Sixty Years of Casting Research

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The 60 years of solidification research since the publication of Chalmer's constitutional undercooling in 1953 has been a dramatic advance of understanding which has and continues to be an inspiration. In contrast, 60 years of casting research has seen mixed fortunes. One of its success stories relates to improvements in inoculation of gray irons, and another to the discovery of spheroidal graphite iron, although both of these can be classified as metallurgical rather than casting advances. It is suggested that true casting advances have dated from the author's lab in 1992 when a critical surface turbulence condition was defined for the first time. These last 20 years have seen the surface entrainment issues of castings developed to a sufficient sophistication to revolutionize the performance of light alloy and steel foundries. However, there is still a long way to go, with large sections of the steel and Ni-base casting industries still in denial that casting defects are important or even exist. The result has been that special ingots are still cast poorly, and shaped casting operations have suffered massive losses. For secondary melted and cast materials, electro-slag remelting has the potential to be much superior to expensive vacuum arc remelting, which has cost our aerospace and defense industries dearly over the years. This failure to address and upgrade our processing of liquid metals is a serious concern, since the principle entrainment defect, the bifilm, is seen as the principle initiator of cracks in metals; in general, bifilms *are* the Griffith cracks that initiate failures by cracking. A new generation of crack resistant metals and engineering structures can now be envisaged.

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I. INTRODUCTION

ONE of the central tenets of metallurgy has been that for metals "the microstructure controls the properties." At worst this is not true, and at best it is a half truth. More often, the history of cast metals confirms "defects (actually bifilms) control the microstructure and properties." This alarming and revolutionary proposition has yet to be universally accepted. However, it is quite obviously true: even rudimentary experiments to determine strength and elongation of cast metals using simple (badly designed) test bar molds show scatter which usually greatly exceeds the effects of conventional metallurgical efforts such as alloying or heat treatment. The scatter may be from porosity, but is more often from defects which are not easily seen; bifilms. These features are natural products of turbulent transfers and stirring operations.

Casting is a complex manufacturing process, and is influenced not only by conventional metallurgy such as alloying and heat treatment, but also by the manufacturing hardware such as the plant and the processes because of their major involvement in the generation of different populations of bifilms. Some effort will be made to look at the influences of all these factors. However, the central role of casting defects controlling structure and properties represents a major revolution in metallurgical thinking and is emphasized in this review. A few preliminary examples are listed below from (Reference 1).

II. MICROSTRUCTURE AND PROPERTIES

1. The benefits of reduced grain size in Al alloy castings appear to be mainly the effect of an action in which the melt is cleaned from bifilms, rather than any benefits from the Hall–Petch relation. An example is given below.
2. Grain size itself is sometimes controlled by bifilms, large bifilms from turbulence in the mold can cause large grains by interlacing the melt and suppressing thermal convection, thus suppressing the normal processes of dendrite arm remelting which would normally lead to finer grains.
3. The traditional attribution of improved properties by chilling to create refined secondary dendrite arm spacing (SDAS) has been proven to be merely a provision of addition time for bifilms to unfurl, effectively growing as cracks, and thereby reducing properties.
4. The effect of Fe in reducing the properties of Al alloys is seen to be the result of the straightening of bifilms by the growth of beta-iron particles on them. In contrast, the benefits of Mn to reduce the effect of iron are explained by the conversion of the iron-rich intermetallics to the cubic lattice alpha-iron phase which has sufficient symmetry to enable it to grow around the initially convoluted bifilm

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and so prevent it unfurling to become a serious engineering type of crack.

5. The modification of Al-Si alloys by Sr is explained in detail for the first time, as a result of the deactivation of bifilms as substrates for Si by the addition of Sr, converting the Si phase from a cracked, irregular plate-like growth (known as an unmodified eutectic structure) to a fine classical coupled eutectic which we know as a 'modified' structure.[1–3]
6. The conversion of flake graphite cast iron is similarly explained as a conversion from growth of graphite on bifilms in suspension in the iron, to the removal of bifilms by reduction with magnesium addition, leaving only compact nuclei around which graphite can now grow as spherical nodules.[1,4]
7. The dramatic effect of properties, particularly elongation and fatigue, on bifilm reduction by simple mechanical techniques such as a dwell time after melting, allowing bifilms to sediment, and avoiding subsequent pouring of the melt, cannot be explained by conventional metallurgy, but is now widely practiced, appearing successful for nearly all engineering metals.[1,5]

III. METALLURGICAL ADVANCES

Mg alloys enjoyed the major advance associated with the introduction of Zirconium,[6] first successfully achieved in Germany in 1947 (just prior to Chalmer's announcement of his constitutional undercooling theory; these were exciting times!) Zirconium metal was an efficient grain refiner, so that significant benefits to strength and toughness resulted from the Hall–Petch effect[1] as a result of the basal plane being the only effective slip plane in these alloys (in contrast Al alloys, being face centered cubic, had a profusion of slip planes, so that boundaries provided almost no hindrance to the propagation of slip to a new grain. Thus, grain refinement has only a weak benefit.). Interestingly, the most successful technique for the introduction of Zr was *via* a fluoride-based salt which had to be stirred into the melt, then allowed to settle to the bottom of the crucible (as a result of its high density conferred by its content of $BaCl_2$). It seems probable, therefore, that an important, but as yet unrecognized, contributor to the property benefits would be the reduction in bifilms by their absorption into the flux.

The Al alloys have seen some modest advances associated with increasing purity of alloys available, in particular with low iron contents. The alloy originally called KO1, but now A201, containing approximately 5 Cu and 0.7 Ag has yielded excellent tensile properties, but has been plagued by its difficulty to be cast without defects. Thus, casting issues have hampered the development of this remarkable alloy. Its near cousin containing no silver, A206, is nearly as good but similarly problematic. Both are sensitive to the impurities Fe and Si, almost certainly as a result of the precipitation of beta-Fe and Si intermetallics straightening the cracks intrinsic in oxide bifilms, since both tend to grow on bifilms and both adopt a planar growth habit.[1]

The development of Al-Li alloys, started in the late 1950s, heralded the first significant advance in specific stiffness in a metal alloy, since the great volume of rigid Li-rich compounds both reduced the density of the alloy and increased the stiffness.[7] However, it is interesting that some major users of the aerospace industry have been reluctant to take up these alloys for their intended purpose as wing structures because of minute cracks which have been observed around rivet holes, even though the analysts affirm that the cracks are of a subcritical size and therefore will not spread. The cracks will have originated from oxide bifilms, as a result of the less-than-perfect metal treatment and casting processes.

The expansion of work on partly solid mixtures has grown exponentially since the discovery of rheocasting and its cousins by Flemings and co-workers in 1972.[8] The introduction of particulates into metallic matrices has expanded beyond the scope of this paper to address them. However, some general comments are appropriate. The casting of slurries, in contrast with the casting of liquid metals, appears to favor a relatively non-turbulent flow regime which has reduced the entrainment defects when pouring or injecting. This significant advantage is, unfortunately, offset by the introduction of non-wetted oxide films into the melt during the creation of the solid/liquid mixture by solid additions and/or oxide bifilms introduced by the stirring processes. This natural population of bifilms in most solid/liquid mixtures will have the benefit of regularizing properties, and possibly even generating some strength benefit as an MMC. They may therefore generate properties improved compared with conventional metal castings. Ultimately, however, the maximum attainments of practically all of these interesting composite materials will be limited by the presence of bifilms. In comparison, advanced liquid casting techniques in which bifilms can be eliminated offer the promise of ultimate theoretical properties which should, in principle, be attainable.

Melt treatments have played an important part of light alloy casting. The use of filters has in general been very beneficial, the ceramic foam filters becoming available for the first time in industrial quantities in the early 1980s.

Rotary degassing techniques followed quickly in 1990 which have purported to deal with the hydrogen content of liquid Al alloys. They have undoubtedly improved properties of castings, although there are good reasons to believe that these properties are far from optimized. Development effort has concentrated on the reduction of hydrogen content, whereas it seems the far more important action in the elimination of primary oxide skins from charge materials. Unfortunately, although eliminating the relatively few but large primary oxide bifilms, the process clearly replaces them by millions of small, new oxide bifilms, as confirmed by theoretical expectations and by the sharp change of chemistries of the oxides, from spinels to pure alumina as the process proceeds. Rotary degassing is therefore a process currently optimized inadequately, and requires to be further developed or possibly abandoned.

The wide adoption of Sr additions to Al-Si alloys has also been accompanied by mixed benefits as a result of the neglect of bifilm issues.[1–3]

The action of Ti additions (together with C or B) to Al alloys again appears to have been effective as a grain refiner, but has a negligible effect in raising mechanical properties (although Al-Si alloys are exceptional as a modest beneficiary). Once again the increased properties from Ti additions have been shown to result from the cleaning of the melt from bifilms as a consequence of the heavy $TiAl_3$ precipitating on the bifilms in suspension in the liquid, and sedimenting these to the bottom of the crucible prior to pouring. If the bifilms are mixed up once again by stirring immediately prior to pouring, most of the benefits of the addition are lost.[1] This effect is clearly seen in the work of Katgerman and colleagues[9] who found that grain refiner added to the ladle both grain refined and eliminated cracks in a high-strength Al alloy, but when added into the flowing stream of metal into the continuous caster resulted in beautifully grain refined but cracked billets.

The cast irons have a venerable engineering history, but have enjoyed a number of extraordinary revolutions. An early development dating from the 1920s was the process of *inoculation* in which the flake graphite was controlled in size and shape.[1] The next, probably more famous event was the discovery in 1943 and announcement in 1948 of the discovery of spheroidal graphite iron, appropriately known in the States as ductile iron.[10] More recently, the realization that compacted graphite iron, known for years but essentially unused, had useful properties intermediate between flake and ductile irons developed into a significant engineering class of irons. More recently still, the development of austempering has resulted in irons with strengths measured in GPa, exceeding those of many steels. This succession of metallurgical developments in cast irons is a rarely paralleled success story. Only in 2009 was a theory attempting to explain the structure of cast irons proposed. It was based on the role of silica bifilms as substrates for the formation of graphite.[1,4]

Steel castings have enjoyed the benefits of independent control of carbon and oxygen, and the lowering of hydrogen and nitrogen by argon-oxygen-degassing (AOD) treatment.[11] Even so, the use of the newly developed naturally pressurized filling system for shaped castings has illustrated that AOD might be redundant if melts can be cast sufficiently free from bifilms by careful design of the filling system of the casting.[12]

A. *Engineering Developments*

The machinery to make molds has seen a steady increase in molding speed; the record currently held by the Disamatic machines from Denmark for a mold every 10 seconds as a result of them using both the front and back of the mold to contain part of the mold cavity, and arranging the molds in a continuous line, each front to back. The other advance in the fast greensand molding technique is the use of shock consolidation of the mold to create uniformly dense molds. In this way, the reproducibility and accuracy of castings have greatly improved.

The vertical parting plane in the Disa molds allows, in principle, the molding of a good nicely formed series of filling channels. In practice, at this time, only disgracefully poor filling systems are generally employed. For most of the other high production greensand molding plants, the filling system design has been given no priority in mold production, with the result that such plants often struggle to achieve a reasonable quality of castings and struggle to control a tolerable scrap level.

The chemically bonded sand molds dated from about the 1970s achieved high accuracy of cast products. This was further enhanced by the appearance in the 1980s of gas curing systems in which the hardening of the core or mold could be carried out in situ in the mold and core tooling (*i.e.*, the patternwork). In the first decade of 2000, the silicate binders were newly formulated as environmentally friendly, and yet efficient and economical. These sand binders represent a major step forward in creating user-friendly casting environments, while sometimes bringing additional benefits such as solubility in water, allowing sand reclamation systems to be revolutionized.

B. *Filling System Design for Gravity Casting*

There have been multiple efforts over the years to develop rules for the design of gravity filling systems for shaped castings. Nearly all of these have suffered the fundamental problem of pouring into a cone-shaped funnel, and so aspirating air.[1] All subsequent variations of the filling channels introduced downstream of this quality-destroyer were overwhelmed by the damage introduced by this disastrous first stage of the filling system design. This failure illustrates a general explanation of all this early work; it was not clear to experimenters that every stage of the progress of the melt through the filling channels had to be correctly designed. Any feature at any stage which introduced air would cause the whole system to fail.

For this reason, offset step basins, which work as well or better than any other design of filling basin, have been routinely abandoned because they did not appear to work. The reality was that they had no chance to reveal their benefit to a filling system because the remainder of the filling system was so bad as to overwhelm and conceal any benefit.

The widespread use of wrongly tapered sprues has been tolerated simply because experiments which have been performed with difficulty on automated plants to provide correctly tapered sprues have shown little or no benefit. Once again, the benefits were overwhelmed by the rest of the badly designed system.

A somewhat similar history goes with the universal use of the ‘well’ at the base of the sprue. Many experiments carried out in the US and UK over many years using cine photography and water models have been poorly optimized because the sprues and runners have been oversized to a degree, so far from optimized, that an optimization was fundamentally impossible for these systems. The subsequent adoption of wells, in spite of the poor experimental results, was justified by the use of comforting language such as the well providing a 'cushioning' effect for the metal as it impacts the base of the sprue. Clearly, it is easy to be influenced by the words we use to describe physical phenomena, despite all the facts.

The author set up the Cosworth Process Foundry in 1978 for the casting of cylinder blocks and heads for the Cosworth racing engines. This was the first foundry world-wide to use an electromagnetic pump as the sole production technique for filling molds.[5] Some powerful lessons were learned by this experience. Initially, the foundry was set up in two halves:

(i) The first half made castings by traditional pouring by gravity;
(ii) The second half produced castings by only uphill (counter-gravity) filling using the electromagnetic pump.

For both halves of the foundry, the alloys were identical, the sand and binder identical, heat treatment identical, both sets of cast products were substantially free from porosity, and appeared perfectly sound by X-ray radiography, but their properties were totally different. The gravity castings leaked, but the counter-gravity castings did not. By pouring with gravity, it was a difficult to make a good part, whereas by counter-gravity, it was difficult to make a bad part. Furthermore, for uphill cast cylinder heads on the engine test bed, failures by thermal fatigue dropped from 50 pct to zero overnight and fatigue properties from the uphill cast aerospace castings exceeded the highest fatigue resistance of all other aerospace foundries in Europe at that time. Clearly, these different behaviors were not the result of *metallurgy*, nor *solidification*, but only the result of *casting*.

These were substantial differences in materials which, from the same foundry, would normally have been expected to be identical. Incidentally, the gravity pouring side of foundry was closed after about 3 months, and subsequently, all castings were transferred to the counter-gravity system. The lesson was never forgotten. The powerful benefits of counter-gravity casting were to be explained later by research at the University of Birmingham. This work introduced the concept of *surface turbulence* and quantified the concept by Weber and Froude numbers which led to the related concept of a *critical ingate velocity* in the region of 0.5 m/s.[13] These concepts were to revolutionize the production of shaped castings and ingots. Continuous casters have yet to convert fully to these concepts.

The concept of a critical ingate velocity explained why counter-gravity could be so successful: it was able without difficulty to provide filling at velocities not exceeding this value, whereas pouring with the liquid metal accelerated by gravity automatically accelerated the metal to unwanted high velocities, greatly exceeding the critical velocity, and providing sufficient energy to create masses of entrainment defects. The gravity pouring process was, and always would be, fundamentally different in performance from counter-gravity.

It was with the help of the video X-ray radiography unit that a detailed study of gravity filling system design resulted eventually with designs which limited the damage significantly. The latest improved designs of filling systems for gravity castings were called a *naturally pressurized* design. It contrasted with previous concepts of the so-called pressurized and non-pressurized designs. Although the naturally pressurized system was more than 10 times better than conventional designs in the numbers of defects created in the castings,[14] it was clear from this work that the damage created by gravity pouring could never be completely eliminated; this optimized gravity system was at best merely a damage limitation exercise.

The revolutionary conclusion from this work was that for the best castings, any pouring of metals could not be allowed. Only counter-gravity had the potential to produce perfect products.[15]

The attainment of perfection, once ridiculed as impossibly difficult, seems to be attainable with sufficiently well-designed melting systems and melt handling, teamed up with casting by some non-turbulent technique such as counter-gravity.[15] Furthermore, the corresponding attainment of ultimate properties of metals, never yet achieved, now seems achievable. Extremely high strengths coupled with extremely high elongations should be routine, as a result of the absence of bifilms, or bifilm residues in wrought metals. It seems there are sound reasons why bifilms represent the *only* natural source of cracks in metals; they constitute the *Griffith cracks* required for all fracture processes.[16] Without bifilms, it is predicted that a metal should never fail by cracking. All failures would be by plastic extension (but possibly at extremely high stress).

C. *Secondary Remelting Processes (VAR and ESR)*

For the highest duty steels and Ni-base alloys, they are remelted for at least a second time. There are many such processes, including such exotics such as plasma processing of various kinds. However, the two major processes are vacuum arc remelting and electro-slag remelting. Occasionally, the attribution 'remelting' is replaced, presumably for marketing reasons, by 'refining.'

Vacuum arc remelting has been the principle process for the highest duty applications almost certainly as a result of the association of the word 'vacuum' with quality. This unfortunate promotional advantage of the inferior process has cost the aerospace and defense industries dearly.

(Occasional specifications call for limitation of the hydrogen content, but after hot working to most sizes of engineering interest, hydrogen is easily able to diffuse out of the product within a few hours or days. Thus, the vacuum is essentially redundant from the point of view of usefulness in reducing hydrogen in most products. As a further aside with respect to hydrogen in metals, the embrittling effect has been widely researched, but remains not well understood; confusion in the experimental literature abounds. This is highly suggestive of a strong but essentially unsuspected role of bifilms, present in different quantities and qualities in different batches of materials.)

Huge development efforts have been made into VAR specifications for all the strongest steels, despite (i) the high costs of the process which often includes prior melting by ESR or other processes such as vacuum induction melting (VIM) to produce consumable electrodes, and (ii) the great costs of 'scalping' (the peeling off of its skin, being 20 pct or more of the volume of the ingot) to enable it to be processed by hot plastic working of various kinds. In fact, the VAR ingot requires to be scalped because of the oxidized laps, actually deep bifilm cracks, which characterize the surface of the ingot, and which lead to cracking during plastic working. As in nearly all industrial vacuum processing, the vacuum is not sufficiently good to prevent the oxidation of the surface of the liquid metal.

In contrast, ESR has no such oxidized lap problems, and a difficult alloy, such as the Ni-base Waspalloy, which cannot be forged after VAR without peeling, forges like butter with its as-cast surface when remelted by ESR. The fact that ESR has been downgraded with respect to VAR is because of its fatigue performance. However, this is hardly the fault of the ESR process but the fault of the grossly poor electrodes from which the ingot has to be made. The electrodes are commonly formed in tall ingot molds which are top poured. The entrainment defects in this electrode material can be seen by the unaided eye from a distance of at least 100 m. The defect density in the electrode overwhelms the remelting process, so that at least some of this defective material survives the remelting process. Sometimes, the extensive bifilms allow large chunks of the electrode to fall prematurely into the melt, creating large defects which are entirely uncharacteristic of this process when operated with an appropriately sound electrode. It is a tribute to the fundamental soundness of the ESR process that it can usually take such dreadful melting stock and transform it in a single remelting operation into a material competitive with VAR which has enjoyed the additional processes of prior refining of its electrode, often by ESR or VIM.

In an exchange of recent correspondence,[17] it has become clear that not a single comparison of VAR and ESR material appears ever to have been tested for the condition in which both processes have enjoyed the benefit of similarly well cast electrodes. If such a test had ever been made the ESR process would have clearly demonstrated its superiority over VAR and saved the world and its high-tech industries untold fortunes. Both would benefit from electrodes produced from simple bottom gated ingots in which air had been excluded from the filling system by contact pouring, plus, preferably, correct geometry of downsprue, runners, and gates.

One of the advantages listed for VAR is its ability to produce castings of large diameter but avoiding the creation of channel defects as a result of its higher temperature gradients. Once again, this is hardly an excuse for selecting VAR over ESR. ESR ingots can be produced in a continuous fashion cast into short collar molds with the ingot exposed below the collar directly cooled by impinging water jets. If the presence of the solidified slag layer is found to inhibit heat transfer significantly, it is not difficult to think of mechanical techniques for eliminating this layer to gain the maximum benefit from water impingement direct on to the ingot surface. The temperature gradients achievable by ESR would then exceed those attainable by VAR. For the occasional requirement of a hollow ingot, ESR can produce such a shape as-cast by use of a central water-cooled mandrel. In this case, the attainment of a high temperature gradient to suppress channel defects is once again solved. There is no excuse for the adoption of VAR over ESR. This has been an expensive mistake.

D. *Ablation Casting*

Ablation casting[18,19] is the name given to the new casting process which aims to have solved much of the standard casting problems suffered by the industry up to now. The mold is made from an aggregate (not normally silica sand) bonded with a water-soluble binder which is ablated (eroded) away by water jets after the mold is filled with liquid metal. In this way, for the first time, the air gap limitation to cooling is avoided since there is direct contact between the cooling water and the casting. Also, for the first time, the mold only defines the casting shape, but in an ideal case will provide no cooling. Cooling is independently controlled by the casting engineer, who plans the cooling pattern and rate of cooling to match the geometry of the casting; heavy bosses and sections are given longer to solidify prior to moving the cooling to thinner sections. The process, being water based, is free from dust, fume, smoke *etc*, is a pleasant, highly productive working environment, and delivers castings with excellent properties and good economy. It is currently subject to patent cover. It promises to be an important development, typical of what we might expect for future casting operations.

E. *Bifilm Reduction Technology*

Both the aluminum and steel industries are making some headway to achieve cleaner melts and less turbulent casting processes. The properties are seen to be improving steadily in both of these industries.

However, one of the most spectacular demonstrations of the benefits of nearly zero bifilms is seen in the example of Ni alloy single crystal turbine blades. All blades, whether polycrystalline or single crystal, are currently cast using relatively poor gravity casting techniques which ensure a plentiful population of bifilm cracks in the casting. In the case of the polycrystalline blade, the rapid growth of grains from all directions traps the bifilms in place, keeping properties down. In the case of the single crystal, the slow rate of freezing, taking perhaps 2 hours or more, allows many bifilms to float out. In addition, those that remain are pushed by the advancing dendrite array, so the final casting is relatively free from bifilms and enjoying spectacularly good reliability and creep properties.[15] (The old explanation of the elimination of weak grain boundaries has been shown to be false from recent molecular dynamics simulations). If melting and casting techniques were improved by relatively simple and low cost methods, then the Ni alloy would be expected to solidify with even better properties than the single crystal, even as a polycrystal (of course it would not exhibit the special directional properties), but could be produced at a fraction of the time and cost. The single crystal itself could be produced more quickly, at reduced cost, and with improved properties.[16] The huge promise of bifilm-free metallurgy has yet to be explored.

IV. CONCLUSIONS

1. The development of casting has lagged behind the development of metallurgy and the study of solidification as a result of investors and researchers being unaware of the control that casting defects exert on both the structure and the attainable properties of both cast and wrought metals. This lack of awareness of the profound controlling nature of casting defects has hampered the development of casting, which in turn has hampered the whole of mechanical and civil engineering, costing foundries, and the engineering world untold billions in poor attainment and failures.
2. In summary, it is concluded that the familiar mantra the properties of metals and alloys are controlled by their structure is partly illusory, since both the structures and properties of cast metals (and many resulting wrought metals) are mainly, if not wholly in some cases, controlled by bifilm defects.
3. Although some commendable and encouraging initial efforts have been made, properties in the absence of bifilms have yet to be fully demonstrated in metallurgy.

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