cup wall angle did prove to provide further enhancement in terms of final

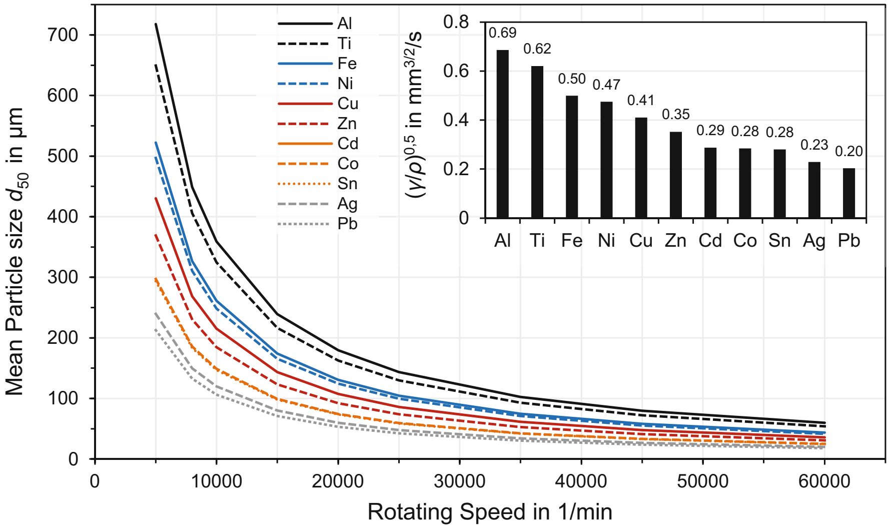

Fig. 2.7 (**a**) Theoretical particle size dependent on the rotation speed of various pure metals r = 20 mm and (**b**) square rooted surface tension to density ratio

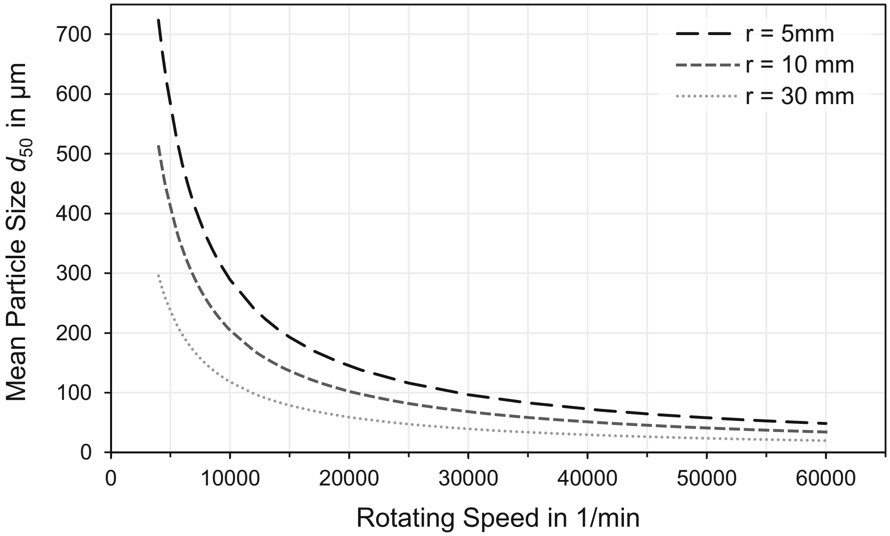

Fig. 2.8 Theoretical particle size dependent on the rotation speed and variation of disc diameter from 5 to 30 mm of SnAgCu alloy calculated after Plookphol [27]

particle size. Figure 2.8 is derived from the theoretical expression of Eq. (2.27) and does not account for technical boundaries. One can see that an increasing disc diameter improves the rotational force at the geometry rim generating smaller droplet units at constant melt flow rate.

2.4.2.5 Feed Rate, Wettability and Oxygen Content

Melt feed rate has been reported as an important process parameter having a major impact not only on mean particle size, but also particle size distribution [27]. An increasing of melt flow will inevitably affect the apparent disintegration mode. Melt layer thickness may increase at the rim of the geometry and coarser particles result. On the other hand, an unwanted premature solidification may occur when choosing a very low melt flow rate. Hence, careful evaluation and adjustment of the melt flow rate will support the process outcome in terms of particles size and throughput.

Another very important system property affecting the continuity and yield of the atomization process is the wettability of the atomizer material [32]. In terms of contact angle of the melt, and also surface roughness of the geometry, it becomes evident when improving wettability, slippage of the melt can be reduced, resulting in steady atomization [27]. Often the atomizer is pre-coated with the melt and mechanically roughened before running the process.

One parameter recently receiving more attention is the amount of oxygen in the processing chamber [33]. Theoretically, oxygen will support the surface oxidation form metal oxide layers. As a result the restoring surface tension of the melt flow increases hence, hindering the capability of secondary disintegration of droplets. Yet, this phenomenon remains unclear. Process safety (i.e. explosion in aluminum atomization) and also consumer specifications demand that the oxygen level be as low as possible [27].

2.4.2.6 Process Design

The design of centrifugal atomizers is mainly driven by the centrifugal forces necessary for the materials to be atomized. The most crucial dimension of an atomizing chamber is the diameter required for sufficient centrifugal radial path. Figure 2.9 shows a common type of industrial atomizer. The melting unit is attached on top of the spray chamber consisting of a crucible, induction coil, furnace, stopper, and motor for the crucible. The melt is introduced through the nozzle onto the rotational geometry. The rotational geometry is powered by a motor, which rotates the disk.

Process monitoring via a camera or a window allows for the process to be controlled and regulated manually, which is especially important at the beginning and the end of a batch. As discussed earlier, surrounding gas may have an influence on the stability of the process and the final product. Commonly, an air supply unit is mounted to the spray chamber. The granular product can be extracted from the unit either continuously or discontinues at the bottom of the metal tank.