Lucalox* Alumina: The Ceramic That Revolutionized Outdoor Lighting

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Introduction

In 1966 the General Electric (GE) Company introduced the Lucalox lamp, a high-intensity, high-pressure sodium-vapor lamp that has the highest light-producing efficacy (more than 100 lumens per watt) of any lamp with a polychromatic spectrum. That lamp (shown in Figure 1) and similar lamps produced by other manufacturers have revolutionized highway and roadway lighting. The annual worldwide market for them is now about $500 million dollars, and they are used for the great majority of outdoor lighting.

The envelope of the new lamp was a new pore-free polycrystalline aluminum oxide ceramic with excellent corrosion resistance and light-transmitting properties that had resulted from earlier research at the GE Corporate Research and Development Center in Schenectady, New York. The new ceramic was developed in the course of a program, the initial goals of which were to better understand the sintering process. The material was widely spoken of as a product of research purely motivated by a desire for understanding, and this was the case, at least initially.

The development started with the observation and interpretation of an interesting ceramic microstructure using understanding from earlier work on metals. At all times, there was serious effort to understand the observed phenomena. However, when a potentially valuable product was identified, the research program shifted, first to demonstrate that a truly pore-free product was possible, and second and much more difficult, to develop processes for making the material reproducibly and economically in the shapes needed. As in other materials-development efforts, the work required a considerable amount of enlightened empirical work, but it raised many questions the answers to which contributed to an understanding of the competitive processes that occur during sintering.

This article** will emphasize the research that led to the new ceramic, the circumstances under which it was performed, and the new understanding that it produced. Doping with MgO plays an important role in permitting the development of the necessary microstructure in the ceramic, and the mechanism by which it operates is still being argued. Toward the end of the article, I briefly discuss this problem.

This is primarily a personal narrative because I was closely involved with nearly all the steps. The development of the lamp itself followed the ceramic development by several years and was done independently at the Large Lamp Division of GE. I shall discuss that work only briefly.

Early Work

By the mid-1950s, the great effort to make metallurgy scientific, which started at the end of World War II, was in full swing. Its goals were more to understand and quantitatively explain well-known properties of metals than to develop improved alloys. Solid-state diffusion, the origins of microstructure by the nucleation and growth of new phases, crystal growth, grain growth and sintering, and the role of microstructure and dislocations in controlling properties such as flow resistance, work hardening, and fracture were among the phenomena studied.

Ceramics then, to the purist, were materials made from clay mixed with other minerals and then fired to consolidate them. The final product consisted of crystalline phases bonded by a silicate glass. A few new single-phase, nonmetallic, polycrystalline inorganic materials such as barium titanate ferroelectrics, ferrimagnetic ferrites, and uranium dioxide nuclear fuel had been developed during or after World War II. Some older materials such as "recrystallized" alumina were sometimes called ceramics, particularly in the United States, but the usage was rare except there. However, this class of materials was becoming technologically important.

I had been active in metals research for most of the previous decade. In 1954 I was asked by Herbert Hollomon, manager of the Metallurgy Department of the GE Research Laboratory, to form a group to study ceramics. The goals were to understand the origin of properties and to learn to control them by using the ideas and techniques then being used in metals research as well as any other appropriate methods. A group of people with a variety of backgrounds in metallurgy, crystallography, chemistry, and of course ceramics was assembled, and we selected some topics for study.

The properties of classical ceramics were commonly related to composition and the identity of the phases present. Microstructure was sometimes examined on petrographic thin sections, but the primary goal was to identify the phases from their optical properties. The technique of the metallographer—examining polished surfaces at high magnification to determine the shape and arrangement of grains or phases—was rarely if ever used on ceramics to relate microstructure to properties. One of our innovations was to use this technique to study the microstructures of a variety of ceramics.

Substantially all ceramic materials are made by sintering powders. There was already a considerable amount of research being done on sintering, particularly of metal powders, so it seemed appropriate that we select it as one major area of study.

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* Lucalox is a trademark of the General Electric Company.

** Part of this paper draws upon the manuscript for a seminar on the new lamp and new ceramic given to the Massachusetss Institute of Technology Metallurgy Department in December 1967.

Sintering Research in the Mid-1950s

One of the major goals of sintering research in the mid-1950s was to understand particle joining and the growth of necks between individual particles. Less attention was paid to the more complex transformations that occur when a multiparticle array is sintered. That was reasonable because powder metallurgists were doing most of the work, and the coarse powders they used (usually >20 μm) did join and shrink a little but would not sinter to high density by heat treatment alone, so neck formation was the major phenomenon observed. Metallurgists had long believed that plastic flow (i.e., dislocation movement in the individual particles) was the major matter-transport mechanism for neck formation. However, a few years earlier, Kuczynski[1] had concluded, from an analysis of the growth of the neck between a sphere and the plate on which it lay, that matter transport occurred by solid-state diffusion. He recognized that the process involved the creation and annihilation of lattice vacancies, but he did not identify a source or sink for the vacancies or discuss the approach of particle centers (specimen shrinkage). Although his conclusion was accepted by many, there was strong continuing support for the plastic-flow hypothesis. One argument was that if the mechanism of pore elimination in a compact was the evaporation of lattice vacancies from a pore and their diffusion to the surface where they would be eliminated, then it should take longer for a large specimen to sinter than a small one. Since no such size dependence is observed, it was concluded that the mechanism must be plastic flow.

Alexander and Baluffi[2] had observed that pores lying on a grain boundary in a sintered copper compact might be eliminated while pores not on grain boundaries remained. They suggested that the atoms that filled the pores came from the grain boundary. Many workers interpreted this to mean that the atoms had diffused in from the surface along the grain boundary—that is, that vacancies from the pore had diffused out along the grain boundary to the surface where they were annihilated.

In most respects, oxide particles are better for sintering than metal particles because they are smaller (about 1 μm or less) and with suitable heat treatment, compacts made from them can shrink to achieve 95% or more of theoretical density. Kingery and Berg[3] were among the first ceramists to study sintering. They too were concerned with the development of necks between particles and observed the behavior of several nonmetallic materials. They made the useful observation that the proposals of Nabarro[4] and Herring[5] for microcreep, by the generation of lattice vacancies at grain boundaries subjected to a tensile stress and their annihilation at grain boundaries subjected to a compressive stress, more generally implied that grain boundaries could be both a source and a sink for vacancies. They postulated that the centers of two sintering particles could approach each other if atoms at the surface of contact were to diffuse away from there and be deposited at adjoining free surfaces. In a porous compact, the equivalent statement is that lattice vacancies could evaporate from a pore and be discharged at an adjoining grain boundary to permit the centers of the boundary-defining grains to approach each other and the whole piece to shrink.

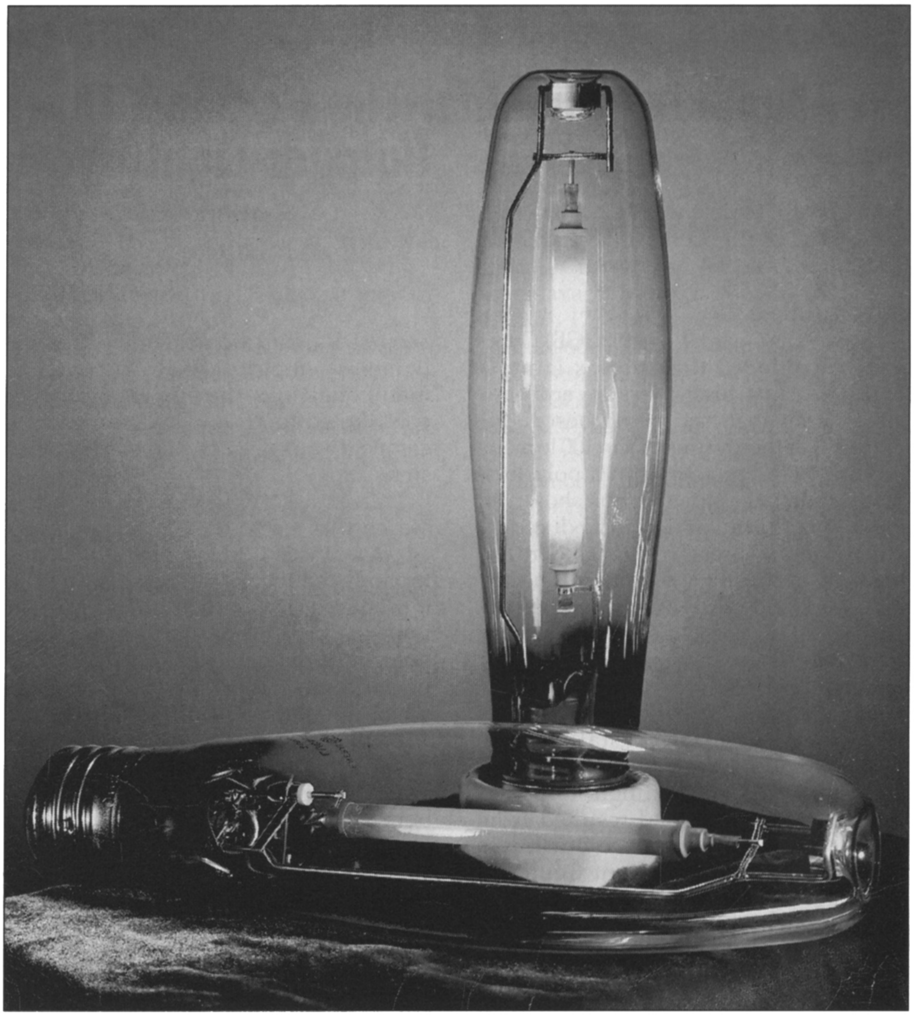

Figure 1. The Lucalox high-pressure sodium-vapor lamp. A discharge lamp with a cylindrical envelope of translucent Lucalox ceramic that resists high-temperature sodium vapor. The lamp proper is enclosed in an evacuated glass envelope for protection and to prevent oxidation of the metal end cap of the ceramic tube. The lamp is now available in a wide range of sizes. (1962 press release photo.)

Grain Growth and Pore Capture

Although the declared goals for the new group were to provide new understanding, there was of course hope for new materials. Probably the highest hope was to make ceramics ductile so that their high-temperature corrosion resistance and strength could be used in heat engines without fear of fracture. Somehow it was overlooked that ceramics had high flow resistance at high temperatures because dislocations could not easily move in them. That desire led to one wildly unsuccessful very early experiment: A piece of sintered (then usually called "recrystallized") alumina was heated for a long time above 1800°C and then hit with a forging hammer. It broke into many shards, which settled the argument of that approach to ceramic ductility.

I examined one of the shards using the metallographic technique of examining a polished and etched surface (as I have already mentioned, that technique was rarely if ever used on ceramics) and found the interesting structure shown in Figure 2. It has large grains full of pores, with some striking pore-free regions over which a grain boundary had apparently swept. The dotted line in Figure 2 shows the presumed initial position of the grain boundary. (The large voids that apparently lie on the present grain boundary are the result of surface chipping and are not large pores.) The interpretation[6] of the microstructure of Figure 2 emphasized that competitive reactions such as grain growth, and particularly discontinuous grain growth, can occur during sintering of a multiparticle array, and that they can markedly influence pore removal.

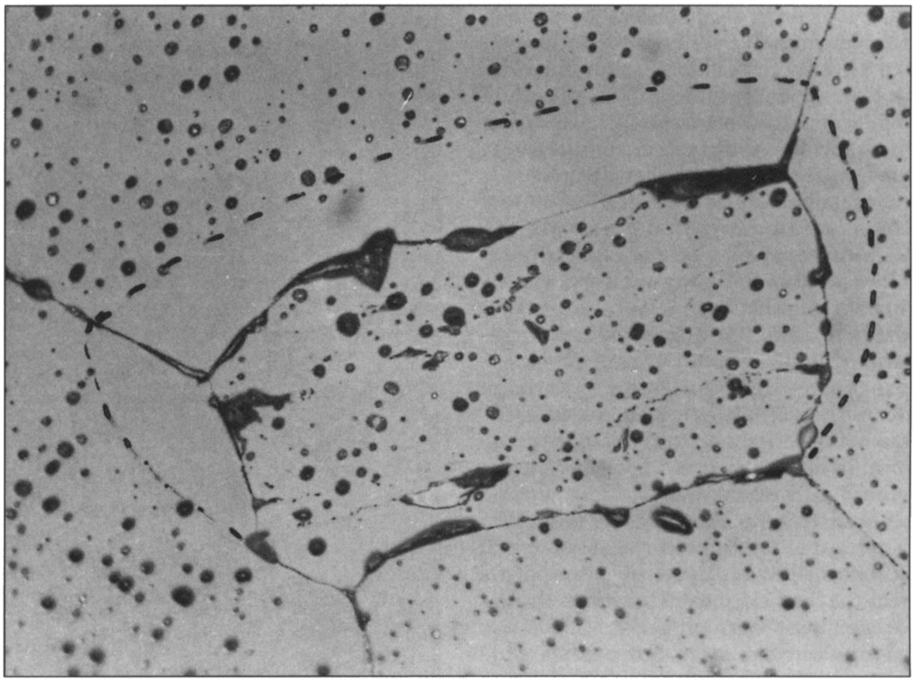

Figure 2. Microstructure of a shard of alumina that failed the forging test. The pores inside the large grains are the residue of misfit regions between the original powder particles. By discontinuous grain growth, the large grains grew by consuming the smaller grains generated initially and the pores between them. Pores on the new set of boundaries pinned the boundaries in position but were preferentially removed, permitting the boundaries to move to new positions where the process was repeated. The moving boundary thus left a pore-free wake. The dotted line shows the presumed initial position of the moving boundary in this micrograph. Original magnification is 250X. Etched in molten $K_2S_2O_7$.

The pores of Figure 2 must initially have been on grain boundaries because they were initially the regions of misfit between the original powder particles. That is consistent with an initial particle size of a few micrometers. The current average grain diameter is a few hundred micrometers. Therefore enormous grain growth must have occurred, quite certainly by the process called discontinuous grain growth or secondary recrystallization. Discontinuous grain growth occurs when some grain boundaries are more able to move than others, and there are several sets of conditions under which it can occur. An important one, pertinent to the interpretation of Figure 2, requires the presence of just enough second-phase inclusions of suitable size[7] to prevent grain-boundary migration and grain growth in that specimen (the Zener condition). In metals the common second-phase inclusions are nonmetallic solids, but in a sintering ceramic, pores act in the same way. Now if the effectiveness of these boundary pinning inclusions gradually decreases—by Ostwald ripening of solid inclusions or reduction in pore size by sintering, the time will come when a sufficient number of inclusions (pores) are removed at a few local sites to liberate a few high-mobility grain boundaries. Then those boundaries will move rapidly to absorb a few neighboring small grains and create some relatively large grains. The boundaries between a many-sided large grain and an array of small grains are strongly curved and can migrate past inclusions or pores that prevent the movement of less strongly curved boundaries. Hence the few larger grains consume their neighbors, and as they grow, their boundaries migrate past pores and the pores are thus entrapped inside the growing grain. The result is a new generation of relatively enormous grains filled with pores. Since the process involves the growth of a few grains from special sites, it superficially resembles the nucleation and growth by recrystallization of a new generation of grains in a deformed metal, hence the alternate name of secondary recrystallization used by many metallurgists.

When the process is completed, the boundaries of these large secondary grains are only slightly curved and are easily pinned by the few pores they find upon them. However, those pores can be easily removed with further heating, as Alexander and Baluffi had earlier observed. The boundary is thus freed to move a little until it encounters more pores that in turn are removed, also eventually encountering an area swept free of pores as shown in Figure 2.

Discontinuous grain growth requires high-mobility grain boundaries. If boundary mobility is low, a boundary can move only slowly when the disappearance of some inclusions frees it, giving time for more inclusions to be eliminated and more boundaries freed so that extreme grain-size contrast necessary for discontinuous grain growth never develops. The grain size remains uniform, and continuous grain growth occurs at a rate limited by the rate of disappearance of boundary pinning inclusions. A critical corollary of this is that only in discontinuous growth do boundaries migrate past large numbers of inclusions. Hence the presence of a large number of pores inside grains is a certain indicator that discontinuous grain growth has occurred.

At that time, these observations were of great interest because they supported the diffusion model rather than the plastic flow model for matter transport during sintering. They also indicated that the grain boundary rather than the surface was the sink for vacancies. There is no reasonable mechanical-flow model that would predict that pores at grain boundaries should be preferentially removed. The diffusion model does assume that vacancies evaporating from the pores could be annihilated at an adjacent boundary with grain centers approaching and specimen shrinkage occurring. If lattice vacancies were removed by diffusing to the surface to be annihilated, then pores near the surface would disappear faster. Observation showed them to be removed from grain boundaries throughout the specimen at approximately equal rates, so it appears that the boundary itself serves as the vacancy sink. Pores on grain boundaries are removed readily, but pores inside grains seem to be quite stable, indicating that grain-boundary diffusion is the preferred mode of matter transport.

Of more importance for the production of a pore-free ceramic, observations show that if discontinuous grain growth could be prevented then all pores could be removed by sintering because they all lie initially on grain boundaries. I first quickly attempted to do this by introducing "nonceramic" inclusions of platinum into the specimen to pin all grain boundaries as "nonmetallic" inclusions do in metals. My approach of imbibing a partially sintered specimen with chloroplatinic acid and then heating it further did not work. The nonceramic inclusions were coarse and did not inhibit grain growth so a black, porous ceramic resulted.

Identification of an Opportunity

Thus far the work was really very purely motivated by the desire to obtain scientific understanding of sintering. However, in the course of other work, an accidentally greatly overfired specimen made from a commercial alumina powder, which contains some undetermined impurities, did not undergo discontinuous growth and did sinter to a nearly pore-free state. The product was noted and the guess made that some unknown impurity had reduced grain-boundary mobility but no immediate attempts were made to identify the impurity, though its existence was mentioned in the paper[2] that included the previously mentioned analysis.

Shortly after that, a group from the Large Lamp Department of GE visited the laboratory, and more or less incidentally, this slightly translucent overfired specimen was shown to them. George Inman, manager of Advanced Engineering for that group, expressed considerable interest and suggested that if the product could be improved and fabricated into suitable shapes, it might be valuable for lamp envelopes. With that possible use established, we decided to start a program, the goal of which was the manufacture of pore-free alumina.

> An accidentally greatly overfired specimen made from a commercial alumina powder did not undergo discontinuous growth and did sinter to a nearly pore-free state.

In the interim, Bob Coble, one of Dave Kingery's students from Massachusetts Institute of Technology, had joined the group and was studying the initial stage of sintering. Kingery and Berg had devised a good model for neck formation between two spheres by diffusion and annihilation of lattice vacancies at grain boundaries and had shown that it was possible to test such a model in a multiparticle array by just observing the kinetics at the early stages of shrinkage. By studying shrinkage, you automatically rule out any contribution to neck growth by surface diffusion or vapor transport. The model seemed to hold well in the very early stages of sintering when the shape changes were small and the simple assumptions of the model were fulfilled. Coble had been working for some time observing initial sintering kinetics in several materials and was polishing his models and extending them to later stages in sintering. With only a little arm-twisting, he was persuaded to set aside these fundamental studies for a time and perform some experiments in an attempt to make the pore-free product.

The Search for a Pore-Free-Material Manufacturing Process

As was suggested in the section on Grain Growth and Pore Capture, the initial approach to making a pore-free alumina was to try to inhibit discontinuous grain growth and thus to avoid the capture of pores inside grains where they appeared to be quite stable. Again earlier work was helpful. I had observed several years earlier that discontinuous grain growth in high-purity zinc (containing a small dispersion of ZnO) could be prevented by the addition of 0.7-wt% silver and had attributed the effect to the reduction of grain-boundary mobility.[8] In addition we guessed that the accidentally overfired specimen that sintered to nearly theoretical density had an impurity that reduced grain-boundary mobility. Finally a greatly unrecognized paper of Cahoon and Christenson[9] led us to the identity of the impurity. They investigated the effect of many additives on sintering alumina by examining microstructures in petrographic thin sections (then the only technique used by ceramists). They had found that the small additions of MgO were remarkably efficient in eliminating the very large grains they found in most of their specimens.

An alumina powder made by the calcination of ammonium alum (Linde A) had been procured earlier and was being used for much of our sintering work, including the initial sintering-kinetics work, because it was so pure. It was used by Coble in the first additive experiments.

This selection of powder was later realized to be critically important because it possessed three characteristics not possessed by other powders available at that time. First it was pure, and the grains in sintered material had the characteristic foam-cell structure typically found in metals. Many commercial alumina powders yield elongated or even faceted grains that have been attributed to the presence of a liquid phase at sintering temperature. (See for example Figure 20 in Reference 14.) Second the particle size of the elementary particles was small ($<1\ \mu m$) and uniform. Third, although the powder is somewhat agglomerated, the agglomerates were readily broken up by small amounts of milling. Other potentially useful powders required so much milling that they became contaminated.

Following Cahoon and Christenson, the first additive Coble tried was a fraction of a percent of MgO,[10] and it worked! Discontinuous grain growth did not occur, and virtually all the pores were removed. Incidentally, MgO additions only inhibit discontinuous grain growth. They do not prevent continuous grain growth from occurring, for the reasons mentioned a few paragraphs earlier.

At the time, pore-free alumina was a very impressive product. A photograph

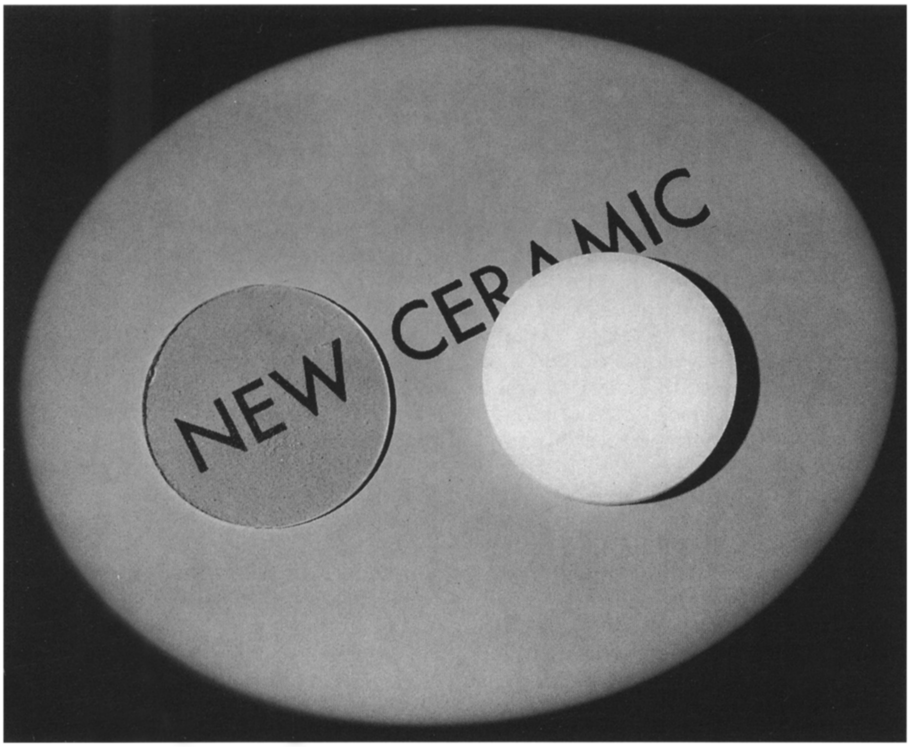

Figure 3. Translucency comparison of pore-free and normal porous alumina (press-release photo).

demonstrating the high translucency[11] of this material with a normal good specimen of porous sintered alumina is shown in Figure 3, and a comparison of microstructures between porous and nonporous alumina is shown in Figure 4. These photographs were used in publicity at the time the ceramic was announced. The pores in the pore-free specimen were eliminated when the grain size was small. Obviously a considerable amount of grain growth occurred upon later sintering.

Further Development of Lucalox

Given an approximate recipe for making this product, it was still necessary to optimize the composition and processing and firing conditions so that the product could be made reproducibly and economically in the shapes needed. This work continued for a couple of years with contributions from a number of people with primary responsibility held by Charles Bruch. Some of it is reported in a 1962 paper by Bruch.[12] Over most of that period, a major contributor was Nelson Grimm of the GE Lamp Glass department. To aid in process development, Grimm moved to Schenectady for a number of months and at the end of the development period returned to Cleveland to develop the final process used for making the tubing for lamp envelopes.

The new ceramic was announced in September 1959, and the name selected for the product was Lucalox from transLUCent ALuminum OXide. For a few days, it was pronounced "loose-alox" but then the little known rule of English pronunciation that C before A is always hard prevailed, and the present pronunciation "luke-alox" was adopted.

The Role of MgO

The mechanism by which MgO operates to prevent discontinuous grain growth has long been a matter of debate. A myriad of explanations have been offered, and many of them have been recently reviewed by Harmer.[13] As indicated previously, we were led to use MgO doping by the desire to find an additive that would inhibit discontinuous grain growth. The observations of Cahoon and Christenson[9] that magnesia additions inhibited the appearance of large grains in their (relatively impure) alumina sintered specimens also led to this choice. Additions above the solid-solution limit form magnesium-aluminate particles at the grain boundaries. We first thought it might have worked by second-phase pinning of the boundaries. The observation that additions within the solid-solubility range also worked ruled out this explanation. After some internal disagreement, we decided the mechanism must be Lucke-Cahn[14] drag caused by magnesium ion[15] adsorbed at the grain boundary. Then several workers[16,17] were unable to find any evidence of adsorption of magnesium ions at the grain boundaries, and the mechanism remained in limbo for a time. However, others, particularly Monohan and Halloran,[18] and then Burke, Lay, and Prochazka,[19] demonstrated quite conclusively that the MgO addition *did* reduce grain-boundary mobility. Since it was also believed that there is no, or very little, adsorption of magnesium at the grain boundary then a different mechanism was needed to explain the reduction in grain-boundary mobility it accomplished. In 1990 I suggested a mechanism based on poisoning of dissolution or deposition sites,[20] as described in the next paragraphs. There has been little discussion of it in the literature.

The simplest version of grain-boundary motion assumes that the disordered region separating two differently oriented lattices migrates by atom movements of less than an interatomic distance to reorient the lattice of the consumed grain to that of the growing grain. In this version, adsorbed atoms at the interface must jump a full interatomic distance to keep up with the moving boundary. This more difficult process provides the grain-boundary drag. There is, however, considerable evidence that grain boundaries, or at least low-angle boundaries, are highly structured. Accepting this, Gleiter[21] has suggested that grain-boundary motion is a more complex process requiring an atom from a grain being consumed to separate from its site and diffuse along the grain boundary to a suitable attachment point in the boundary of the growing grain. Grain growth is thus reasonably assumed to occur by a mechanism very similar to the Burton-Cabrera-Frank[22] model of crystal growth from a fluid.

The growth or solution of crystals in a fluid, or in aqueous media at least, can be profoundly modified by small impurity additions. A textbook example is the ability of a trace of urea in solution to change the habit of sodium-chloride crystals growing from solution from cubes to octahedra. In dissolution, Gilman, Johnston, and Sears[23] observed that a few parts per million of ferric fluoride in water can change the shape of

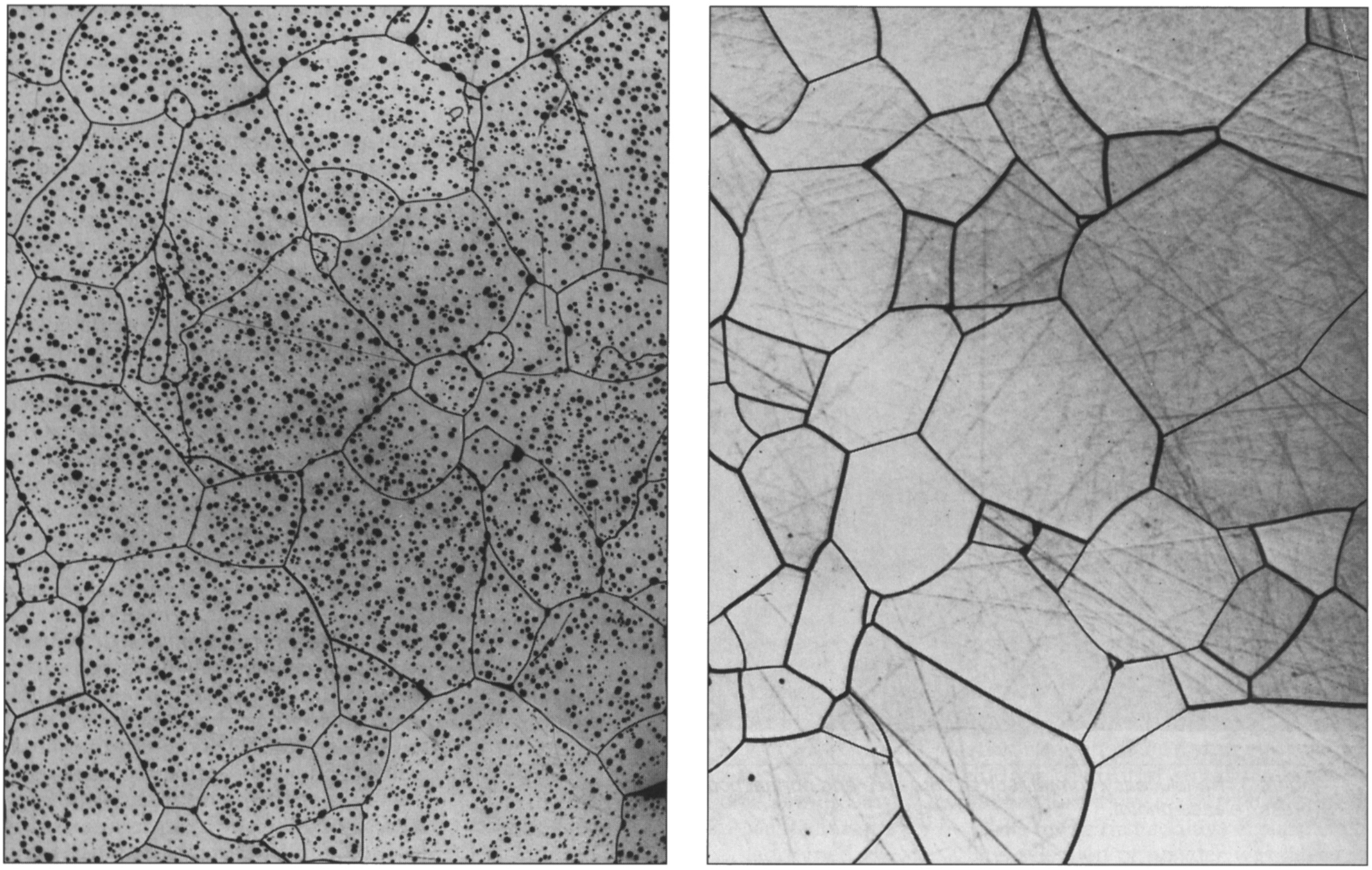

Figure 4. Comparison of microstructures of porous, sintered alumina, sintered without MgO addition (left) and the pore-free ceramic in which a MgO addition prevented discontinuous grain growth (right). Original magnification is 250X. (Press release photo.)

etch pits at dislocations from broad depressions to sharp pits. Both of these effects are significant, and the additives are highly specific. In the crystal-growth case, the poison works by poisoning growth sites on cube faces but not on octahedron faces. In the etch-pit case, shallow depressions at dislocations form because once a new lattice plane step is nucleated, the step moves rapidly and the layer is removed to quite a distance. If the dissolution steps are poisoned and the rate of dissolution is decreased but the rate of nucleation of new steps is unaffected, a pit with much steeper walls is generated. Grain-boundary mobility reduction by MgO doping of alumina is attributed to this poisoning.

With the preponderance of the evidence pointing to the fact that there was no detectable adsorption of magnesium at alumina grain boundaries, but with the certainty that magnesium did indeed reduce grain-boundary mobility, the poisoning hypothesis was the only proposed mechanism that would explain the effect in 1990.

Most recently Soni et al.[24] have observed, using a scanning ion microprobe and secondary ion mass spectrometry, that there is indeed segregation of magnesium at the grain boundaries of MgO-doped alumina if the alumina is slowly cooled from its 1800°C sintering temperature. Faster cooling produces less segregation. The work also showed that in slowly cooled CaO-doped specimens, calcium is adsorbed almost as much as magnesium under similar conditions.

The first additive Coble tried was a fraction of a percent of MgO, and it worked!

Although these findings confirm the long-held suspicion that some magnesium is adsorbed at alumina grain boundaries, they raise another problem. They confirm that calcium is adsorbed to about the same extent from calcium-doped specimens, yet calcium oxide doping of alumina does not inhibit discontinuous grain growth,[25] and therefore it presumably does not reduce grain-boundary mobility. For comparable concentrations of adsorbate at the grain boundary, there is no reason why one ion should be substantially more effective than another in causing Lucke-Cahn drag. The alternate suggestion that magnesium ions are specifically able to poison growth and/or dissolution sites overcomes this difficulty.

Development of the Lucalox Lamp

The Lamp Division began experimenting with the new material in 1956. A metal-vapor discharge lamp was planned, and it was hoped that with a strong corrosion-resistant envelope that could withstand higher temperatures, a lamp could be designed with higher efficiencies than had been previously obtained. By 1960 K. Schmidt had found that sodium vapor could be contained in the Lucalox ceramic material at higher temperatures than before, and that it produced golden-white light at high efficiency. W.C. Louden worked extensively on lamp components: the metal ends and electrodes, and the sealing techniques for bringing electrical leads into the interior of the ceramic envelope. All of this work was carried out in Cleveland with only intermittent consultation with the Research and Development Center on materials problems.

In December 1962, the new lamp was announced, and the results were outstanding: an efficacy of 105 lumens of light per watt, the most efficient source of light of acceptable color in the history of electric lamps.

For purposes of comparison, the conventional incandescent lamp has an efficiency of about 18 lumens per watt. The conventional mercury-vapor lamp used in outdoor lighting produces about 50 lumens per watt, and the fluorescent lamp about 75. In its day, an efficiency of 100 lumens per watt was more or less the equivalent of the four-minute mile. The low-pressure sodium-vapor lamps that were popular in the United States some years ago have been improved until their efficacy is over 100 lumens per watt. They were extensively used in Europe, but the light was exclusively at the wavelength of the sodium D line so all objects viewed in their light are either yellow or black. By raising sodium vapor pressure and temperature, other transitions than that giving rise to the sodium D line are excited, all the lines are broadened toward the red end, and a quite continuous spectrum a little deficient in blue is obtained so that the light has a golden color. Sodium vapor at such high pressures and temperatures rapidly attacks glass lamp envelopes but is readily contained by Lucalox ceramic.

Conclusion

A number of new ideas about technique and starting materials came out of this development:

- Microscopic examination of polished and etched surfaces can be as useful in ceramic process development as it is in metallurgy.
- Steps should be taken to prevent discontinuous grain growth during sintering because it entraps pores inside grains where they are very stable. However, there is no simple rule to define what is an appropriate additive to prevent such growth. Magnesium is a very specific addition for aluminum oxide.
- The initial powder selection is critical. It should be pure, its particle size uniform, and any agglomerates easily deagglomerated. Prior to this work, it was assumed that a high green density in a pressed compact would yield the highest density sintered product, and that a powder with a range of particle sizes would pack to a higher green density than a monodisperse one. However, shrinkage results from the annihilation of lattice vacancies from pores at an adjacent grain boundary. This must be a cooperative process if particle centers are to approach each other. All the atoms between a pair of grains must be removed if their centers are to approach. To accomplish this, vacancies must diffuse for about one-half the pore-to-pore distance to remove the full layer of atoms. (There may have to be a little grain-boundary sliding also.) If a range of particle sizes is used to get good packing in the green compact, there will be a range of pore sizes in the partly sintered compact. As the smaller ones are eliminated, the separation between the larger ones increases so that not only are the pore curvatures and driving force for evaporation smaller, but the diffusion distances are larger, greatly increasing the time for sufficient diffusion to permit the grain centers to approach each other. The optimum configuration is to have all particles, and hence all pores, the same size, so pores can disappear simultaneously leaving no orphans.
- The powder agglomerates must be broken up. In the development work, a defect initially encountered was "measles"—small white regions scattered throughout the specimen. It was quickly found that these could be eliminated by properly milling the powder to break up all agglomerates, and it became obvious that each measle was a small cluster of larger pores initially formed by the poor fit of large agglomerates. It had been long known that milling would increase the final density of a sintered compact. This observation provided a more reasonable explanation than the one sometimes made at the time that the energy imparted by the grinding process also served as a driving force for sintering. The only important driving force remains surface energy.
- The agglomerates in the powder must be soft enough to be broken up with mild milling. If extensive milling is required, contamination is introduced and uniformity of particle size is difficult to achieve.

Polycrystalline alumina ceramics of the Lucalox type are now made by a number of manufacturers. Its combined properties of high light-transmitting ability and excellent resistance to hot sodium vapor make it the only successful material found to date for containing the plasma of the high-pressure sodium-vapor lamp. Single-crystal sapphire tubing made by the edge-defined growth process was tried briefly, but its higher cost and difficulties with strength and light transmission prevented its displacing the polycrystalline material. While there are minor uses for pore-free alumina, the lamp-envelope applications are the only important ones.

Acknowledgments

I thank two of my colleagues for the contributions they made during discussions about parts of this article. Both have made important contributions to further improvements in Lucalox ceramic processing, and each has been responsible for the development of another commercially important pore-free ceramic. Svante Prochazka[26] developed silicon carbide for structural purposes, and Charles Greskovich[27] was the leader in the development of the first polycrystalline ceramic scintillator for computer-assisted-tomography scanners.

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7. J.E. Burke, in *Grain Growth in Industrial Metallurgy* (American Society of Metals, Cleveland, 1954) p. 1. The volume fraction of inclusions just necessary to prevent grain growth in a uniform array of grains with a uniform dispersion of inclusions was subsequently specified by Zener as $f = 4/3(r_i/G)$ where f is the volume fraction of inclusions, r_i is their radius, and G is the average grain diameter (quoted by Cyril Smith, *Trans. AIME* **175** [1947] p. 15). In the present case where the pores are the operating inclusions and must initially lie on grain boundaries, they are much more effective. Therefore under those conditions, $f = 8(r_i/G)^2$ (J.E. Burke and J. Rosolowski, *Treatise on Solid State Chemistry*, vol. 4, edited by B. Hannay [Plenum Press, New York, 1976] p. 621.)
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11. Pore-free alumina is not transparent because aluminum oxide is birefringent. An entering ray is split into polarized ordinary and extraordinary components vibrating in planes normal to each other and showing different refractive indices. Upon entering each new grain, all rays are again split, so a point source viewed through the ceramic emerges as a circle of confusion, which is greater the smaller the grain size and the thicker the specimen.
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