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New Developments in Specialty Metals Processing
- the Role of the Science Base

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1. Introduction

It is an honor to participate in SIMAP 88 and be asked to prepare an assessment of advances in specialty metals processing. At the same time it is a daunting task to be original in treating this topic, in view of the numerous excellent publications on this very topic (1-4). For this reason, the processing of specialty metals and alloys is a very interesting subset of the materials field, because to a considerable extent rather different rules will apply here, providing an unusual set of driving forces for change.

Many applications of specialty metals are in the defense, nuclear, aviation and energy fields, where performance may be an overriding criterion; this is in sharp contrast to commodity type materials, e.g. reinforcing bars where price is the dominant factor. This facet of specialty metal applications does allow one to employ "exotic technologies", the development of which may be largely supported by "outside" research contracts. At the same time the producers "will have to look over their shoulder" to watch for competition from new materials, such as ceramics and composites.

Another unusual feature of many specialty metals applications is that materials have been "qualified" for certain uses, not only through their intrinsic properties, but by the processing route, such as vacuum arc remelting, powder metallurgy and the like. This may have a certain ossifying effect, followed by an abrupt change, once a new process is qualified.

Finally, in many instances the research emphasis has been on characterization rather than on processing; as a result, highly sophisticated

characterization techniques are being routinely used, while the fundamental understanding of the process technology may not be at a comparable level. Indeed, many superallow processing techniques have to be regarded as "overgrown" laboratory scale installations, which have not fully benefitted from proper scale-up techniques.

Figure 1 shows a "road map" of specialty metals processing operations using traditional technology. It is seen that this includes vacuum induction melting as the first step, followed by (plasma) vacuum arc or electroslag refining, with electronbeam or plasma cold hearth refining as alternatives. In the traditional route the ingots thus produced would be both hot and cold worked. Some more exotic variations do exist on this general theme, that could include electroslag casting, ESR followed by VAR and the like.

Figure 2 shows some of the new technologies that are either being implemented, or at least are in the development stage. It is seen that these include:

- new melting arrangements, such as cold crucible and cold hearth melting

- new forming arrangements, which may be collectively termed as near net shape casting. These include the powder, the metal spray routes as well as thin slab or sheet casting.

For the sake of completeness, we should also mention directional solidification, the casting of single crystal turbine blades (5) and rheocasting (6), although these topics will not be discussed in detail.

In order to complete the listing of new technologies with which the specialty metals community must be concerned, we need to mention:

- structural ceramics

- metal matrix composites

- surface modification and coating technologies.

These new materials and the associated processing technologies represent major potential competition for the specialty metals producers both in terms of price and performance. As an example of potentially superior performance, Figure 3 shows a comparison between the stiffness and the specific strength of various metals and metal matrix composites (7).

The ideal response to this quite real and potentially devastating competition would have to be the combination of:

- significantly improving existing operations (PCHRC, Spray Forming, etc. being excellent examples); and

- attempts at merging these technologies through exploiting cladding, surface modifications and metals matrix composites, which would still be excellent end users of the metal products.

Our main attention in this review will be confined to three stages in the processing sequence, namely
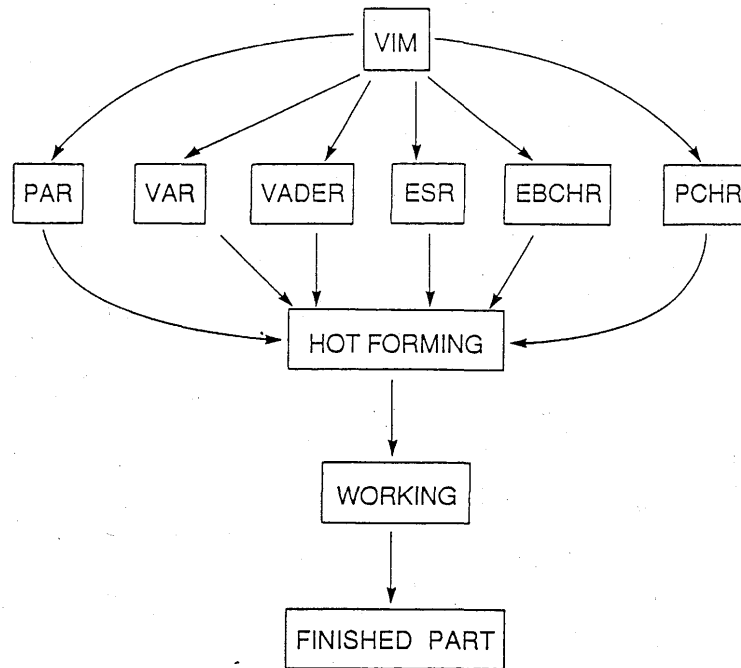
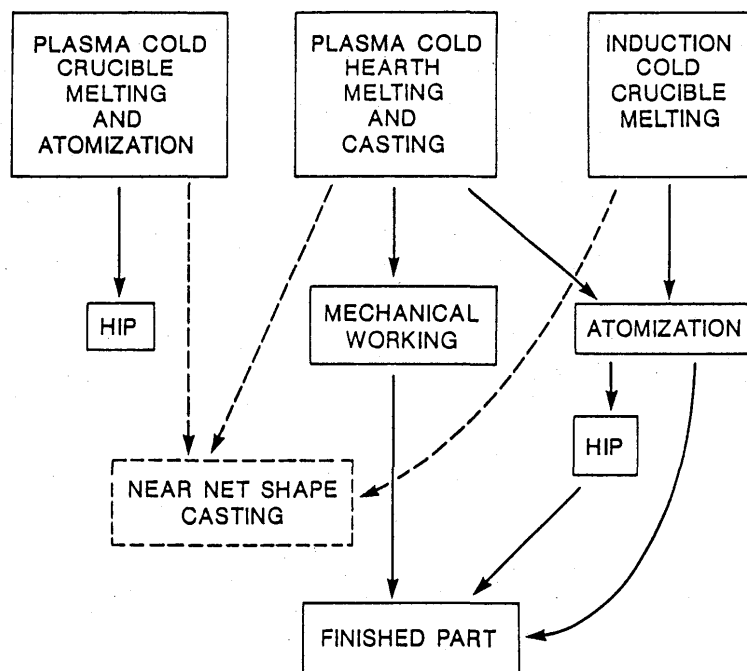Figure 1   A general "road map" of specialty metals processing, following the conventional route.

Figure 2   Some novel concepts in specialty metals processing.
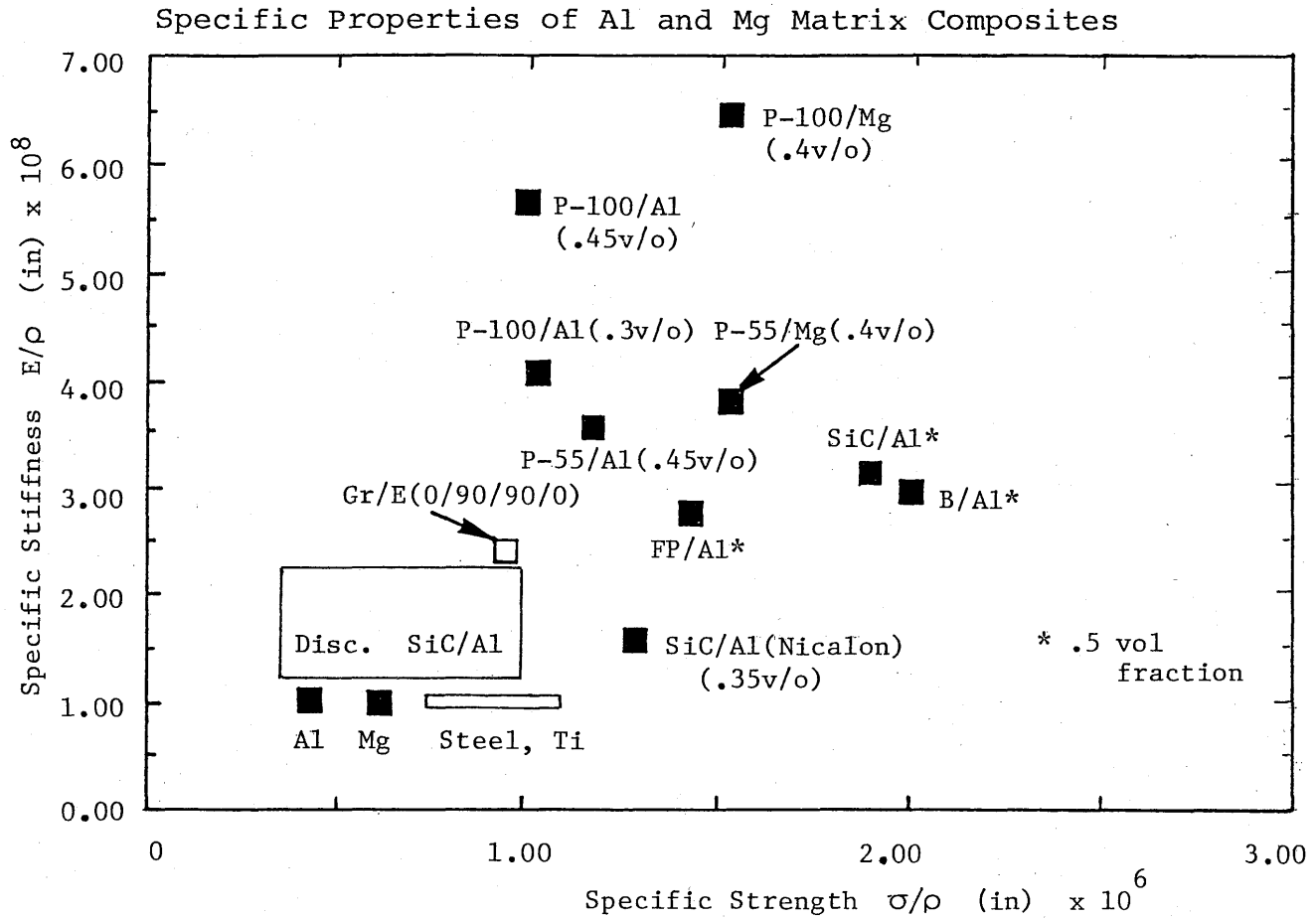
Specific Properties of Al and Mg Matrix Composites

Figure 3 Some properties of aluminum and magnesium matrix composites, compared to aluminum, magnesium, steel and titanium, after Mortensen, Cornie and Flemings.

- melting

- refining

- solidification processing.

The fact that these are very closely interrelated cannot be overemphasized, as is the need to fully integrate the total operational sequence. The effect of impurities introduced into the system by a poor melting or refining practice or the results of inappropriate primary solidification processing (e.g. poorly formed electrodes in VAR or ESR) will manifest themselves in the final product quality.

What are the prime objectives in these three stages?

Melting

The objective of melting operations is to transform a solid feedstock, e.g. scrap, sponge and the like to a melt of specified composition and temperature, while avoiding contamination. Vacuum induction melting has represented a major advance in its time and a great deal of progress is being made in this direction, with the development of improved vacuum induction melting technologies; however, contamination by the crucible walls still cannot be completely avoided. The skull melting techniques, which have been pioneered by the U.S. Bureau of Mines, rely on the formation of a solidified shell between the crucible wall and the melt. This shell effectively protects the wall and will prevent contamination due to melt-wall interactions. Several variations of this technique exist. The heat source may consist of electron beams, a plasma or induction.

As fully discussed by several authors (8,9), these techniques have both advantages and disadvantages.

EB melting, sketched in Figure 4, has the advantage of providing an extra clean environment at greatly reduced pressure. The disadvantages are associated with the high cost and with the possibility of volatile alloying element losses in the beam impingement area.

Plasma melting is appealing, because of the reduced cost. Furthermore, the less well-focused energy source will lead to reduced volatilization; nonetheless, vaporization losses will still occur.

Cold crucible induction melting (10) is a newer technology, but one which can offer many potential attractions, including greatly reduced volatile element losses and good agitation, which may help in the refining. As an example Figure 5 shows a schematic sketch of an inductively heated cold crucible furnace, employed at the Duriron Company. The segmented wall design, necessary to avoid excessive eddy current generation in the copper cooled walls, is clearly illustrated. An even more recent idea, shown in Figure 6, suggests that the cold crucible technology may be adopted to levitation melting, thus avoiding any contact with the crucible wall altogether.

It would appear that the cold hearth melting processes offer major technological attractions and we can look forward to many other applications of this technology.

Beam targets and effect:
Gun A:
    1 Ingot pool rim for flat pool
    2 Trough spout lip for avoiding stalactites
    3 Electrothermal barrier for slag removal
    4 Pool superheating in front of the skimmer
Gun B:
    1. Slag EB-heating
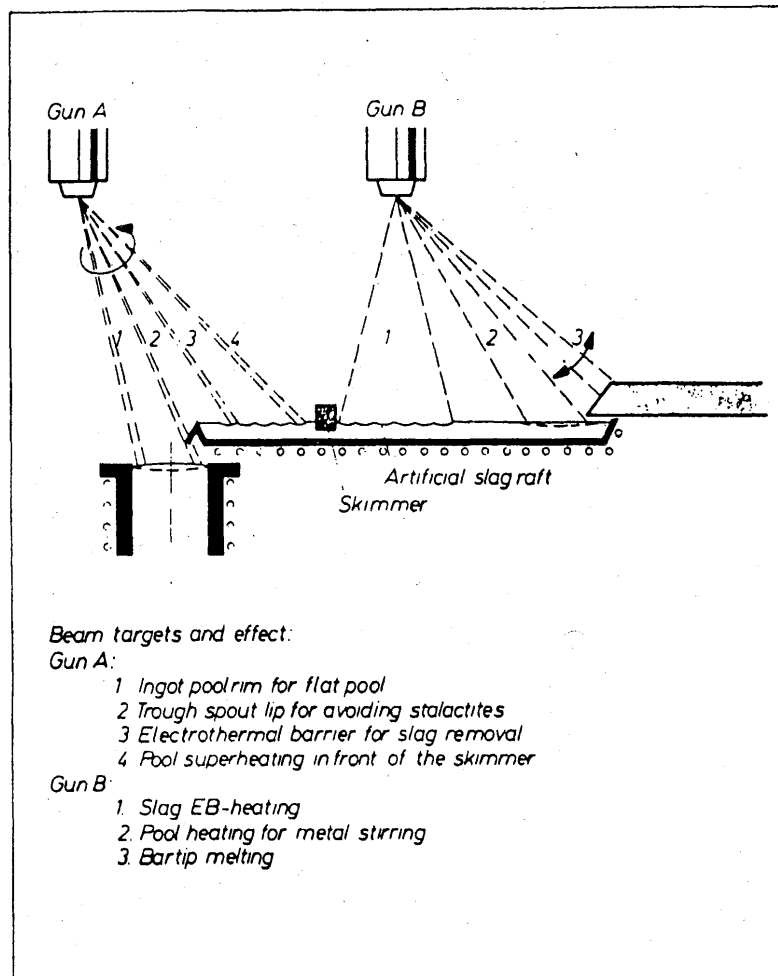   2. Pool heating for metal stirring
    3. Bar tip melting

Figure 4   A schematic sketch of an electron beam refining furnace, used for nickel based superalloys; courtesy of Dr. Georg Sick, Leybold Hereaus
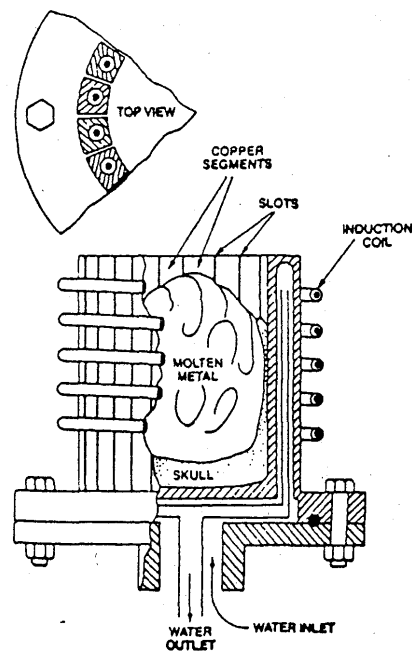
Figure 5   An induction heated Cold Crucible Melting Operation (10)
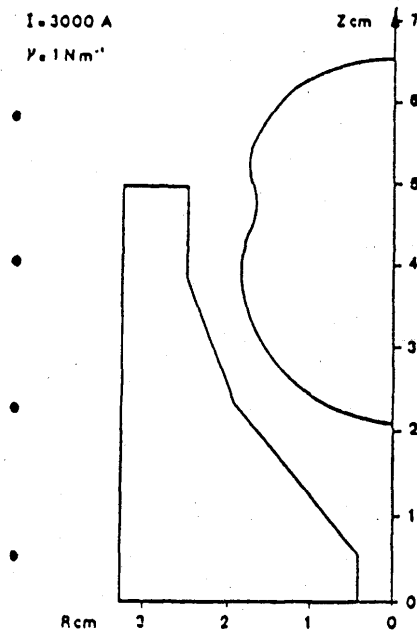
Figure 6   A schematic sketch of levitation melting using a cold

crucible, after Ganoud et al. (11)

Refining

In the usual metallurgical sense the term refining means the removal of undesirable impurities. In the majority of specialty melt refining operations (VAR, ESR, VADER PCHR etc.), the actual composition changes are quite small and are usually confined to degassing (in the vacuum systems) and possibly the removal of the inclusion particles. Indeed, in many instances the letter R in ESR, VAR etc., may equally stand for remelting or refining.

The whole question of refining is being brought rather more sharply into focus by the need to provide progressively cleaner, i.e. inclusion free, melts of tightly defined composition for the subsequent solidification processing operations.

While the thermodynamic, equilibrium criteria for refining are quite well established, the actual kinetics and the factors that govern these kinetics, are rather less well understood. We shall return to this point in the discussion of the science base.

Solidification Processing

During the past decade major advances have been made and are being made in solidification processing. The classical remelting techniques, VAR, PAR and ESR involved relatively deep pools, quite long solidification times, hence large crystals and the possibility of macro and microsegregation. Thus significant deformation processing was a necessary step in the operating sequence.

The recent trends, as were illustrated in Figure 2, are toward near net shape processing, that is, transforming the melt into the desired final shape in one step or at least in a reduced number of steps. On the traditional steel scene this involves the direct casting of thin slabs (say 25-50 mm thick) or sheet (say 1-2 mm thick) (12).

In specialty metals processing the attractive near net shape processing technologies include:

-    the directional solidification and the casting of single crystal turbine blades

-    the formation of rapidly solidified powders, which are then hot isostatically pressed

-    the spray forming of near net shapes, e.g. plasma spraying or the Osprey Process

-    the forming of rapidly solidified ribbons or sheet.

These near net shape processes offer many potential advantages. Due to the much shorter solidification times the grain sizes are greatly reduced; furthermore, segregation is also minimized. Last but not least, the elimination or reduction in mechanical working can offer substantial cost savings.

These new technological developments may be summarized by stating that key

advances are being made in skull melting (and refining) and in the near net shape casting areas, and that both of these are having a major impact.

As an example Figure 7 shows a titanium pipe, produced by low pressure plasma spraying (34).

2. The Need for Innovation

Some Technical and Economic Considerations

The need for technological change is well illustrated by using the well-known "S" curve (13), which as shown in Figure 8, provides a relationship between the effort (resources) put into the development of a product or a process and the performance or financial return. This typical "S" shaped curve shows that initially resources have to be sunk into a development project, without any immediate return, but then, if a new market is being captured and/or major cost reduction is possible through new technology, there will be a sharp increase in the profitability. As the technology matures we shall reach the realm of diminishing returns. At that point, or even before, it is essential to consider new technologies.

Indeed in many real life situations discontinuities will occur and the history of a product (group) performing a given duty may be described by a series of "S" curves, as shown in Figure 9. It is seen that the new product or process may rapidly displace the old operation. Within a metallurgical context, we may cite VIM replacing air melting and continuous casting replacing ingot casting as typical examples. In a more consumer oriented field, aluminum cans replacing steel cans and the possibility of aluminum being replaced by plastic containers may be quoted.

The time scale of replacement will necessarily vary with the application; in case of the integrated steel mills in the U.S., it took some 20 years or more for continuous casting to be substantially implemented, and the disastrous results of this slow rate of change are well documented.

For the sake of argument, it may be interesting to show a broad brush plot of some specialty metals processing operations on an "S" curve, as done on Figure 10. One may make a plausible case for placing ESR, PAR and VAR processes in the mature category, spray processing, near net shape casting in general and cold hearth processing on the steep part of the curve, where rapid advances are being made with corresponding economic benefits.

Cold crucible melting, and the whole field of composite materials processing are operations with a major potential, "ready to burst" on the scene.

3. The Science Base of (New) Materials Processing Operations

In a general sense, one should state that the science base of materials structure, properties and characterization, tends to be much more advanced than the science base of the processing operations themselves. This may seem paradoxical when one considers the very high degree of sophistication involved in the design of processing equipment, such as electron beam guns, plasma torches and the like.
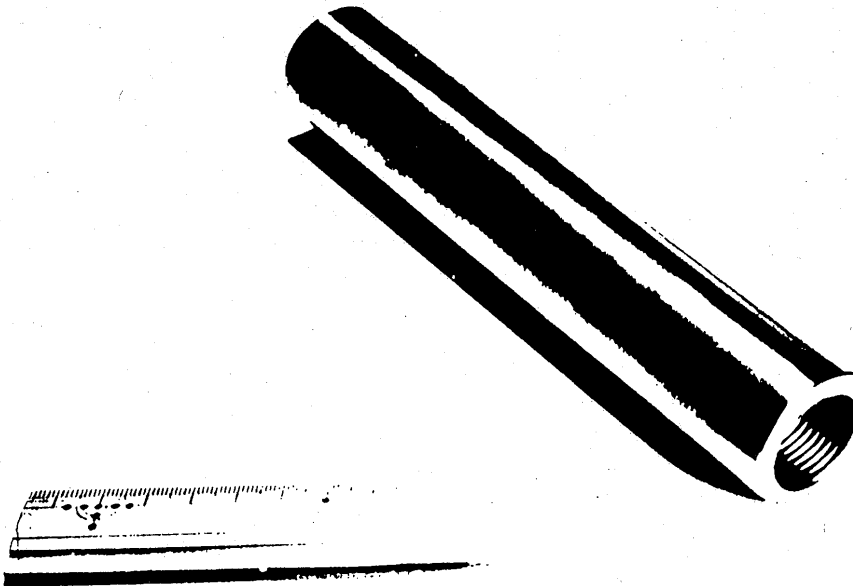
Figure 7   Photograph of a Titanium pipe, produced by low pressure
           plasma spraying. Courtesy of Nippon Steel Corporation (34)
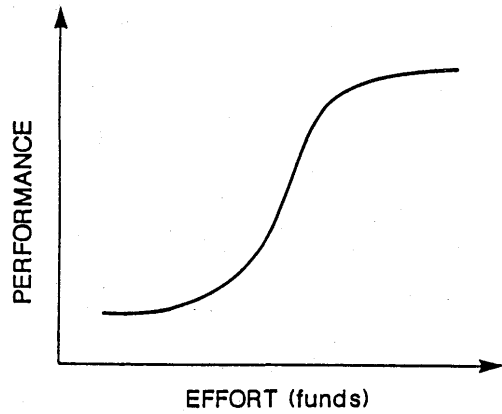
Figure 8   The "S" curve, depicting
the relationship between
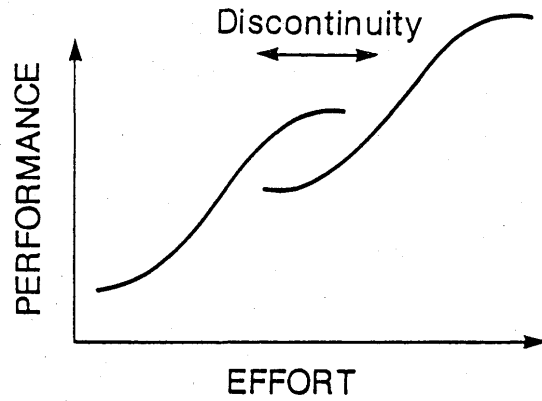effort and performance.

Figure 9   The discontinuity in "S" curves
as we jump from one process or
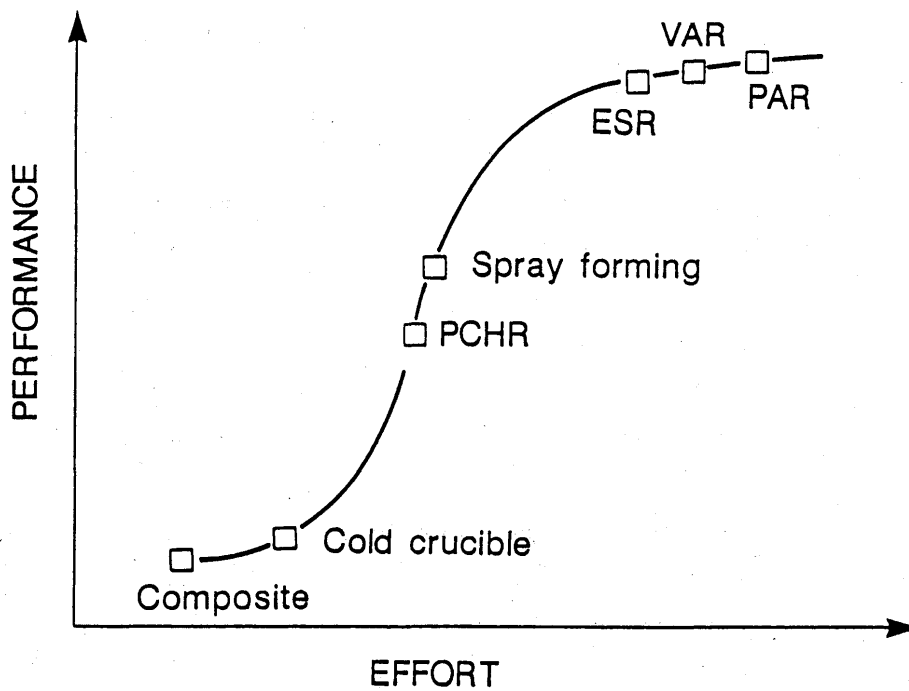product to another competing one.

Figure 10   A tentative "S" curve for specialty metals processing
operations.

In the following we shall make brief comments on the science base of some selected processing steps with emphasis on the knowledge:

-   currently applied

-   potentially available from other fields

-   that has to be developed.

The following generic categories will be considered:

3.1 Plasma Systems

3.2 Melt Behavior

3.3 Atomization

3.4 Solidification Processing

3.1 Plasma Systems

Plasmas are produced by the passage of a current between two or more electrodes, which results in a partially ionized gas. Plasmas are an essential ingredient in VAR, PAR, VADER and in many of the cold hearth melting and refining operations. A good description of plasma behavior is available in the literature (4,14), and elegant observations on the VAR system have appeared in previous Vacuum Metallurgy Conference Proceedings (15). Our present state of understanding of plasma systems may be summarized as follows.

There exists a good general understanding of the behavior of plasmas in a qualitative way and we have a good idea of the types of velocities and temperatures and heat transfer rates that one may expect in these systems (16,17). As an example, Figure 11 shows a comparison between the experimentally measured and the theoretically predicted temperature profiles in a transferred arc system.

However, the actual design of plasma guns is still largely empirical and there is a great deal of uncertainty regarding the precise behavior of the regions near the electrodes, e.g. anode and cathode fall, anode and cathode spots, and the like. This lack of knowledge hampers both optimized plasma gun design and a realistic description of the region where a plasma impinges onto a molten metal pool, e.g. surface deformation, volatile element vaporization etc., such as sketched in Figure 12, where it is seen that the impinging plasma jet may cause a depression in the free surface and possibly the formation of free surface waves. The temperature and the velocity fields in the vicinity of this depression are quite difficult to predict and yet may have an important influence on the local heat and mass transfer rates. We should mention here that the rapidly growing welding literature (18) may be an excellent source of ideas for modelling plasma-melt interactions.

3.2 Melt Behavior

In virtually all melting and refining operations we encounter a molten metal pool, which as illustrated in Figure 13, will undergo circulation due to a combination of factors, that may include:
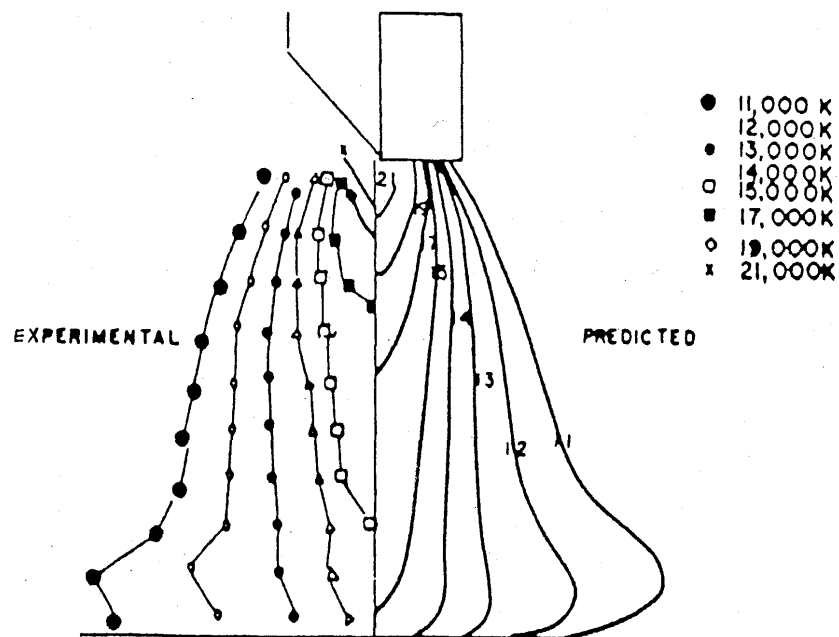
Figure 11   A   comparison   between   experimentally   measured   and

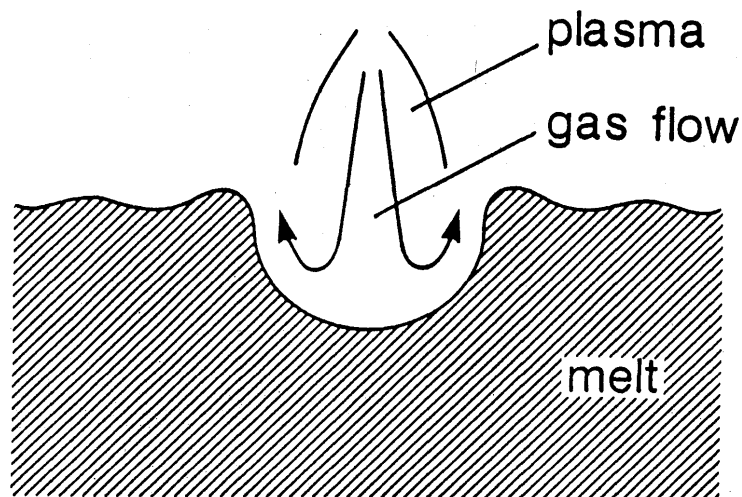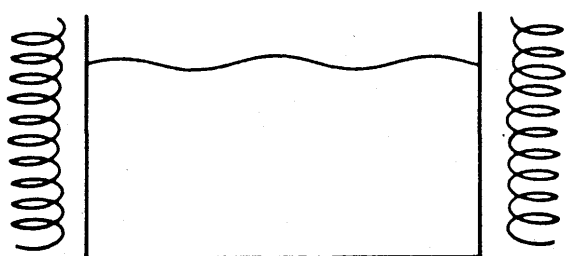theoretically predicted temperature profiles in a transferred
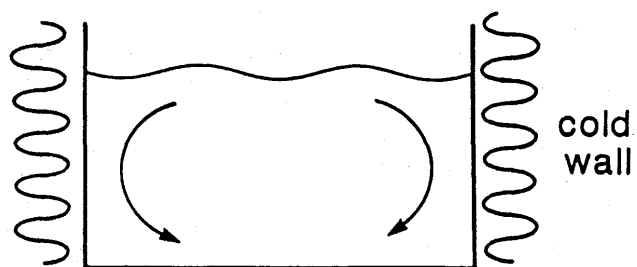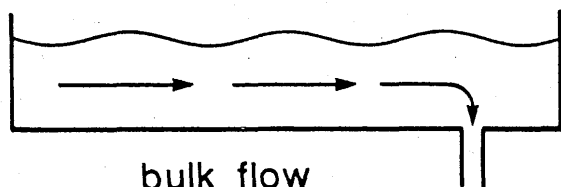
arc system (16)

Figure 12   Sketch showing the free surface deformation when a plasma jet

impinges onto a melt surface.

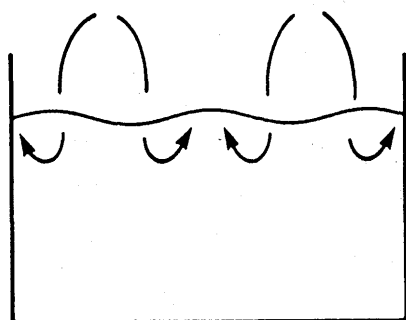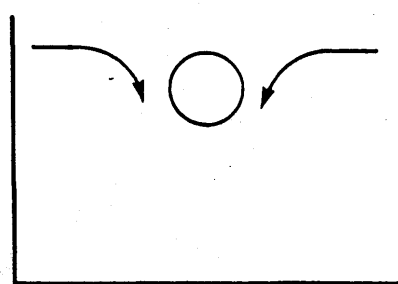induction stirring

buoyancy

cold wall

bulk flow

EM forces due to plasmas

surface tension driven flow

Figure 13   Sketch of some basic fluid flow situations encountered in melting

- electromagnetic forces (induction of direct passage of a current)

- buoyancy (i.e. natural convection)

- bulk transport (e.g. in hearth flow systems)

- surface tension driven flows.

Problems of this type have been very extensively studied by engineers in secondary steelmaking and ladle metallurgy (19,20).

It is understood in a qualitative sense by the specialty metals community that this convection may play an important role in affecting the performance of the system. More specifically, as illustrated in Figure 14, convection will modify:

- the temperature profiles

- the shape of the solid skull in skull or cold crucible melting and refining operations

- the flotation of inclusion particles in CHR systems, among others.

However, the quantitative description of these velocity fields has yet to be systematically undertaken within the context of specialty metals processing.

In principle these problems are quite readily formulated by writing down the (turbulent) Navier-Stokes equations:

$$\nabla \cdot \rho u = 0 \tag{1}$$

$$\rho \frac{D u}{D t} = - \nabla \rho - \nabla \cdot \tau + \rho g + \rho F_b \tag{2}$$

where:

$$\frac{D}{Dt}(\ ) = \frac{\partial}{\partial t}(\ ) + u_x \frac{\partial}{\partial x}(\ ) + u_y \frac{\partial}{\partial y}(\ ) + u_z \frac{\partial}{\partial z}(\ )$$

u is the velocity vector
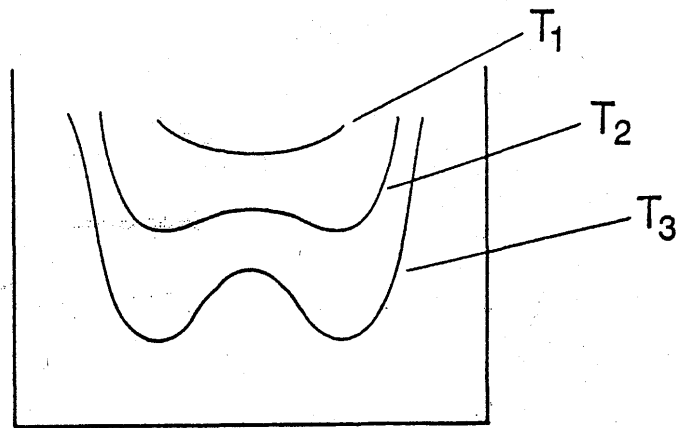
t is time

ρ is density

τ is the stress tensor

g is acceleration due to gravity; and

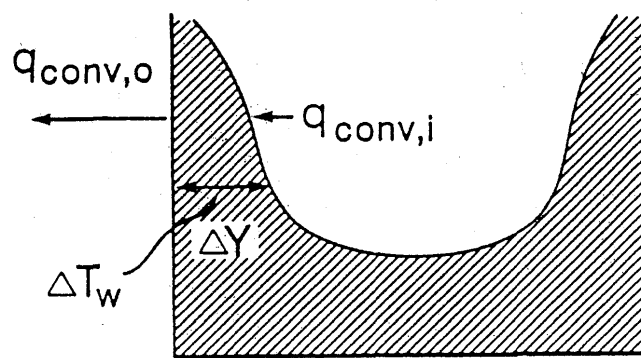F_b is the electromagnetic body force.

together with the thermal energy balance:

Figure 14   Some important effects of convection in

T_1

T_2

T_3

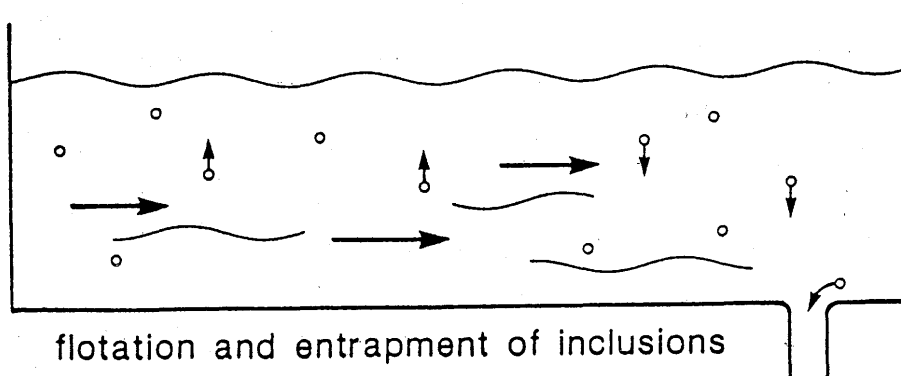## convection modifies isotherms

(a) modifying the temperature fields in melts

$q_{conv,o}$

$q_{conv,i}$

ΔY

ΔT_w

skull formation

$$q_{conv,i} = \frac{k_w \Delta T_w}{\Delta Y} = q_{conv,o}$$

(b) affecting skull formation

flotation and entrapment of inclusions

(c) affecting the flotation and entrapment of inclusions

$$\rho \, C_\rho \, \frac{DT}{Dt} = \nabla k \nabla T + \dot{q}''' \qquad (3)$$

where:

T   is temperature

C_ρ is the specific heat

k   is the thermal conductivity; and

$\dot{q}'''$ is the rate of heat generation.

We note that Equs (1-3) are coupled, because the velocity terms appear in the heat flow equation and in case of natural convection the temperature gradients drive the flow. Furthermore, in order to complete the statement of the problem we need to specify or obtain solutions for:

- the electromagnetic force field (through the solution of Maxwell's equations)

- surface tension gradients, in case of surface tension driven flow

- and the boundary conditions in general.

In principle these solutions may be obtained using various software packages, e.g. PHOENICS, FLUENT, FIDAP, TEACH or "homemade" differential equation solver subroutines (21,22). Many such solutions have been published in the literature; some recent symposium volumes (23,24,25) are a good source of information on results of this type.

As an illustration Figure 15 shows a comparison between experimental measurements and theoretical predictions for an electromagnetical casting system (26). This set of plots is interesting, because it clearly illustrates the level of sophistication reached in modelling systems of this type. More specifically, it is seen that the position of an electromagnetic shield will play a key role in affecting the velocity fields in the system and that predictions and measurements agree very well. This work has important practical implications, because for the satisfactory operation of the system a critical shield position must be employed, which provides both an adequate restraining force and at the same time minimizes the melt velocity perpendicular to the free surface.

Figure 16 shows the computed temperature profiles in a steelmaking tundish, both in the absence and in the presence of auxiliary heating (27). Generically, this latter class of problems is quite similar to the cold hearth processing system, as is the quite well studied Hall-Cell Simulation problem (28).

One should mention on a cautionary note that the use of computational packages and that the solution of two and three dimensional partial differential equations requires considerable experience, as does the intelligent interpretation of the results.
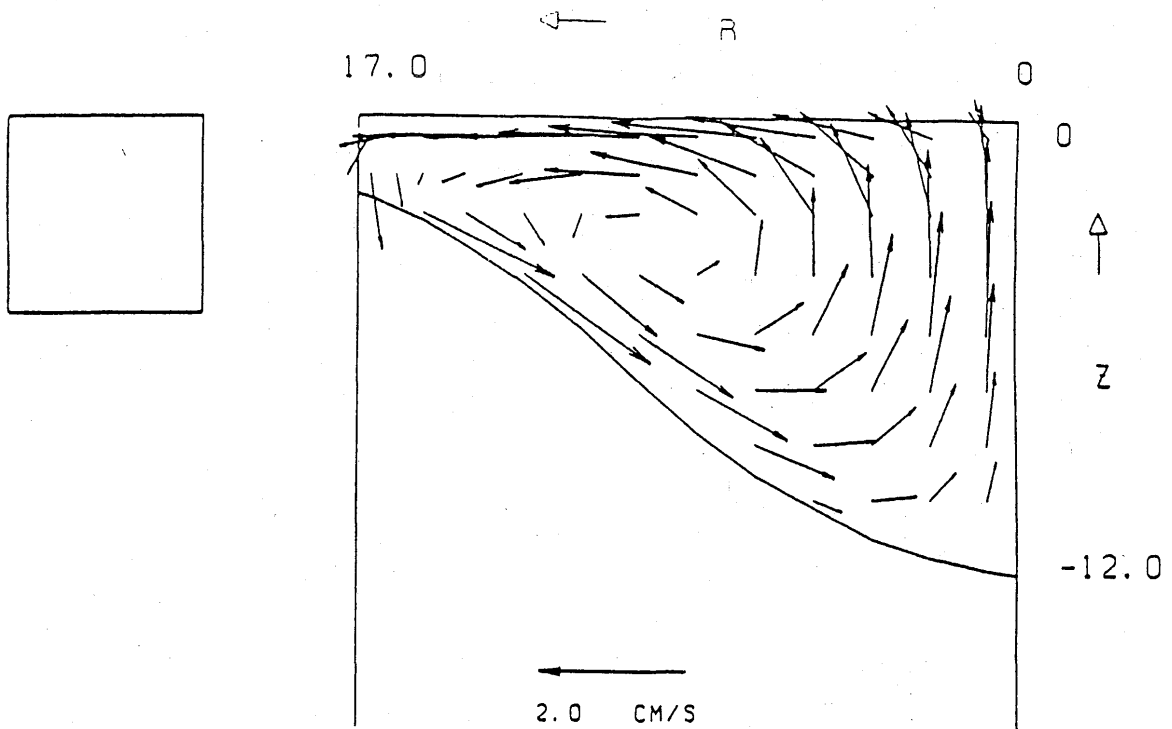
17.0

R

0

0

Z

-12.0

2.0 CM/S

Figure 15   A comparison of the theoretically predicted (upper part) and the experimentally measured (lower part) velocity fields in a model of an electromagnetic casting system (36). The successive figures show the effect of different electromagnetic shield position.
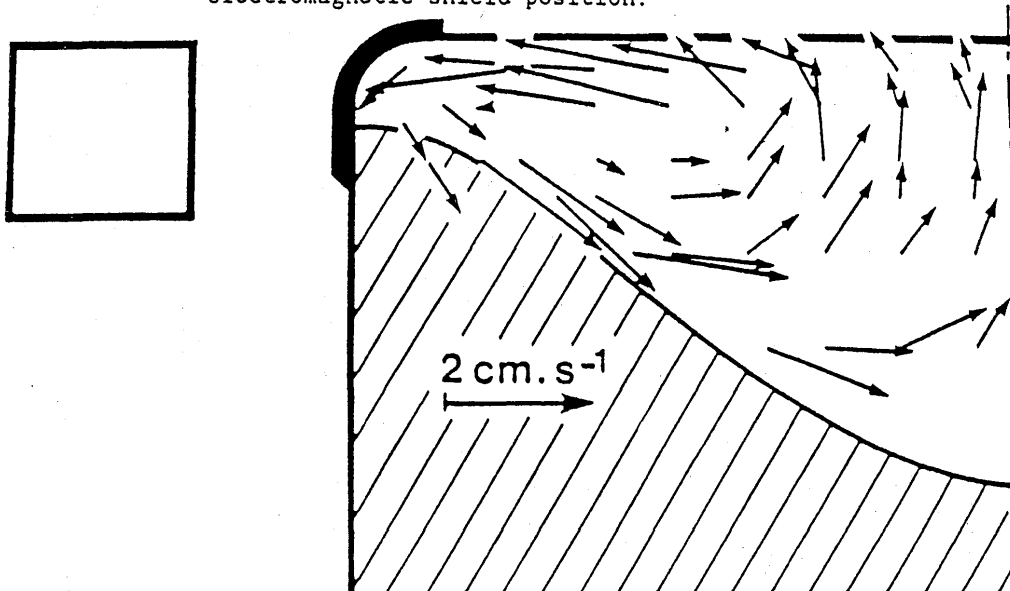
2 cm. s⁻¹

Figure 15A

17.0

R

0

0

Z

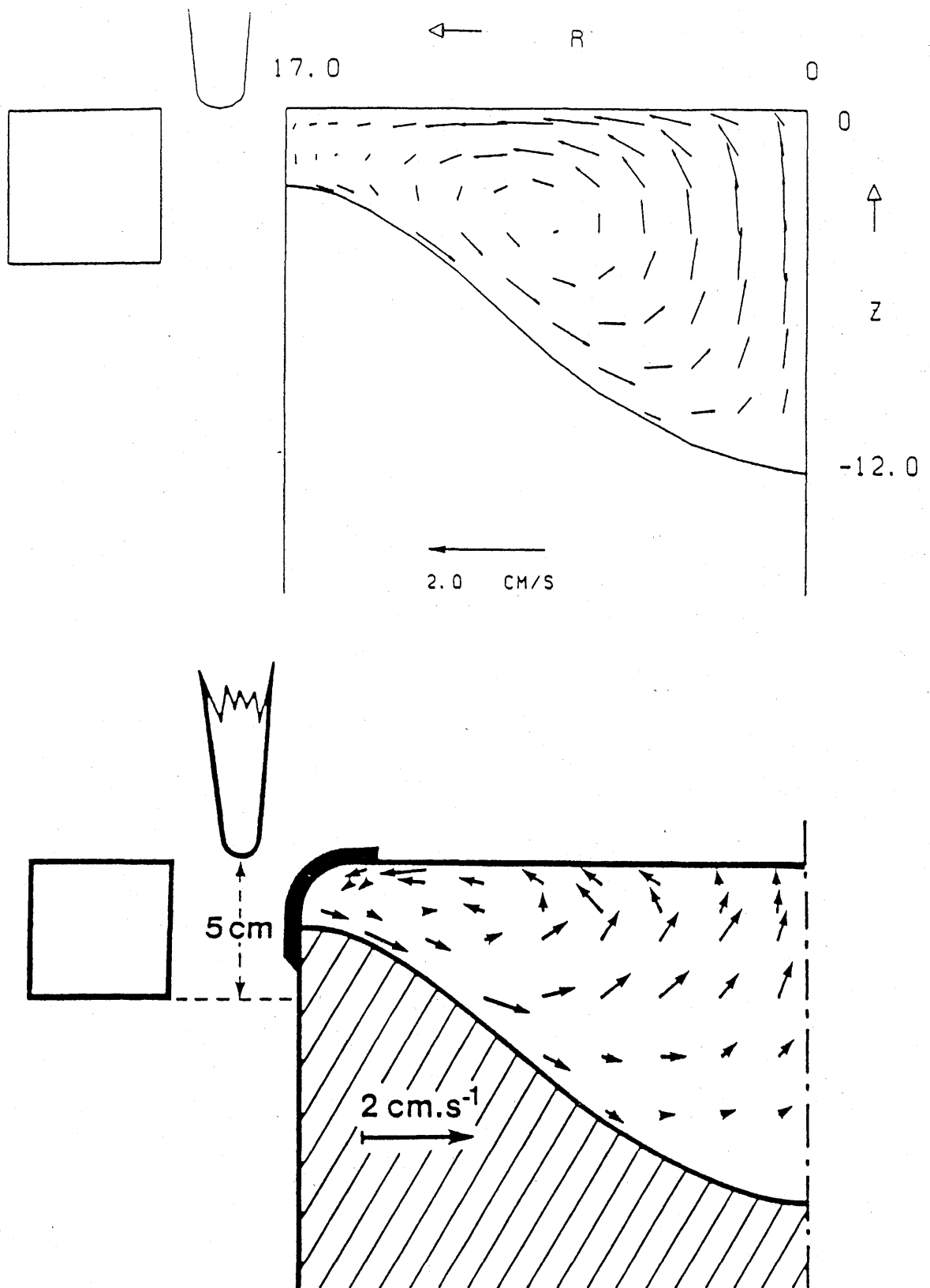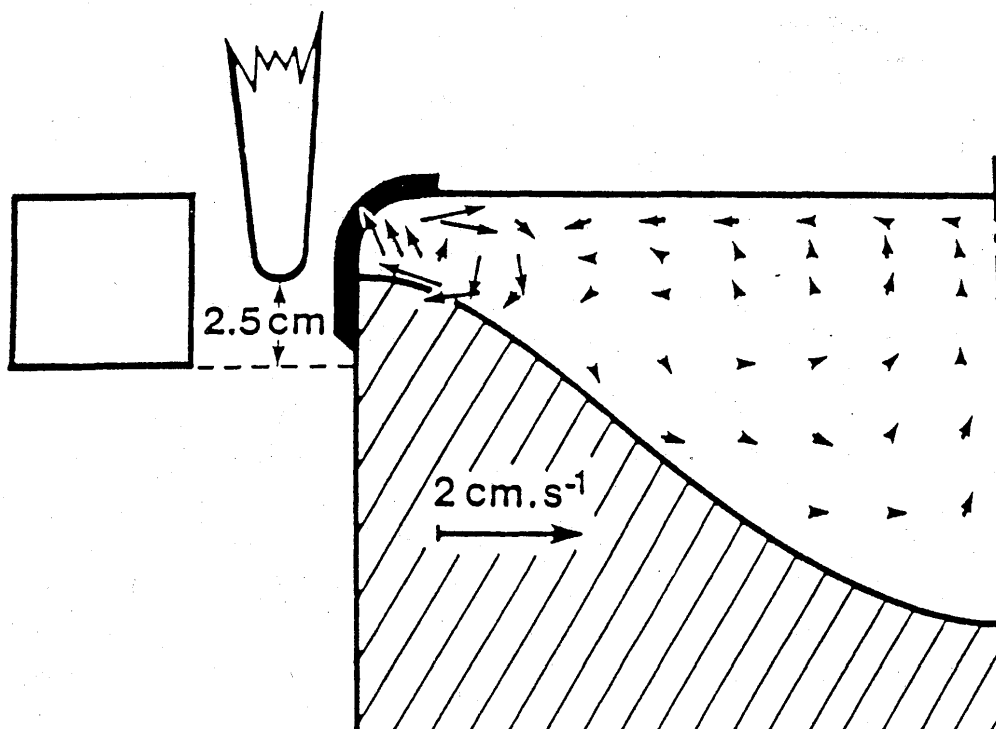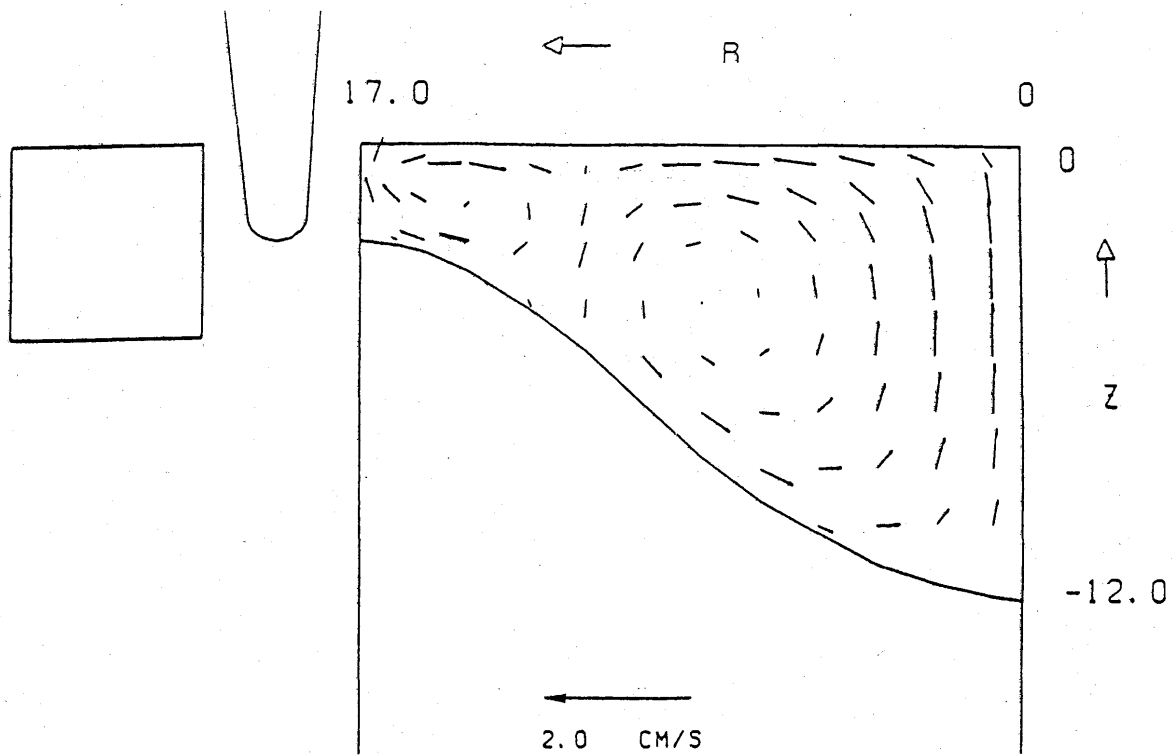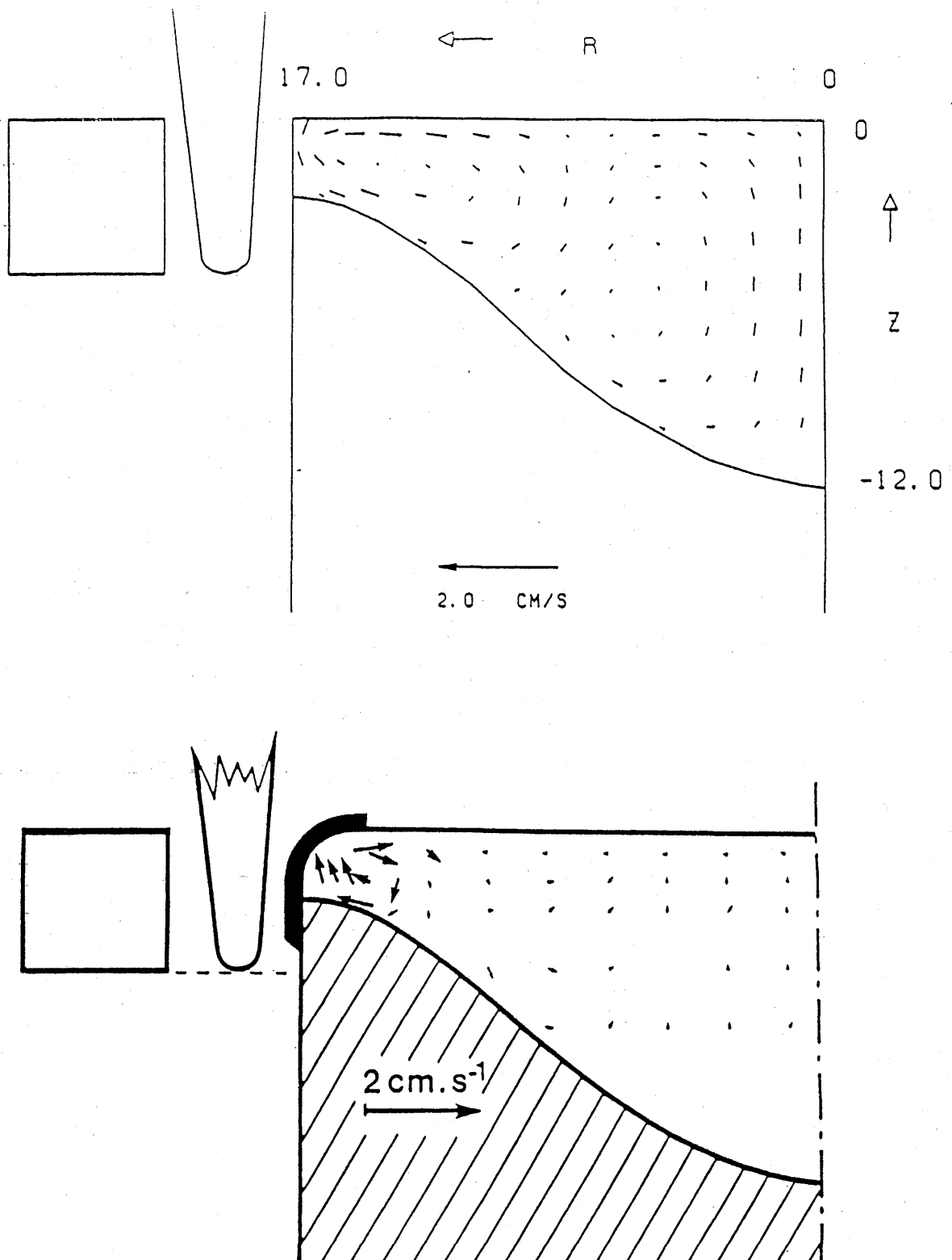-12.0

2.0 CM/S

5 cm

2 cm.s⁻¹

Figure 15B

Figure 15C

Figure 15D

Some recent work, carried out in our laboratory (29), modelling the behavior of shallow tundishes (e.g. about 1 m long, 0.5 m wide and a few cm deep), is even closer to the cold hearth refining problem. These calculations have shown that the flow behavior of these shallow pools may be markedly affected by the positioning of the inlet and the outlet streams. Unless considerable precautions are taken, recirculating flows may occur, causing by-passing and short circuiting.

Simple scoping and scaling (sometimes called back of the envelope type) calculations, with a good knowledge of transport phenomena can often provide very valuable insights. As an example, let us consider a plasma fired cold hearth refining process for titanium, where the hearth is some 40 cm long, 23 cm wide and about 2 cm deep, with a processing rate of 180 kg/hr.

The mean linear melt velocity, due to this bulk motion, is quite small, of the order of mm/s; at the exit, or at the pouring lip the velocity will be higher, perhaps of the order of tens of mm/s.

However, flow will develop in the system due to other driving forces, thus we will have:*

- flow due to thermal natural convection, driven by the difference in the temperature between that of the frozen crust and that in the bulk. This may be estimated to be of the order of tens of mm/s

- electromagnetically driven flow, due to the passage of a divergent current through the melt, which again may be of the order of several tens of mm/s or higher; and finally

- flow driven by surface tension gradients, caused by the temperature gradients at the free surface. These flows can be significant because of the shallowness of the bath and may reach values as high as several hundred mm/s.

Two immediate conclusions have to emerge from these simple considerations:

(1) Various recirculating flows will exist, the magnitude of which will overwhelm the bulk flow of the material through the hearth. It follows that the overall flow pattern may vary a great deal depending on the design detail and that the nominal residence time of the metal in the hearth may differ significantly from the actual residence time. In other words, short circuiting and by-passing are likely to occur. The use of dams or baffles, e.g. a slag dam which is employed in some designs, should be helpful in providing flow control.

(2) From a mathematical modelling standpoint, this will be a stiff or ill-posed problem, which might give difficulties with the computation.

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* These considerations have been developed on the analogy of problems in welding.

Nevertheless, there appears to be a substantial, as yet untapped, resource "out there" which would enable us to address many of the melt flow problems in specialty metals processing in a very rapid and cost-effective manner.

3.3 Atomization

One of the major developments in near net shape processing has been the atomization of molten metal streams, and their controlled deposition to produce the desired preforms. Several such processes exist today, the best known being the Osprey (30) system. A critical issue in these operations is to produce small, say 50-100 micron, liquid droplets of uniform size. This is usually done by impacting the molten metals streams by high velocity gas jets, such as sketched in Figure 17. While the principles of atomization are quite well understood, and may be explained in terms of hydrodynamic stability theory (31,32), the design of atomization equipment is still being done largely empirically. There seem to be outstanding, as yet not fully explored opportunities here for a more extensive use of computational fluid dynamics for atomizing nozzle design. Once the atomized droplets are formed, their interaction with the carrier gas is well understood and can be readily represented (33).

3.4 Solidification Processing

The understanding of structure-property relationships and the identification of the appropriate phases, has been one of the mainstays of specialty metals processing. Controlled solidification in ESR, VAR and the development of directionally solidified alloys and single crystal turbine blades all owe their existence to a good fundamental understanding of solidification theory.

The advent of rapid solidification, that is the ability to produce microcrystalline solids, by employing cooling rates in the region of 10^4-10^5°C/s (and the subsequent consolidation of these materials by HIP), provided a major advance in materials technology. The cooling and undercooling phenomena that occur while these droplets travel with the carrier gas have been extensively studied and are quite well understood.

The more recent technology of spray forming is perhaps less well studied, although an excellent qualitative picture has been drawn in an earlier paper by Singer and Evans (30). It has been suggested by these authors that two basic physical situations may occur, as illustrated in Figure 18:

(i) At low spray rates (a) and also for small droplet sizes, the droplets hitting the solid surface spread very rapidly (over a matter of microseconds), and then solidify, over a period of milliseconds, before the arrival of the next droplet. Under these conditions very fast cooling rates (or very short solidification times) are obtained, giving very fine grains, say around or below 10 microns.

(ii) At high spray rates (b) solidification is not completed before the arrival of the next droplet at a given site. Under these conditions, as analyzed by several authors (37), a thin liquid film will form at the top of the deposit and the cooling rates will be much slower. Such an arrangement will give a rather larger grain size, say several tens of microns.

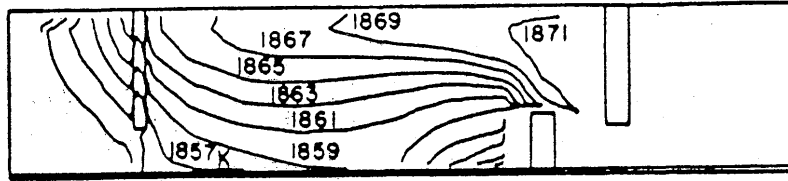The transition from (i) to (ii) is not quite obvious in many applications,

Figure 16   The computed temperature profiles in a steelmaking tundish, 7

m long, 0.85 m wide and 0.65 m deep

(a)   in the absence of external heating

(b)   in the presence of external heating

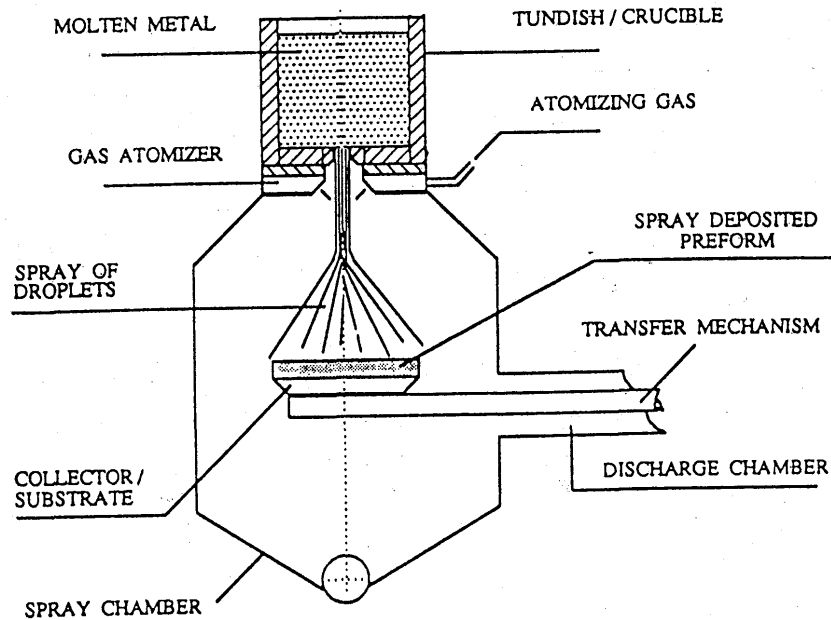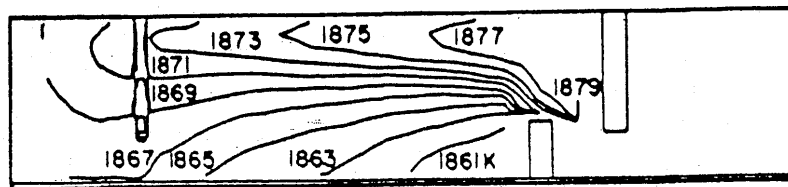
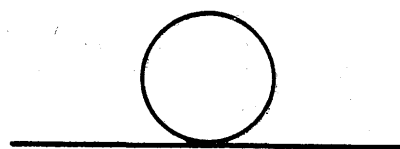Figure 17   Schematic   sketch   of   a   metals   spraying   system,   original

picture kindly supplied by Professor Apelian of Drexel

University

Figure 18   A schematic sketch showing the impact and subsequent
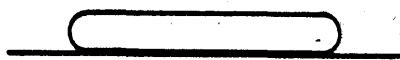solidification of

(i) one droplet on a solid surface
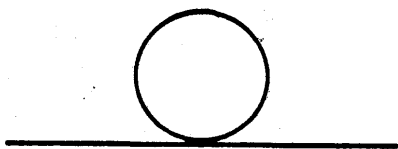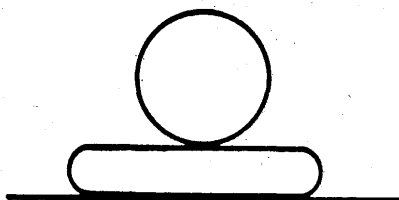
(a)

(b)

(c)

(d)

(ii) two subsequent droplets, with the second impacting
before the first had an opportunity to solidify

(a)

(b)

(c)

(d)

which may account for the disparity in the grain sizes observed and the difficulty in predicting the experimentally measured grain sizes.

The spray forming technique is a very attractive avenue for the near net shape processing of specialty metals; mathematical modelling and some related basic process studies should help identifying the optimum combination of the many process parameters, such as superheat, substrate preheat, gas flow rate, deposition rate, scanning arrangements and the like.

Other forms of near net shape processing such as the direct casting of plates, thin slabs and sheet, has aroused a great deal of interest in the steel industry, and some major advances are being made in the casting of stainless steel strip (12). However, the "process science" aspects of this technology are rather less well explored.

4. Discussion

In this paper we sought to present a brief review of some of the new developments in the melting, refining and solidification processing of specialty metals.

Technological change is inevitable and is proceeding at an ever increasing pace; thus even some of the "far out" new ideas such as cold crucible melting, the various cold hearth processing operations and spray forming may well become part of the accepted technology in the near future.

In terms of identifying the "best bets", there is a clear need to provide molten metal feedstock of progressively greater purity - which should favor skull melting techniques. Induction and plasmas may turn out to be more cost effective than electron beams in many applications.

In solidification processing, the direct spray forming techniques would seem to offer several advantages, including fine solidification structure and a reduction in the deformation processing steps.

The third, perhaps equally important step in specialty metal processing is refining. Once a charge has been melted, so that outside contamination has been minimized, there could be an opportunity of removing impurities contained in the original charge by the addition of reagents or gas purging under vacuum, all of which could benefit from mixing or the careful regulation of the flow patterns within the system. Some of these ideas are now being discussed (1), but probably a great deal more could be done by fully implementing concepts from ladle metallurgy and secondary steelmaking.

The implementation of these new processing ideas will require more process research and the effective utilization of modelling techniques.

At the present modelling appears to be a very attractive option, because of the ready availability of computing hardware and software. As discussed in the preceding sections of the paper, some of these modelling tasks could be quite routinely undertaken, using readily available computational techniques and methodologies.

Problems within this category include induction furnace design, the study of some aspects of heat flow, fluid flow and electromagnetic phenomena in cold

hearth remelting, electro-slag refining and casting and the continuum aspects of spray forming. In these areas modelling could play a major, highly cost effective role in process optimization.

Yet other problems, which will require an improved science base, include the fundamentals of atomization, plasma-melt interactions, EB-melt interactions and the microscopic and structural aspects of spray forming. Here basic research needs to be done before purely "mechanical" computational efforts will become appropriate.

In closing, one should reiterate that we are living in a rapidly changing world with quite fierce competition both between different materials and the producers of the same types of materials. Some of the new technologies discussed in this paper will be adopted at a fairly rapid pace, indeed the survival of many producers may well depend on their ability to adapt. Over the longer haul, it is quite likely that structural ceramics and metal-matrix composites may replace some or a sizable fraction of the specialty metals currently in use. The rate at which these changes will come into play will, of course, be greatly affected by the changes in product and process specifications. New alloy development and perhaps more important new, more efficient processing technologies of the types we discussed should help the competitive position of the specialty metals producers.

The development of a better process science base and the more extensive use of modelling could play a major role in these developments, both with regard to maintaining the competitive position of metals and in helping with the transition into other materials, such as composites.

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