

Thermal plasma processing in the nineties

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Abstract - In the context of materials which are expected to be one of the major technical issues in the nineties, an attempt is made to describe the role which thermal plasma processing may play in these years. Newer developments in plasma melting/remelting, extractive metallurgy, plasma deposition, plasma synthesis, and plasma densification are briefly described since it is anticipated that these developments will carry over and expand in the nineties. The major research needs associated with these developments are delineated and it is concluded that some areas in thermal plasma processing will develop into viable technologies in the nineties.

INTRODUCTION

In this paper, an attempt will be made to forecast developments in the field of thermal plasma processing in the nineties. Forecasts are always risky, unless there are trends already pointing towards such developments. Fortunately, this is the case for most of the newer developments.

It should be realized that thermal plasma processing must be viewed in the context of much broader technology trends; some of them are clearly visible today. There is no question that materials and materials processing will be one of the most important technical issues in the nineties. This will not be restricted to the development of new materials, but will also include the refining of materials, the conservation of materials (by hard facing, coating etc), and the development of new processing routes which are more energy efficient, more productive, and less damaging to our environment. Thermal plasma processing will play an important role in these developments. Its potential for developing new materials-related technologies is increasingly recognized and many research laboratories all over the world are engaged in advancing the frontiers of our knowledge in this exciting field. An interesting example of the utility of plasma processing has been recently demonstrated in connection with a breakthrough in the field of high temperature superconductors. It has been shown that superconducting films of these new materials can be deposited by plasma spraying [1].

In spite of great strides over the past 20 years, the number of successful industrial applications in thermal plasma processing is still relatively small. There are two major reasons for the relatively slow growth of this technology. First of all, industrial efforts have not been sufficiently paralleled by basic studies at universities and, as a consequence, the required engineering base for many processes is still poorly developed or non-existent. This problem is directly linked with the nature of thermal plasma processing as a highly interdisciplinary field which cannot succeed without extensive interdisciplinary endeavors. Knowledge in plasma physics, gaseous electronics, fluid dynamics, and heat transfer has to be paired with experience in surface chemistry, electrochemistry, and material science. Universities are usually not structured for this type of interdisciplinary cooperation. And secondly, the potential of thermal plasma processing has been frequently overestimated, resulting in widespread disappointment and, even more damaging, in cuts in R & D funding. For a realistic assessment of the potential of thermal plasma processing it is essential to take competing technologies into account.

The first part of this paper will focus on some of the more recent developments in the field of plasma melting/remelting, plasma extractive metallurgy, plasma deposition, plasma synthesis, and plasma consolidation. It is felt that these developments provide guidance for what may be

expected in the nineties. These developments, however, will be closely linked with advances in basic research. Therefore, the most pressing research needs will be discussed in the second part of this paper, followed by some conclusions.

Because of space limitations, plasma decomposition (for example of toxic waste) and plasma chemical processing are not included in this paper, although these areas may see some interesting developments in the nineties. Plasma decomposition especially is considered to become a growing technology in the nineties.

Reference material for this paper has been primarily derived from a recent workshop [2] and from two reports [3,4].

EXAMPLES OF RECENT DEVELOPMENTS

(a) Melting and remelting technologies

In contrast to conventional arc furnaces with consumable cathodes which have been known for almost 100 years, newer developments - known as arc-plasma or plasma furnaces - employ plasma torches with non-consumable cathodes in a controlled furnace atmosphere. There are several advantages to this approach, including reduced noise levels, elimination of graphite electrode consumption, higher yields which is particularly important when processing valuable alloying elements such as nickel and molybdenum. In general, the key advantages of thermal plasma techniques as specifically applied to melting technology include

- the ability to achieve a steady-state, uniform flow of partially ionized gas with bulk gas temperatures well above those obtainable with chemical (combustion) flames or resistance heater systems;
- operation in an inert or reactive environment, thus providing complete control of the atmosphere, and
- a compact system that can process material in a variety of forms at high throughput rates and with relatively high electrical/thermal efficiency.

Today, a wide variety of arc plasma torches are in use or projected for scrap melting, alloying, iron melting in cupolas and remelting technologies. These plasma torches operate with D.C. or A.C. either in the non-transferred or in the transferred mode and at power levels up to almost 10 MW. In the case of non-transferred arcs, the plasma torch is essentially an arc gas heater [5] producing extremely hot gases which emanate from the torch in the form of a plasma jet. The more common approach, however, makes use of transferred arcs where the molten pool serves as one of the electrodes and the major energy input is at the arc root of the molten bath surface. As an example, Fig. 1 shows the Linde arc

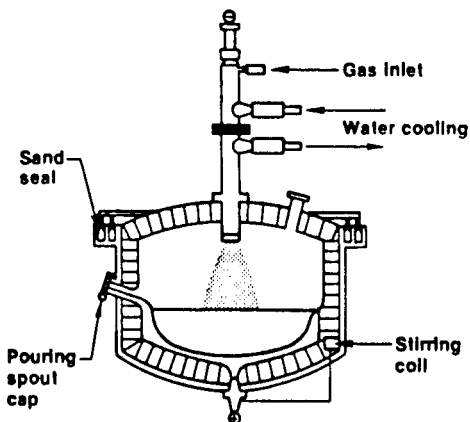


Fig. 1. Schematic of the Linde Plasma Arc Furnace.

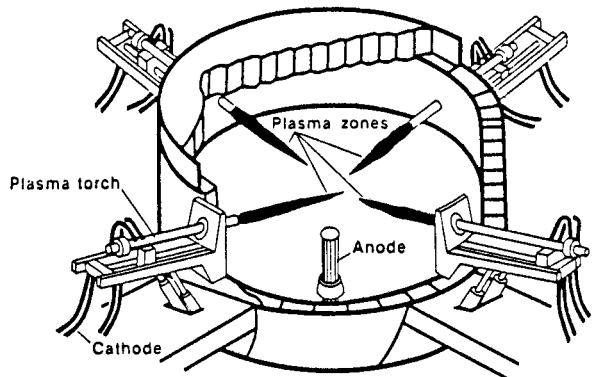


Fig. 2. Schematic of the Freital Steel and Voest-Alpine D.C. Arc-Plasma Furnace.

furnace [6]. Although this furnace is not in use, it may be considered as the prototype of modern arc-plasma furnaces. The water cooled D.C. arc plasma torch mounted in the furnace roof is operated in the transferred arc mode with the molten bath as anode. A continuous supply of argon passing through the torch provides an inert furnace atmosphere. This particular torch has been operated with a hot (thermionically emitting) cathode at current levels around 10^3 A.

Based on this principle, powerful plasma furnaces have been developed by Freital Steel (East Germany) and Voest-Alpine (Austria). In Freital, 15 and 35 ton plasma furnaces are currently in operation for making high quality steels [7]. The 35 ton furnace uses four plasma torches mounted in the sidewalls of the furnace and tilted at 30° from the horizontal as shown schematically in Fig. 2. The arc length can be adjusted by axial movement of the torches. Thermionically emitting cathodes are employed consisting of tungsten alloyed with lanthanum, thorium, or ittrium. These furnaces have been used to melt carbon steels, nickel based alloys, and tool steels.

An interesting development has been reported by Krupp [8], based on plasma furnaces operated with A.C. arcs. These furnaces have been designed for melting of scrap, production of alloy steels, and for reheating of steel in the ladle. In contrast to the previously discussed plasma furnaces, the frequently troublesome bottom electrode is no longer required, and compared with conventional arc furnaces, these furnaces provide higher yields, lower noise levels, and reduced flicker. A schematic of the plasma torch used in such furnaces is shown in Fig. 3. The thermionically emitting, hot electrode consists of tungsten and has been designed to withstand A.C. operation. During the positive half-wave of the applied voltage (with the electrode as anode), the electrode heat transfer is substantially enhanced which, however, provides favorable conditions for the following half-wave where the electrode serves as cathode. Thermionic emission is now spread over a larger surface area resulting in lower current densities and correspondingly longer lifetime of the electrode.

Actual furnaces use three plasma torches mounted nearly vertical into the roof of the furnace. A 3 ton pilot furnace equipped with 3 torches, each operated at 4 KA (1.6 MW) is being tested and a 10 ton furnace is under construction with three 6kA torches.

Another type of arc plasma torch using a "cold" cathode (usually Cu) in combination with vortex-stabilization of the arc is frequently found in modern plasma metallurgy. In this design the arc voltages are substantially higher than in typical hot cathode torches and the corresponding arc currents are much lower, resulting in enhanced electrode life.

A typical example of such a torch used for ladle heating is shown in Fig. 4. The D.C. arc is operated in the transferred arc mode which requires a

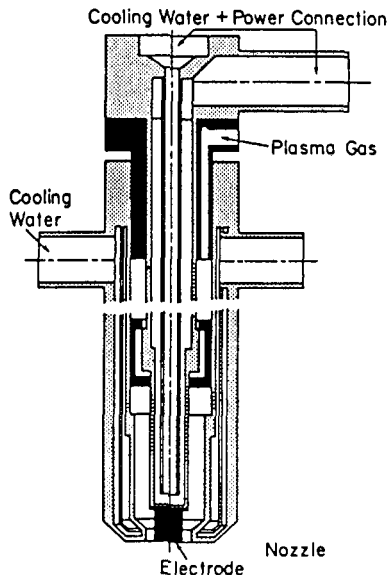


Fig. 3. Plasma Torch for A.C. Operation Developed by Krupp.

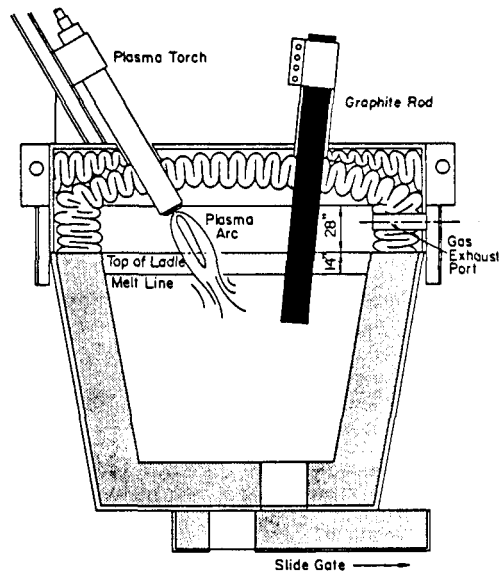


Fig. 4. Schematic of Arc-Plasma Ladle Heating (Plasma Energy Corp., U.S.).

return electrode (graphite rod). The main objective in this application is to hold steel in the ladle at constant or slightly increasing temperature levels.

In a joint development project of U.S. Steel and the Plasma Energy Corporation (U.S.), a 220 ton ladle with a specially designed cover (see Fig. 4) is heated by two 6 MW arc plasma torches which have an electrode life of more than 200 hours.

The Japanese steel industry has been very active in developing new and improved technologies for plasma melting and remelting. Daido Steel, for example, developed over the past 10 years two melting and refining systems with the objective of replacing the conventional vacuum arc melting and vacuum induction melting systems by a more cost-effective process.

The Plasma Induction Furnace (PIF) is a hybrid system shown schematically in Fig. 5. Conventional induction heating is augmented by arc heating using a transferred arc.

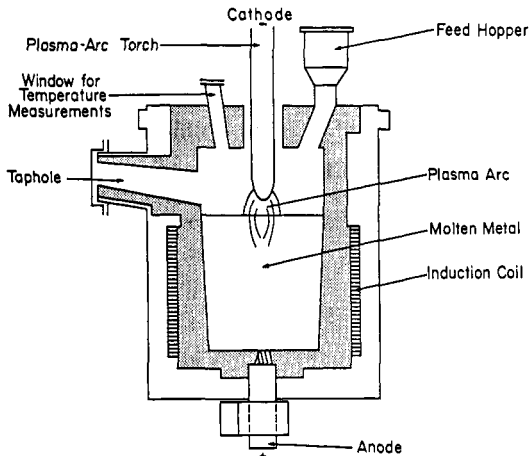


Fig. 5. Schematic of the DAIDO Steel Plasma Induction Furnace (PIF).

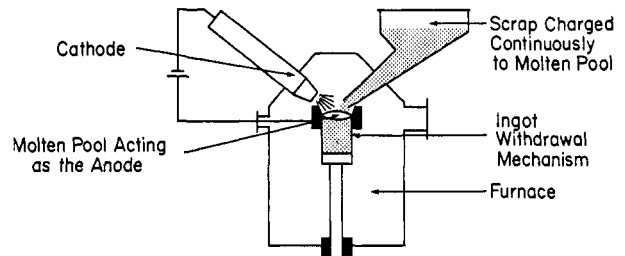


Fig. 6. Schematic of the DAIDO Steel Progressive Casting Furnace (PPCF).

The second type of furnace known as the Plasma Progressive Casting Furnace (PPCF), shown schematically in Fig. 6 has been designed for melting and casting as a competitive process for vacuum arc melting, electroslag refining, and electron beam melting. Typically, six 90 kW torches are used and operated in the transferred arc mode and the solidified material in the bottom of the crucible is continuously withdrawn into the ingot chamber (see Fig. 6). In contrast to vacuum furnaces, this approach allows alloying with high vapor pressure metals without excessive losses.

Both the PIF and the PPCF use D.C. plasma torches operated in the transferred mode as all previously discussed arc heaters.

In the following, an example of a non-transferred arc heater will be discussed which has been developed by SINTEF in Norway. This arc heater is immersed in the liquid pool of the metal [9]. This heater is particularly useful for ladle heating and, in general, for liquid metal heating.

The arc operates inside a pair of concentric, consumable graphite electrodes immersed into the liquid metal bath up to 100 cm below the surface. The torch gas is a mixture of Ar and N₂ ranging from 0 to 100% N₂. A prototype arc heater has been operated at a power level of 1 MW in liquid ferrosilicon.

The major advantage of this design over a transferred arc system is the higher efficiency and the elimination of excessive heating of the refractory liners of the furnace.

Because of space limitations, other interesting developments in the field of plasma metallurgy cannot be included in this section. The interested reader is referred to a number of excellent surveys on this subject [3, 10-12].

(b) Plasma extractive metallurgy

There has been considerable interest in extractive metallurgy as evidenced by numerous laboratory and pilot plant scale studies reported in the literature (extraction of iron, titanium, molybdenum, and ferro-alloys).

Two different types of furnaces have been used for plasma extractive metallurgy: transferred arc reactors for ferroalloy production (similar to those used for melting), and reactors where a reducing gas is preheated and upgraded by using plasma torches. The reduction of the ores to be treated is performed in a furnace filled with coke, which is mainly used as a refractory material for providing a sufficiently long residence time for the injected ore particles to be reduced.

Transferred arc reactor

A number of different reactors have been designed to replace traditional submerged arc furnaces. Such reactors have been operated on a pilot plant scale for the production of ferrovanadium, ferrochromium and manganese, of pure or alloyed molybdenum [12, 13], for the melting/smelting of direct reduced iron and iron ores [14].

Industrial processes are now under development for ferrochromium production in South Africa:

- at Middelburg at a 10.8 MW power level with a Tetronics type furnace;
- at Minteck at a 3.5 MW power level;
- at Samancor at a 8.5 MW power level, with a Voest Alpine type furnace.

Plasma reformers for direct reduction [12, 14, 15]

Direct reduced iron in the form of pellets is an attractive source of fresh iron units for specialty and modest volume steelmakers whose output is insufficient to justify large blast furnaces. For example for ball bearings, a fresh iron source is required to abate the problem of accumulation of residual elements upon repeated remelting of scrap.

Most production systems for direct reduction consist of shaft furnaces in which iron oxide pellets are heated and reduced by an upward flowing stream of carbon monoxide and hydrogen. Instead of methane reformers, plasma reformers are attractive for direct reduction processes, as they offer the potential to utilize virtually any source of reductant including coal. This is true for the case of the Plasmared plant of SKF at Hofors in Sweden which started up in January 1981. This plant started with LPG as a reducing agent and was converted to coal within a year. With the plasma reformer, the productivity of the original Wiberg-Soderfors system which operated initially at 25,000 tons of iron per year, has been increased to 70,000 tons per year. At the beginning the gas was heated with six SKF plasma torches of 1,300 kW each using 350 Nm³/h of plasma gas, SKF now uses units of 5 to 6 MW each with a gas flow rate of 1,600 Nm³/h (with enthalpies between 4 and 8 kWh/m³).

Smelt reduction

Injection of powdered ores into plasma heated reducing gas streams is an effective means for hot metal production. Simultaneously, gaseous by-products are generated which essentially consist of carbon monoxide and hydrogen with energy requirements in the range of 2.5 MWh/ton of hot metal. The hot gaseous by-products may be used for fluid bed preheating and partial prereduction of the incoming raw material or, as expected with the Plasma Dust process of SKF at Landskrona (Sweden), for district heating and other purposes.

In its Plasmazinc process at Landskrona in Sweden, started in September 1984, SKF has been recovering metals from waste oxides collected in steel mill exhaust filters and from secondary oxides. The raw materials are injected into a coke filled shaft furnace where liquid metals and slag are collected at the bottom of the shaft and tapped in a conventional manner.

Gaseous zinc and lead are carried with the hot off-gas to condensers where they are collected as liquid metal. The condensation of the vapor is very efficient even with a low zinc-content charge, since there is no carbon or water vapor present in the off-gas and consequently no reoxidation of the zinc takes place. Since there is no combustion air or oxygen used, the off-gas does not contain nitrogen and its volume is small. The Plasmazinc plant at Landskrona has a capacity of approximately 70,000 tons of dust per year when operated with three plasma generators of 6 MW each.

(c) Plasma deposition

Although plasma spraying is already a well-established commercial process, its science base is still in the developmental stage. Plasma spraying is considered to be one of the prime candidates for producing high temperature resistant coatings for turbine blades, anti-wear and anti-corrosion coatings, especially for high temperature applications. Recent experiments indicate that plasma spraying using high frequency (r.f.) plasma torches may be advantageous for producing near net shape parts of metals and alloys. The latest tests demonstrate the feasibility of depositing the new high temperature superconducting materials by plasma spraying [1].

Fig. 7 shows a schematic diagram of a D.C. plasma spray torch. The plasma jet emanating from the spray torch provides the environment for melting and acceleration of powders injected into this jet. Fig. 8 shows some typical temperature and velocity distributions in the plasma jet [16] without powder injection, and Fig. 9 shows the velocities of particles injected into the plasma jet [17].

Recent developments in the field of arc plasma spraying, in particular the introduction of the low pressure plasma deposition (LPPD) process, have resulted in a renewed interest in the capabilities of plasma spraying. In conventional plasma spraying the deposition is carried out at atmospheric pressure, whereas in low pressure plasma deposition (LPPD), the process takes place in a low pressure environment. In contrast to conventional plasma spraying, LPPD provides major benefits such as

- higher particular velocities which create greater than 98% dense deposits;
- broad spray patterns which produce large deposit areas, and
- transferred arc heating of the substrate which improves the deposit density and adhesion of the coating.

These characteristics provide LPPD with the potential of becoming a viable method of consolidating powders for high performance applications. Moreover, the LPPD process can be automatically regulated to make controlled deposits on complex geometries at reasonably high deposition rates (up to 50 kg/hr).

(d) Thermal plasma synthesis

Over the past years, thermal plasma synthesis of ultrafine and ultra-pure powders has been attracting increasing interest, especially in connection with the synthesis of ceramic materials. High intensity arcs, plasma jets, and high power r.f. discharges are the primary source for producing thermal plasmas required for this emerging technology.

Because of the high temperatures ($> 10^4\text{K}$) which are typical for thermal plasmas, chemical reactions are much faster than those encountered in conventional processing. Also quench rates of the product powders are very rapid which is necessary to avoid decomposition of the products. Fast reaction and quench rates result in very short overall processing times which may be as short as milliseconds. This translates into small reactors with relatively high throughput rates. In spite of this attractive feature, the relatively higher processing costs of using plasma processing must be offset by some superior material's properties. A number of review articles on this subject have been published [3, 18-22].

Plasma synthesis of fine powders may proceed from gaseous, liquid, or solid precursors. Nearly all binary refractory carbides and some refractory nitrides and oxides have been synthesized in plasmas by gas phase reactions using arc or arc-produced plasma jets (see Ref. [22] for an extensive listing of references).

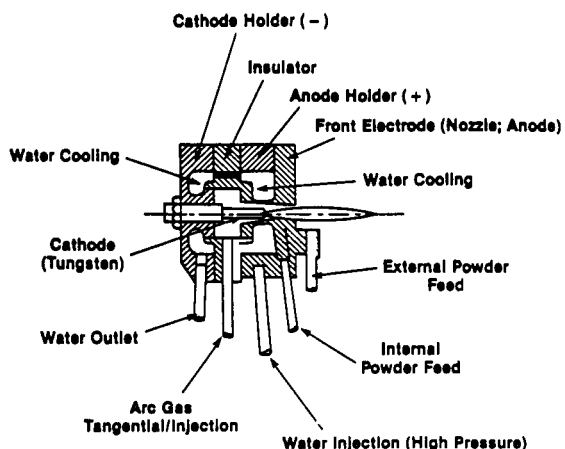


Fig. 7. Schematic of a Plasma Spray Torch.

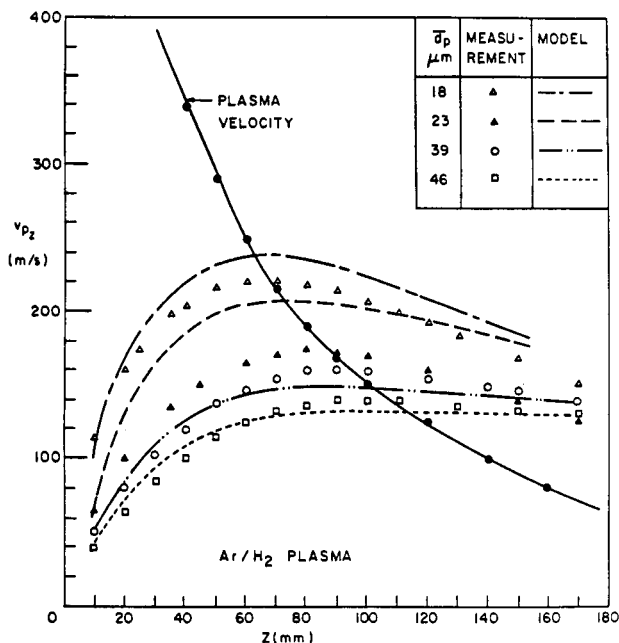


Fig. 9. Particle Velocities in a Plasma Jet.

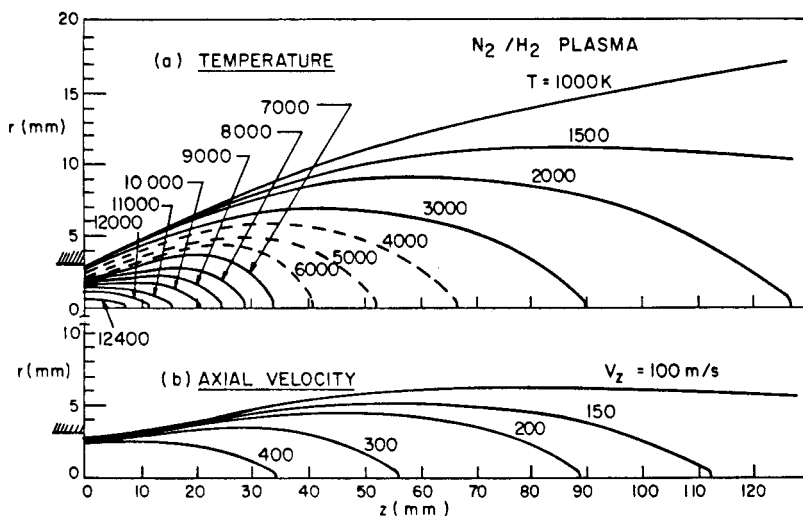


Fig. 8. Temperature and Velocity Distributions in a Typical Plasma Jet.

A number of ultrafine oxide powders have been produced in thermal plasmas. Ultrafine oxides have a wide range of uses in surface coatings, high density ceramics (high tech ceramics), pigments, catalysts, and dispersion strengthening of metals. Three arc-related plasma techniques have been explored: reaction of volatile metal chlorides with oxygen, evaporation and subsequent condensation of oxide powders, and evaporation of bulk oxides. Industrially, TiO_2 and high purity SiO_2 are produced by the chloride process. Mixed oxides of chromia and titania or chromia and alumina have been produced by introducing mixed chlorides into the plasma reactor.

Another route of synthesizing fine powders in plasmas starts with at least one solid or liquid precursor. Such precursors are usually injected into an arc, an r.f. discharge or into a plasma jet by means of a carrier gas. This carrier gas is either inert or it may participate in the desired chemical reaction. Particles which are in the size range from 10-50 μm (typical for plasma spraying) may easily penetrate into the hot core of the

plasma due to their inertia, whereas smaller particles ($< 10 \mu\text{m}$) remain usually in the colder plasma fringes due to the high viscosity of the plasma [23]. Since plasma synthesis with solid or liquid precursors frequently requires particle sizes below $10 \mu\text{m}$, injection of such particles still represents a severe problem.

In general, particulates injected into a thermal plasma undergo a number of processes which may be classified as:

- a) **physical processes, wherein the plasma's heat induces phase changes, i.e. melting, evaporation, and also crystal structure changes;**
- b) **chemical processes, wherein the plasma induces one or more chemical reactions.**

In the first category, the heat transfer process is of particular importance which is much more complex than in an ordinary gas due to the presence of charged and dissociated particles [24, 25].

Processes falling into the second category are even more complex because, in addition to the physical changes typical for the first category, one or several chemical reactions may be induced either on or within the particle itself or in the gaseous phase after evaporation. The kinetics of the various reactions is still poorly understood.

The plasma equipment utilized for plasma synthesis of fine powders encompasses d.c. and a.c. arc heaters, as well as r.f. plasma reactors. Such generators have been developed in a wide variety of designs and over a wide power range and many plasma generators are commercially available today. For some specific applications and for laboratory studies specially designed plasma reactors have been introduced.

(e) Plasma consolidation

Plasma consolidation includes the processes of spheroidization, densification, and sintering. The first two of these processes are already commercially developed, whereas plasma sintering is still in the laboratory stage.

Spheroidization and densification

Frequently, both processes occur simultaneously as porous, irregularly shaped agglomerates are injected into a thermal plasma. Similar equipment as used for plasma spraying is employed for these processes, but the particle size of the agglomerates may be substantially larger ($\sim 100 \mu\text{m}$) than that used for plasma spraying. As the particles sinter and/or melt in the plasma, they assume a nearly spherical shape and densify at the same time. Commercially, fine particles are spheroidized in a plasma for a variety of applications, including materials with a controlled porosity, catalysts [26, 27], abrasives, and materials to transfer ink in photo copiers. A wide range of different materials have been spheroidized, including oxides [27-31] and carbides [29, 32].

In the case of refractory particles of irregular shape injected into a plasma stream, the spherical liquid droplets that are produced rapidly solidify upon cooling. This may be useful in producing free-flowing powders, but it may be even more valuable in certain ceramics, wherein metastable forms, especially the amorphous state are produced. Mullite and certain other aluminosilicates have been found to form as glasses in particle sizes $< 20 \mu\text{m}$.

Plasma densification of presintered agglomerates of metals (e.g. W, Mo) and of carbide-metal mixtures (e.g. WC-Co) has been used to produce spherical, densified powders. Such powders possess excellent flowability which is beneficial to subsequent plasma coating operations [33].

Plasma sintering

Sintering of high-tech ceramics in thermal plasmas has the potential of drastically reducing the time period required for this process, compared to conventional technology. In addition, plasma sintering offers the

opportunity of restrained grain growth and for tailoring heat transfer during the sintering process which may result in desirable structures and properties of the sintered materials.

Plasma sintering is a pressureless sintering process which may cover a pressure range from 760 to a few Torr. For pressures below 75 Torr, the plasma may no longer be classified as a thermal plasma, because of substantial deviations from Local Thermodynamic Equilibrium (LTE).

The essential characteristics of plasma sintering and of any other sintering process are an increase in density and strength of a powder compact upon heating.

Rapid sintering of ceramics has been observed by various investigators [34-44], using gaseous plasmas produced by microwaves, glow discharges, or r.f. discharges. Extensive work by Johnson and coworkers [39-44], using a 5 MHz r.f. argon plasma, shows rapid sintering of various aluminas. They also achieved rapid sintering of MgO and doped alumina using three different plasma devices, namely a microwave-induced plasma, a hollow cathode discharge, and an r.f. inductively coupled plasma [43]. A fine-grained structure of sintered oxide was observed.

More recently, the effects of gas composition and pressure on r.f. plasma sintering of MgO without sintering aids has been reported [45]. Only a few reports about plasma sintering of non-oxide ceramics such as silicon carbide have been published [46, 47]. Kijima [47] has successfully sintered silicon carbide, a most difficult ceramic to sinter, in an argon r.f. plasma to near theoretical density without substantial grain growth. No commercial scale developments of this technology are presently known.

RESEARCH NEEDS

Basic research in the field of thermal plasma processing received relatively little attention in the fifties and sixties, in spite of the fact that space-related plasma technology played an important role during these years. This situation changed drastically over the past 10 years which has seen rapidly increasing research efforts, triggered to a large degree by industrial developments. But one should not overlook the fact that space-related plasma research contributed immensely to the research base in the emerging field of thermal plasma processing. Advances in arc technology, plasma diagnostics, plasma torch and gas heater design are just a few of the more striking examples of such contributions.

In spite of substantial advances over the past 10 years, there are still many pressing research needs which will be addressed in the following. Since some of these research needs are rather generic, they will be discussed independently, but with reference to the anticipated impact on new developments which have been mentioned in the previous section. Efficient research in the complex field of thermal plasma processing requires a two-pronged approach: experimentation and modeling with emphasis on the interaction between these two disciplines. An experimentally verified model which is the final goal of this interaction may prove to be useful for scale-up to pilot plant or even full scale operation.

Although experimental needs and modeling work will be discussed in separate sections, the reader should keep in mind that there is a close link between these two approaches. Because of space limitations, only the major thrust areas will be discussed.

A. Experimental Studies

This section will be subdivided into diagnostics and plasma reactor design, with emphasis on diagnostics.

(a) Diagnostics

Diagnostics suitable for thermal plasma processing must be, first of all, concerned with the plasma itself. Plasma parameters which are of primary concern include temperature, velocity and plasma composition. Well-established methods exist already for determining temperatures in laboratory-type plasmas [4] which are in or close to LTE. Besides emission, absorption, and laser spectroscopy, enthalpy probes have been

used for measuring plasma temperatures. In general, these methods are time-consuming and severely restricted in their applicability to plasmas typically experienced in thermal plasma processing. Therefore, further work is needed to

- adapt or modify existing and develop new methods for measuring plasma temperatures to meet the specific requirements of thermal plasma processing;
- develop methods which allow to cover a wide plasma parameter range in a reasonable time (automation of measurements).

Although thermal plasma processing implies that the plasma is in LTE, more or less severe deviations from LTE may occur, even in atmospheric pressure plasmas. Recent measurements using electric probes in the fringes of a wall-stabilized arc (Fig. 10) indicate strong deviations from kinetic equilibrium ($T_e \gg T_h$) close to the wall [48]. The basic trends according to these measurements are in agreement with analytical predictions, and the electron temperature at the wall is in excellent agreement with the analysis [49]. Even more severe deviations from LTE are expected in the LPPD and the low pressure plasma sintering process, especially in terms of deviations from chemical equilibrium.

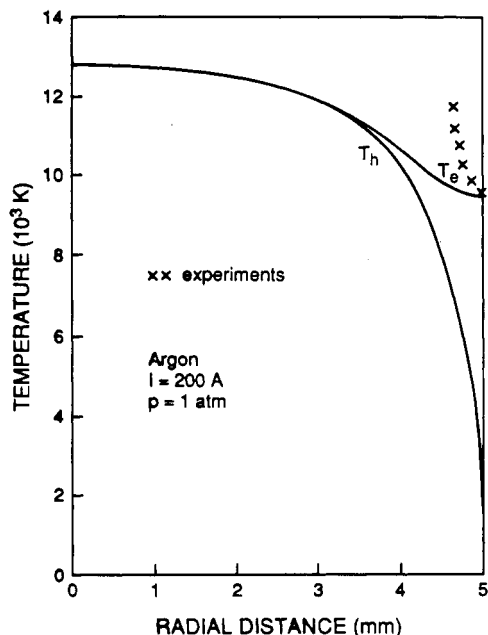


Fig. 10. Calculated and Measured Temperature Distributions in a fully-Developed, Wall-Stabilized Arc.

Since deviations from LTE may play an important role in thermal plasma processing, there is a need to enhance our understanding of deviations from LTE by

- studying such deviations at atmospheric and reduced pressures.

Among the various methods which have been used for measuring plasma velocities, including pitot probes, high speed photography, laser Doppler Anemometry (LDA) etc. [4], LDA has been particularly attractive as a non-intrusive method with excellent spatial resolution. Unfortunately, it is difficult to inject small particles into a plasma ($< 1 \mu\text{m}$) which are needed to ensure that they will readily follow the plasma flow. And even if they can be injected, they will rapidly evaporate. For this reason, reliable measurements of plasma velocities still remain a problem. Further work is needed to

- develop reliable methods for measuring plasma velocity fields without disturbing the plasma. Again, these measurements should be fast so that a wide parameter range can be covered in a reasonable time.

Measuring of the plasma composition in the presence of chemically reacting species is, to a large degree, an unresolved problem. Spontaneous Raman scattering, CARS, and LIF have been rather successful for non-intrusive, in-situ measurements of species concentrations in combustion systems. So far, only the feasibility of Laser Induced Fluorescence (LIF) for plasma diagnostics has been demonstrated [50]. Its application to thermal plasma processing systems, however, is still in its infancy [51]. Therefore, there is a pressing need to

- **develop methods for measuring in-situ species concentrations in thermal plasma processing systems.**

The improvement and development of new diagnostic techniques for thermal plasmas would have a strong impact on any processing system which makes use of such plasmas. A comprehensive data base derived from such measurements would be crucial for modeling work.

Besides plasma diagnostics, diagnostics of particulates exposed to or injected into thermal plasmas plays an important role for understanding plasma/particulate interactions. A number of diagnostic techniques have been developed for in-flight measurements of particulate matter in thermal plasma systems, including methods for measuring particle velocities, particle fluxes, particle diameters, and particle surface temperatures [4].

Without dwelling on details, it may be stated that the level of sophistication of these techniques is high and increasing. One common drawback, however, is the fact that these methods require expensive equipment and highly trained personnel. There is a great need to

- **develop simple and inexpensive methods for measuring particulate parameters in thermal plasma systems.**

The data base derived from such measurements would be particularly important for plasma deposition, plasma synthesis, and plasma extractive metallurgy.

(b) Reactor design

In principle, thermal plasma processing makes use of three different types of reactors: transferred arc, plasma jet, and high frequency (r.f.) plasma reactors. The design of plasma reactors for laboratory-scale experiments is usually governed by diagnostic access rather than efficiency, throughput, proper mixing of the reactants, electrode life, environmental impact etc. These considerations, however, play an important role for industrial reactors, but because of space limitations, they cannot be addressed here.

In the case of transferred arc plasma reactors and arc plasma torches an important design aspect is associated with electrode life. Relatively little basic work has been pursued to understand the electrode behavior in the hostile environment experienced in thermal plasma processing. Electrode design has been more an art than a science.

One of the major drawbacks associated with r.f. plasma processing has been the relatively poor coupling efficiency between power supply and plasma reactor. Presently available r.f. power sources are not designed for plasma generation. Based on these discussions, there is definitely a need to

- **establish the science base for electrode behavior in thermal plasma processing which applies to both cathodes and anodes, in particular to cold cathodes;**
- **develop r.f. power sources geared towards the generation of thermal plasmas. This is more a development rather than a research task.**

Improvement of electrode lifetime derived from such studies would have an immense effect on arc-based thermal plasma processing. A similar statement can be made about r.f. plasma processing if the coupling efficiency can be substantially improved.

Without any question, the growth potential of plasma synthesis, plasma deposition, and plasma densification would further increase with such improvements.

B. Modelling

In this section, selected modeling needs will be discussed related to the developments mentioned in Section 2. As indicated before, modeling plays a key role for the development of the potential of thermal plasma processing into viable technologies.

One of the prerequisites for any modeling work in this field is the availability of thermodynamic and transport properties. Such properties are readily available for single gases and simple gas mixtures. Actual plasma processing, however, is characterized by complex gas mixtures, including trace elements which may have a significant effect on some of these properties [52]. The behavior of the discharge may be drastically affected by the properties of such mixtures. Traces of low-ionization potential material, for example, may change the arc behavior completely [53].

Although the calculation of thermodynamic and transport properties, even of complex mixtures, is well known today, the necessary input data (especially collision cross sections or interaction potentials) are frequently not available. Therefore, there is a need to

- generate data for collision cross sections, especially for neutral/neutral and neutral/ion interactions;
- single out those contributions in complex mixtures which dominate the plasma behavior.

Similar as in the experimental part, modeling should first consider the plasma itself. Modeling of transferred arcs, plasma jets, and r.f. discharges in terms of temperature, velocity and plasma composition has been previously reported, but almost exclusively for simple gas systems. As an example, Fig. 11 shows a comparison between calculated and measured temperature profiles in a free-burning argon arc [54], indicating that non-equilibrium effects ($T_e > T_h$) cannot be neglected, even at atmospheric pressure. This makes modeling substantially more complex.

For many years, modeling of thermal plasma systems has been restricted to laminar flow situations which is of limited validity for thermal plasma processing. Over the past five years, turbulence models have been introduced, especially in connection with modeling of plasma jets, including vortex flows [55, 56]. Quantitative agreement with experimental data, however, is still not satisfactory.

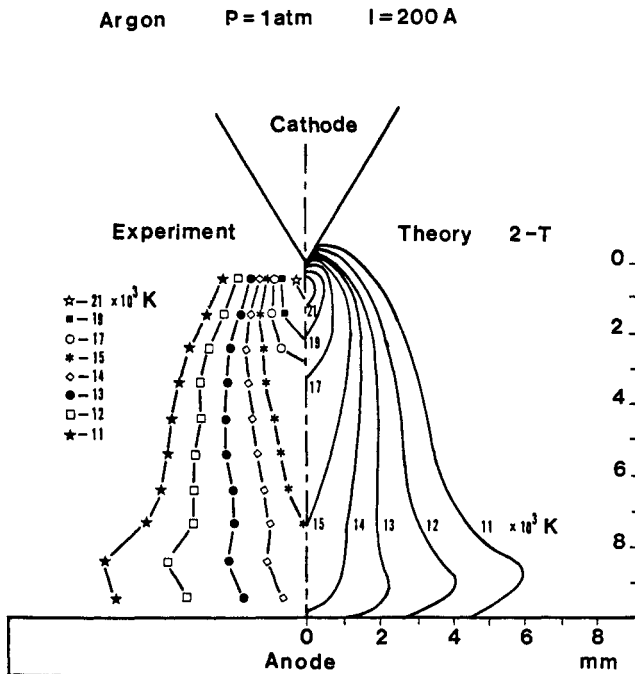


Fig. 11. Comparison of Measured and Calculated (Two-Temperature Theory) isotherms in a Free-Burning Arc.

Recently, first attempts have been reported to model plasma flows experienced in low pressure plasma spraying, using highly simplified models [57, 58].

Overall, significant progress has been made in modeling of plasmas and plasma reactors over the past years, but more work is needed to

- extend modeling to more complex gas compositions;
- improve turbulence models for plasma jets, with and without vortex flow;
- develop non-equilibrium models for the low-pressure plasma spraying and the low pressure plasma sintering process.

This type of modeling work would favorably affect all developments discussed in Section 2.

Modeling of plasma/particulate interaction attracted increasing attention over the past years. Representative publications are summarized in Refs. [23, 25, 59, 60]. These references also describe the relative importance of various mechanisms involved in heat, mass, and momentum transfer to particulates in thermal plasma flows. Some of these mechanisms are well understood, but there is little known about the effect of particle shape, particle charging, radiative heat exchange among particulates under dense loading conditions, and about the synergistics of various mechanisms. Understanding of these basic mechanisms is a prerequisite for realistic modeling of particle trajectories and of the associated particle temperature histories.

Since most of the plasma reactors with particulate injection operate in the turbulent mode, particle trajectories, especially of fine powder particles ($< 10 \mu\text{m}$), can no longer be defined as such. As an example, Fig. 12 shows the effect of turbulence in a plasma jet on particle trajectories, indicating that they move within certain bands rather than following fixed trajectories.

For predicting heat transfer to particulates in plasma flows, heat transfer coefficients are required. A recent survey [61] shows that there is a large discrepancy of predictions among different authors.

In spite of significant progress over the past years, some of the basic mechanisms of plasma/particulate interaction still require clarification, in particular there is a need to

- study the effect of particle shapes, of particle charging, of radiative heat exchange among particles and of the synergistics among the various mechanisms and their contributions to heat, mass, and momentum transfer;
- determine reliable heat transfer coefficients for particulates in plasma flows.

Such studies would primarily affect extractive metallurgy, plasma deposition, and plasma densification.

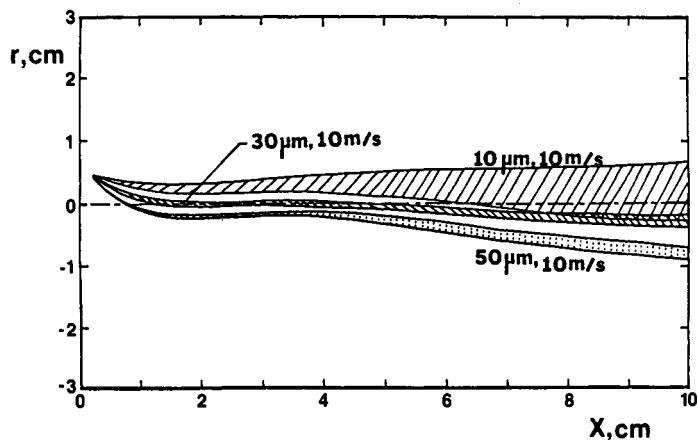


Fig. 12. Effect of Turbulence on Particle Motion in an Atmospheric Pressure Argon Plasma Jet.

Last but not least, modeling of chemical reactions in thermal plasmas has to be considered, because of its importance for plasma synthesis and the formation of fine powders in thermal plasmas. Over the past years synthesis, especially of refractory powders (carbides, nitrides, borides and metal oxides), attracted widespread interest [62]. Although there is a wealth of information in the literature on feasibility studies of synthesizing fine powders in thermal plasma [22], many fundamental aspects are not well understood.

Equilibrium calculations, for example, based on minimization of Gibbs free energy, with consideration of both gas and condensed phases, are inadequate for predicting the yield or even the proper composition of the products from thermal plasma reaction systems. This is primarily due to the dominating influence of nucleation kinetics, a non-equilibrium effect.

The synthesis of fine powders in thermal plasma reactors results frequently in particle sizes in the order of 10 nm. It has been postulated that such particles are formed by homogeneous nucleation associated with the fast quench of the supersaturated vapor phase. Such ultrafine particles show a high rate of densification in the sintering process [63]. One drawback of such ultrafine particles, however, is the problem of handling (agglomeration, contamination). For many applications particles in the size range from 0.1 to 1 μm would be desirable. Unfortunately, particle nucleation and particle growth are poorly understood in the context of thermal plasma processing. Problem areas which need to be addressed include

- studies of precursor injection (gases, liquids, solids) and mixing with the plasma;
- kinetics of chemical reactions (non-equilibrium effects) including effects of quenching;
- particle nucleation and growth.

These studies would be more or less geared towards plasma synthesis.

CONCLUSIONS

In general, thermal plasmas as an electricity-intensive processing tool will be competitive if one or more of the following criteria are met:

- a) High value-added product,
- b) Unique or superior product properties,
- c) Improved processing routes (economics, safety, pollution etc.).

More specifically, it is felt that there is a definite growth potential for the following thermal plasma processing technologies in the nineties:

1. Plasma deposition for producing coatings, graded coatings (metals, alloys, ceramics, super-conductors, amorphous metals, etc.) and monolithic near net-shape parts.
2. Plasma melting and remelting in particular applications which involve ladle and cupola heating. Plasma furnaces for melting steel scrap may overcome their economic barrier in the nineties.
3. Plasma synthesis of ultrafine pure powders of carbides, nitrides, borides and metal oxides is expected to see the first full scale commercial installations in the nineties.
4. Plasma extractive metallurgy will definitely experience some interesting developments in the nineties, but commercial installations will be restricted to areas where economic conditions are favorable for this technology (South Africa, Sweden).
5. Plasma densification of presintered agglomerates into powders of excellent flowability which is already commercialized may experience further growth and it is conceivable that plasma sintering may become a commercial process in the nineties.

These expectations, however, hinge to a large degree on corresponding research efforts and on the support which this field needs to be successful in the nineties.

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