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Recent developments in the design and construction of the cold crucible*

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The concept of the cold crucible is critically re-examined. In order to overcome some of the more serious limitations of the Hukin design, consideration is given to a return to the synthetic crucible; that is one put together from (identical) individual parts. The important features of such a crucible are compared and contrasted with those of one carved from the solid.

1. Introduction

The difficulties associated with the complete melting of the charge contained in a hot crucible are now well documented [1, 2]. Moreover Brice [1] has tabulated the properties of a wide range of materials commonly used to form crucibles, together with compatibility data for a range of refractory materials.

However, contamination problems at high temperatures are generally so severe that considerable effort has been directed toward the elimination of the crucible, as in the classical, pedestal melting technique [3] or its varations such as float zone melting, where intensive development has allowed the production of oxygen-free silicon crystal, routinely and on a commercial basis, with diameters up to 100 mm.

By way of contrast, several workers have contributed to the development of the cold crucible, (used in conjunction with a variety of primary power sources), and this work has been exhaustively reviewed by Jones [4]. However, it is still important to note that, in striving to satisfy the three principles proposed by Lawley [2] and so eliminate contamination problems, the concept of the cold crucible tends to violate the third principle, that of low thermal conductance of the crucible relative to the charge, which is deemed to be necessary for good thermal behaviour in the melt. In most of the early crucible forms it is unlikely that the charge was ever completely molten and the process was akin to skull-melting.

Here we restrict our attention to the case of r.f. induction-melting using a simple, solenoidal coil in conjunction with a cold crucible whose detailed shape and form is governed by three essential requirements:

(a) to contain the molten charge;

(b) to promote indirect coupling between charge and primary electro-magnetic flux;

(c) to give maximum freedom of movement to the molten charge in order that it can assume the optimum shape allowed by the internal electromagnetic field distributions.

1.1. The Hukin crucible

These problems were tackled by Hukin [5] who produced the elegant form shown in Fig. 1. This crucible was machined from standard, two-inch diameter (51 mm), copper bar. Here the essential claim was that all contact, between charge and crucible, was eliminated by means of electromagnetic forces of repulsion; i.e. that the charge was completely levitated. Although there is considerable separation of the charge and crucible it is unlikely that such complete levitation is achieved. By virtue of the shape of a crucible segment the lengths of the necessary current paths diminish towards the inner edge, where they vanish, giving rise to a melt shape resembling the lower half of a

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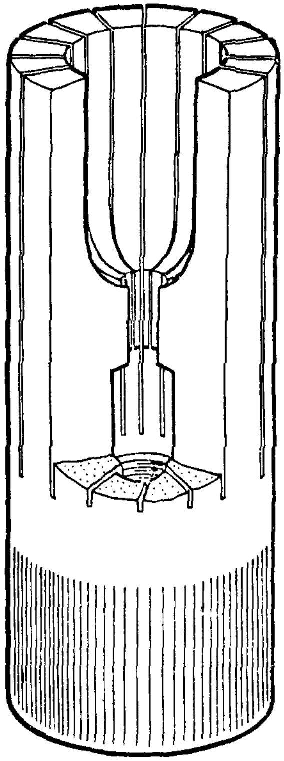

Figure 1 Diagrammatic sketch of the cold crucible (after Hukin).

Figure 2 Hybrid crucible illustrating failure of the levitation mechanism to provide containment.

prolate spheroid. This shape is terminated by a dimple formed at the hole in the central region of the crucible floor. Surface tension forces provide the essential mechanism for containment [6, 7] necessitating the minimization of the width of any aperture needed to permit indirect coupling of the charge to the primary electro-magnetic field.

It is important to emphasize that Fig. 1 is not dimensioned and that improved machining techniques allow the routine production of crucibles with sufficiently small holes in the central region of the crucible floor (about 3 mm diameter) to ensure retention of the molten charge [8].

1.2. Levitation

Failure of the electro-magnetic levitation mechanism to provide containment is illustrated in Fig. 2. This shows one of our early crucibles which could only be used if the central hole through the crucible floor (of diameter about 7 mm) was plugged with a suitable refractory. This plug was only a loose fit and did not need to be positioned with any degree of precision. Indeed the plug could be a hollow alumina tube, with a bore of about 3 mm and an external diameter of 6.35 mm. In the particular example illustrated, the charge material was nickel and the plug was a cylinder of sintered NiO. Progressive deterioration of this plug eventually allowed the molten charge to escape, to become inextricably enmeshed in the crucible structure.

However, this particular crucible (Fig. 2) has a number of interesting features. Firstly, it is based on the traditional tubular construction, brazed to a suitably prepared copper liner. Before slotting, the crucible-section was copper plated externally, in order to ensure uniformly continuous current paths when the fingers were separated. Secondly, its construction also lends itself to a maximization of the number of slots, thought to promote the charge levitation process. In this case 18 slots were possible using a standard slitting-wheel, 0.8 mm thick, consistent with a central hole of about 7 mm in diameter. Clearly, simply increasing the number of slots does not produce levitation, such as to provide containment of the molten charge, at a nominal working frequency of 500 kHz.

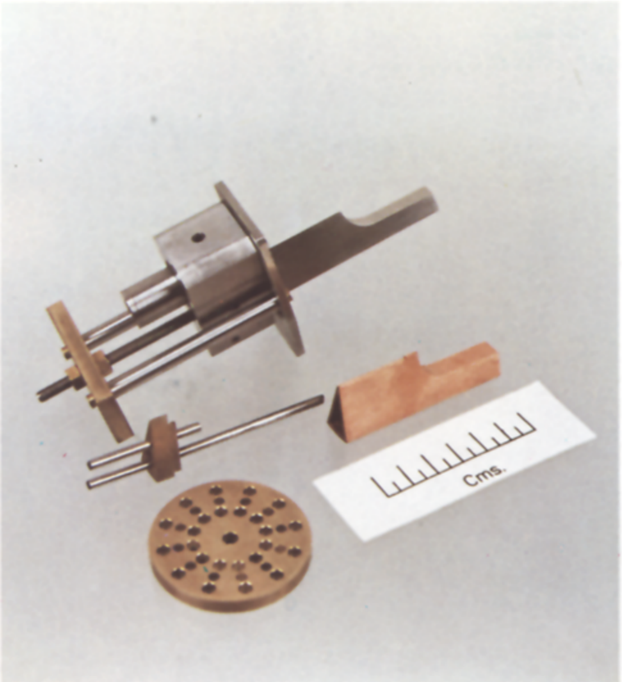

Figure 3 Stainless steel former and the assembly of a complete finger.

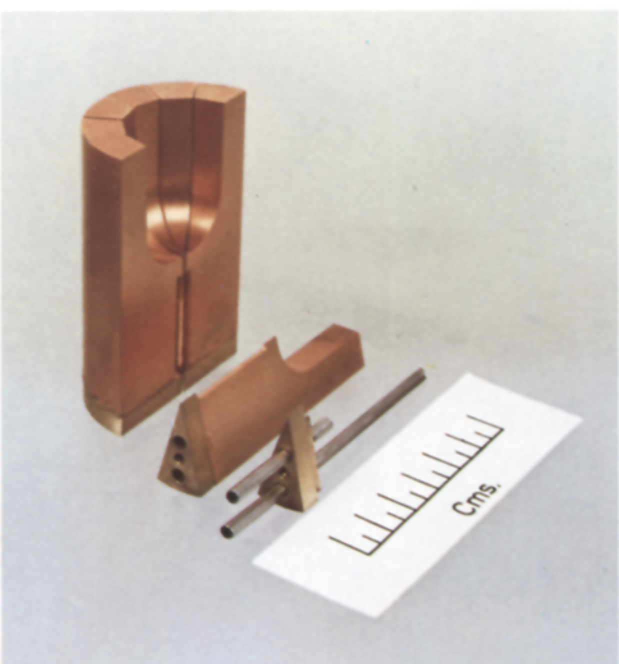

Figure 4 Assembly of the crucible section.

Figure 5 The completed crucible.

1.3. Difficulties associated with the employment of a solid crucible-form

Considering the difficulties which arise when carving a crucible from the solid, it is clear that:

(a) In order to minimize the size of the hole through the crucible floor, a compromise must be reached between the number of slots and the slot width. Besides the simple expedient of reducing the number of slots, improvement may be sought in reducing both the error in slot location and the width of the slot itself.

(b) Variable stress patterns in the copper bar will limit the accuracy and precision of the finished crucible.

(c) Any damage is largely of an irreversible nature leading to total loss of the crucible. This leads inevitably to increased costs, either through an increased failure rate in use, or as a result of the greater precision required in manufacture. During use, any significant damage will render the crucible valueless.

(d) Being of solid construction, the size of the crucible will be limited by such considerations as its weight and the need to produce a stress-free, large diameter bar of uniformly high purity.

(e) By virtue of the method of construction, the water-cooling is highly localized; although copper is an excellent thermal conductor, this feature will also tend to limit the size of the crucible.

2. The synthetic crucible

Considerations such as these led to a re-assessment of the benefits that might be gained by returning to the original methods of crucible construction, whilst retaining the best features of the new crucible form. Thus, a fabricated or synthetic crucible, one put together from identical individual parts to form an integrated whole, see Figs 3 to 5, was considered. As an engineering concept this method of construction poses formidable problems compared with those encountered in simply carving the crucible from the solid. Nevertheless, the method offers considerable advantages if the obvious difficulties can be overcome.

The crucible-section is formed from the appropriate number of identical fingers or segments mounted on a "tranfer plate" and in this case twelve is the largest number of fingers consistent with the production of a 50 mm crucible. It is the manufacture of these fingers, to the required degree of precision, which forms the major part of this work but here only a general discussion is presented. It is hoped to publish a more detailed account of the engineering aspects elsewhere.

2.1. Production of the standard segment

For many years in these laboratories, the technique of electro-forming has been used and developed in order to produce components of an optical nature (for example, microwave cavities and infra-red light guides). In this method a stainless steel former, with a high degree of surface finish, is employed in conjunction with copper plating from a simple bath using an acidified solution of copper sulphate. The former, acting as cathode, is generally rotated continuously about a suitable axis in order to promote the deposition of a uniform layer. This objective is also assisted by the associated stirring action which helps to maintain uniformity in the electrolyte. The best results are achieved as a result of experience, the main guide being the appearance of the freshly deposited copper which has a characteristic salmon-pink colour. It is worth noting that acid copper plating solutions are to be preferred to those based on cyanide complexes, where conditions permit their use, because they are more stable, give rise to less serious effluent problems and are generally simpler to operate and maintain.

It is found that the bond between the stainless steel former and the copper form is largely one of adhesion, comparable to a van der Waals bond. Consequently, provision is included in the design and manufacture of the former to permit the plated form to be separated mechanically from the mould. The stainless steel former used in this work is shown in Fig. 3, complete with such an arrangement in the shape of an extended plate, which also serves to provide additional control over the current distribution in order to ensure a greater degree of uniformity in the thickness of the deposited copper.

Integral with the steel former is a comparatively massive base whose detailed shape assists in the precision machining of the external surfaces of the copper finger to give a uniform wall thickness ($\nless$ 1.00 mm) and to enable it to form part of a right-cylinder.

Each finger is provided with a base plug precision machined from brass and provided with two holes to take thin-walled stainless steel tubing in order to provide for water cooling, see Fig. 3.

A third, blind hole is tapped to provide for single screw fixing of the segment to the base plate, as shown in Fig. 4. This base plate/transfer plate was machined dead flat. In constructing a complete finger therefore, the lower end is squared-off and then secured to the brass base plug. In the prototype "araldite" adhesive was used for this purpose, but a suitable low melting point solder would give a more reliable sealed joint. The complete finger must now be squared-off to a high degree of precision, in order to ensure that when it is secured to the base plate it combines with the other segments to form the complete crucible section which is then entirely demountable.

At this stage each finger is provided with the necessary lengths of stainless steel tubing; these are secured and sealed into the base plug using araldite adhesive and the tubes are lightly grooved to add to the strength of the bond. Finally, each complete finger is tested with water to a pressure of about 20 p.s.i.g. (0.24 MPa absolute).

2.2. Assembly of the completed crucible

The crucible-section was assembled by bolting the uniformly spaced fingers to the transfer plate to give a crucible-form having a high degree of axial symmetry. If desired, the spacing of the segments may be strictly determined during the manufacture of the end-plugs, such that they lock together when bolted down onto the transfer plate.

The manner of construction now follows the procedure outlined by Smith [6, 7] where the prepared crucible-section is secured to a prepared base-section to form a complete crucible which can be attached to a suitable manifold, Fig. 5. The base structure is formed from a single brass section with a flat top edged by a slight lip and provided with two concentric sets of holes designed to accept the base of the crucible-section. In general, the crucible and base sections are secured together by means of a single bolt fixing passing through the base into a tapped hole in the centre of the transfer plate. The crucible-section is then pulled down onto a gasket, formed in situ by using Locktite Superflex RTV2. This fluid material is thus forced into every aperture which requires to be sealed and is then left to cure.

In the prototype, as illustrated here, the water cooling tubes were made somewhat longer than was necessary and the formation of the castable gasket was effected with the aid of an auxilliary plate bolted to the upper face of the base-section. The reason for this was to facilitate the location of any possible leaks which could develop in the experimental, glued joints employed in the assembly of each finger.

Such a failure was experienced as a result of intermittent supply of cooling water to the crucible and the r.f. generator. Location of the leak was no simpler than is universally the case. However, there was little difficulty experienced in dismantling the crucible and extricating the charge material. The damaged finger was then found and the araldite was removed from the defective joints by using the appropriate solvent. The finger and complete crucible were then re-assembled as before. Failure of the glued joint was probably caused by differential expansion (between copper finger and brass end-plug) exacerbated by the brittle nature of the araldite bond. A soldered joint, employing a low melting point alloy, would very probably not have failed under these conditions.

2.3. Advantages of the electro-formed, fabricated crucible

(a) The economy in the use of the metal is considerable and the method of construction lends itself to the use of more expensive metals, such as pure silver.

(b) There is little or no size restriction and very large, lightweight crucibles are made possible.

(c) The individual fingers are relatively stress-free.

(d) Within the attainable limits of precision, the slots and central hole may be made vanishingly small; in larger crucibles there could be room for providing for the adjustment of the relative position of individual fingers. (These apertures are necessary only to allow transfer of r.f. power into the charge from the primary source. In any other physical sense their presence is an embarrassment and may give rise to "feathering" of the charge material into the slots. The resulting mechanical keying will negate any lift forces tending to separate charge and crucible.)

(e) The design philosophy is based on the concept of the demountable crucible, formed from identical fingers. Any finger may be either replaced or repaired as required.

3. Discussion

The advantages of the electroformed, fabricated crucible are considerable. Unfortunately, the engineering difficulties involved to ensure that all of the fingers which go to form the completed structure are identical, to a high degree of precision, are also considerable. Such precision can only be achieved by the batch-machining of a sufficient number of fingers, manufactured on machines which have been brought to an equilibrium condition. In the absence of fully conditioned workshops, the difficulties are exacerbated by temperature variations caused by normal usage of the available work space and the vagaries of the weather superimposed upon the diurnal cycle. In practice, these difficulties are off-set by making use of the fact that a copper form can be separated from the former. This allows that stainless steel former to be used for plating all the forms, under approximately the same condition, each of which may be replaced, in turn, on the former for the machining processes to be carried out. The use of commercial plating baths, with their increased deposition rates and throwing power would greatly speed up the plating process, which at present takes four days per finger, and require the removal of reduced amounts of excess metal at the machining stage.

Further work, which would be of great interest to pursue, would make use of the superior throwing power of commercial plating baths in order to plate the interior of a female mould, of the correct shape and maintained at constant temperature. In this way virtually all of the difficult machining operations could be eliminated. Furthermore, if the correct number of segments could be "jigged-up" to form the required shape, they may be "cold welded" by means of plating-on the necessary amount of metal to form a variety of terminations for the crucible-section. This technique has been successfully used in our experimental series of cold crucibles, however, the advantage of a demountable, repairing/replacement facility is then lost.

It is not often that crystal growers get the chance to grow their own crucibles and so, if only for this reason, the project has been of considerable interest. However, it is to be hoped that with the availability of large repairable crucibles, made possible by the work reported here, increased effort will lead to a wider application of the cold crucible in the fields of crystal growth and materials preparation. This optimistic view is encouraged by the interest shown in the United States ("Studying, Investigating and Developing New and Improved Cold Crucible Techniques" [9]), where a research and development programme is to be instigated, with the aim of growing large diameter, high purity, oxygen-free silicon single crystals and other, refractory materials of interest to the U.S.A.F.

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