POLYCRYSTALLINE MATERIALS FOR ARC TUBE ENVELOPES Svante Procházka

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1. INTRODUCTION

The high-pressure sodium discharge lamp was made possible by the invention and development of highly translucent alumina ceramics in the late fifties [1]. This material combined two essential properties: the chemical resistance to sodium at temperatures above 1000° C and light transmittance. Such material had to exhibit, of course, a number of other properties such as mechanical strength, thermal shock resistance, low vapor pressure and high electrical resistivity at high temperatures and, above all, the fabricability into thin walled tubes to make the new lamp possible. However the combination of the above two properties, not available in other materials, was the essential breakthrough. This contribution focuses on these two subjects, i.e., light transmission in polycrystalline materials and resistance to sodium vapor in alumina and emphasizes advances made in the field over the past decade. Light transmission in polycrystalline solids (with a large enough bandgap) depends on chemical composition, porosity, phase composition, the density and size of scattering centers, surface roughness and in non-cubic substances also on optical anisotropy and grain size. In a ceramic the degree of transmission reflects the perfection of its microstructure, absence of pores, inclusions, impurities, and microcracks. Therefore the translucency of a ceramic body is a manifestation of well-conducted powder processing. In fact, the advent of translucent materials made it possible to observe for the first time all the defects in ceramic bodies introduced in fabrication and also made possible the devising of ways to eliminate these defects and brought about a quantum improvement in perfection of dense ceramics.

2. THE ALUMINA ENVELOPE

Alumina tubing for lamps is made from carefully deagglomerated and processed high grade alumina powders by isostatic pressing or extrusion with binders and subsequent sintering in hydrogen or vacuum near 1850° C. The key conditions for success are:

- the necessary purity, particle size and morphology of the starting alumina powder,
- sufficient deagglomeration to achieve correct particle packing on compaction,
- and conduction of the sintering such as to remove maximum impurities and porosity.

Common impurities in Al₂O₃ viz SiO₂, Na₂O, K_2O , CaO, and Fe_2O_3 are highly undesirable as their presence leads to formation of separate phases that affect microstructure and contribute to light scattering. Studies on the effects of impurities and additions in sintered alumina abound [2, 3]; however, rigorous correlations with concentrations supported by analytical work are rare because of limitations of analytical techniques at the ppm levels, because of frequent complications caused by synergies between impurities and sintering atmosphere, concetration gradients in sintered bodies, and so forth. The purity of the starting alumina powder must be substantially higher than the best metallurgical and ceramic grades and until recently the selection was very small. Over the past 10 years, however several new sources of high purity alumina suitable for arc envelope fabrication became available and are competing with alum derived Al_2O_3 , the prior sole source.

Most serious impurities in alumina are transition metal oxides that show appreciable solubility in Al_2C_3 Their ions form light-absorbing defects in the alumina lattice. The main harm arises when they become reduced in fabrication to metals in the form of a very fine dispersion and thus bring about severe absorption. Fe, Mo, Ni, Co, V, Cr, Ti, Cu are the most prominent, and 20 ppm of these oxides in the starting material is a serious concern.

Calculations and experiments demonstrate that multiple scattering by residual pores is the origin of

most of the attenuation of light in polycrystalline alumina [4]. The scattering by pores is strongly sizedependent. Very small pores, whose sizes are much less than the wavelength of incident light, scatter very little so that bodies with as much as 90% of such pores can be still perfectly transparent [5], which is demonstrated by gels [6]. In sintering, however, such small pores are unstable and disappear. The porosity retained in dense sintered alumina is composed of spherical voids about 0.8 to 3.0 μm i.e., in the range that causes strong forward (Mie) scattering, and therefore has to be minimal -0.01% or less, for use in arc envelopes. We can show that at this porosity level spherical pores of size d uniformly distributed in a slab of thickness 100 d will intercept every passing light beam at least once. With 2 μ m pores and 0.01% porosity, there are 20 milion pores per cc, and a typical wall of an arc tube 0.8 mm thick will show only marginal in-line transmission. Porosities lower than 0.01% are therefore needed and can now be routinely achieved. Larger pores for the same porosity level scatter light less, and therefore, controlled pore growth, if it can be achieved in the sintering operation, is helpful in improving in-line transmission.

Closed pores at grain boundaries that are empty of gas are eliminated during sintering by surface tension. Pores filled with gas which cannot diffuse through the solid may partially shrink but cannot close and will bring about opacity [7, 8]. Argon and helium are such gases. Nitrogen is similarly trapped if sintering proceeds in air, however, and contrary to prior belief, it does diffuse out of pores so long as hydrogene is part of the sintering atmosphere [9]. This observation makes possible use of a safe and inexpensive nitrogenhydrogen furnace environment in manufacturing.

Pores in a sintering body can be removed only as long as they are attached to grain boundaries and only if they do not exceed a certain pore-to-grain size ratio. Once pores are left behind by advancing grain boundaries and are thus swept into the bulk of grains, the diffusion processes responsible for their removal cease to operate. The process of pore removal proceeds by diffusion of vacancies and can be compared to evaporation of a droplet of liquid that proceeds by molecules leaving its surface one by one. The conditions under which pores are removed have been studied extensively [10, 11], yet the reason why a great majority of pores are eliminated and a small but critical fraction isn't, is not positively known and is a subject of active research.

A necessary requirement for entrapped pores to be removed in sintering is grain growth. When grains grow the grain boundaries sweep more and more volume and eliminate entrapped pores on contact [12]. Grain growth assists pore removal by yet another mechanism. There exists an upper pore-to-grain size ratio above which pores are too large to be removed. They behave as cavities and shrink only in proportion to the body. When grains grow, the above ratio decreases, and the larger pores can be also removed [13].

Frequently, residual pores form clusters inside grains as shown in Figure 1. Their origin has been linked to an early burst of grain growth, i.e. rapid grain boundary advancement, which then slowed down to just the rate at which pores could be removed by diffusion [14]. A possible reason for their formation in alumina, according to Halloran and Dynys [15], are hard powder agglomerates that during heat treatment (powder calcination) lost their grain boundaries and formed a porous single crystal, which is incorporated into the microstructure. Still another mechanism of pore cluster formation was traced by Greskovich to rapid local grain growth catalyzed by larger particles such as debris from millwear [16].

The usual grain size of lamp envelope alumina, 25 to 30 μ m, is a compromise between the two trends. Increased grain size, as a rule, brings about increased transmission because of reduced porosity. With large grains, however, strength drops because of internal stresses arising from thermal expansion anisotropy. Also fracture toughness of alumina decreases with increasing grain size [17] and brings about increased failure rate in manufacturing, particularly on grinding and sealing. Consequently an average grain size of 25 to 30 μ m was found to be a practical compromise. Because very large grains generate large stress concentrations and weaken the bodies, a narrow grain size distribution is obviously preferred. The distributions observed in translucent aluminas are mostly log normal, i.e., relatively narrow [18]. Notable deviations in the distribution signal the presence of impurities and/or the presence of extraneous phase particles [19].

Grain growth is controlled by small additions of magnesia that slow down the rate of grain boundary migration and that prevent massive incorporation of pores into grain interiors. It is now agreed that the effect of magnesia is due to its segregation at and near grain boundaries where the larger magnesia cations exert drag on the moving grain boundaries [20]. On the atomic level this effect may be a consequence of bonding of Mg ions to ledges [21]. No other practical addition has been found that would duplicate the magnesia effect.

Small additions of other oxides have been sometimes recommended or used on top of magnesia, and some improvements in properties or processing aspects are claimed, for example, $Y_2O_3[22]$, $Y_2O_3 +$ $La_2O_3[23]$, ZrO_2 , TiO_2 , $HfO_2[24]$, BeO, SrO, and so forth. Doping with MgO + Y_2O_3 + La_2O_3 , for instance substantially, lowered according to Kameno. the firing temperature so that highly translucent bodies could be obtained at 1700° C [25]. The effect is, with all probability, the result of liquid-assisted sintering and therefore some extraneous phases would be expected in such compositions.

Magnesium is lost in part on sintering by evaporation, and this loss has to be taken into account in the formulation of the composition. The loss depends on pO_2 , the furnace, and the type of kiln furniture used for setting the green ware. If the magnesium concentration drops below about 100 ppm, uncontrolled grain growth (exaggerated growth) takes place and results in the formation of large fequently prismatic alumina grains [26, 45]. Since the surface is more depleted of Mg than the bulk of the body, the exaggerated grain growth may affect only the very surface of the body while normal grain growth proceeds in the bulk [27]. The magnesium depletion of the surface is such that Mg cannot be usually detected by microanalytical techniques such as Auger.

On the other hand, when the magnesia addition is more that about 200 ppm, the solubility limit in alumina is exceeded, and magnesium aluminate, MgAl₂O₄ forms as separate grains [28]. These second phase particles having a lower index of refraction contribute also to light scattering. However, the scattering efficiency of a particle is approximately proportional to $\Delta n/n$, the relative difference in indexes of refraction, and is therefore small in comparison to the effect of pores.

Other phases that may be sometimes observed in alumina for lamp envelopes are beta alumina grains which form when sodium, potassium or calcium are present and form lamellar particles of various compositions that may also incorporate magnesium [29, 30]. The contribution of beta alumina grains to scattering and absorption is also small, but they may be harmful by promoting interaction with sodium in service. These phases are unstable at sintering temperatures (1850° C) and may be therefore absent in rapidly cooled bodies but may crystallize on annealing at some lower temperature, such as on sealing.

The contribution of surface roughness to light scattering and transmittance was investigated by Ingold and Taylor [31] and Kaneno and Oda [25]. They observed a correlation between surface roughness measured at short wavelength < 0.08 mm, in-line transmittance, and lamp efficiency. The short wave component of surface roughness is due to grain boundary grooves, growth steps, crystal facets, and in some cases, deposits from the sintering furnace. This type of roughness can be greatly reduced by surface etching with molten borates or other etchants [32]. Removal of 3 to 4 μ m off the surface is sufficient to reduce surface roghness from 2.5 μ m to 0.8 μ m. The surface of sintered alumina is also smoothed and cleaned by an extended dwell in dry hydrogen at high temperature where evaporation of Al_2O_3 and impurities take place.

The many observations, developments, and innovations in translucent alumina now make possible consistent fabrication of arc envelopes with substantially improved transmittance compared to 25 years ago when manufacturing started. Total transmittance of 97%, measured by an integrating sphere, and in-line transmittance of 70% measured on 1-mm thick polished specimens with a sensor detecting light in 30 deg. cone angle at 488 nm, is now routinely achieved. Note that the quoted in-line transmittance is 92% that of polished sapphire. The best specimen prepared in the author's laboratory by sintering, isostatic hot-pressing, and annealing gave 97.5%. The light transmittance of the alumina arc envelope is thus very satisfactory, and room for its impovement is quite limited.

3. SODIUM ATTACK OF ALUMINA ARC ENVELOPES IN SERVICE

HPS arc tubes undergo chemical change in service as result of interaction of their components with sodium vapor and with each other. In the present context the most notable is the formation of sodium aluminates, i.e., NaAlO₂, Na₂O.11Al₂O₃, Na₂O.5Al₂O₃, and other beta alumina-like compounds after long service or at increased power. These reactions mainly slowed down development of HPS lamps with high color rendition, miniature HPS lamps, and other promising derivatives. One can distinguish at least two different modes of sodium attack: the first is formation of beta alumina and NaAlO₂ on the inside wall of the arc tube and illustrated in Figure 2[33, 34], and the second is its nucleation and growth at alumina grain boundaries inside the wall, usually in the hottest section [35] as shown in Figure 3.

The graph in Figure 4 gives the equilibrium vapor pressure of sodium as a function of temperature for the reaction between Al_2O_3 and Na(v) to Na₂O.11Al₂O₃ and to NaAlO₂ and Al. This graph has been based on free energy data by Elrefaie and Smeltzer [36], and although data for beta-alumina from other sources differs it does not qualitatively change the picture. The data was reviewed by Vrught [37], who predicts that for a given sodium vapor pressuc determined by the reservoir temperature and amalgam composition, alumina may react to aluminum and aluminates only in the colder sections of tube below 1050 and 750° C, respectively. Aged arc tubes analyses and model experiments seem to confirm, at least qualitatively, the calculations [38, 39, 40]. Both NaAlO₂ and beta alumina form in the tubes, the latter in the form of lath-like crystals on the inner surface, but do not usually penetrate deep into



Fig. 4. Equilibrium sodium vapor pressure as a function of temperature for the formation of sodium aluminates by the indicated reactions.

the body. Even lamps with sapphire tubing showed slight attack [33, 41]. Aluminum, the other reaction product, was reported in the lamp's spectrum by Reints Bok [42]; however, it was not found in the amalgam where it would be expected to concentrate. It appears that the rate of these reactions is sufficiently slow that only limited attack occurs and is restricted essentially only to the surface. Increased Na vapor pressure, increased temperature, and an imposed electric field with negative bias accelerate the reaction [44]. Well-designed lamps, however, operate 25000 h and more without severe attack. It has been considered, alternatively, that the formation of beta alumina inside the tube is not due to the reaction of sodium with alumina but is the consequence of the presence of a source of oxygen in the arc tube, i.e., caused by the reaction:

$xAl_2O_3 + 2Na + 1/2O_2 = Na_2O.xAl_2O_3$

The possible oxygen sources were discussed by Luthra [43].

Prud'homme van Reine [44], Hing [45], Datta and Grossman [46] and others have implicated impurities, particularly Ca, of accelerating or catalyzing the sodium attack of PCA, perhaps because of the formation of stable binary or ternary compounds and/or strong segregation of Ca in Al_2O_3 grain boundaries. The promoting effect of Ca and Mg on beta alumina formation was confirmed in model experiments by Hodge [47] and Saris [48]. Calcium is a common contaminant of Al_2O_3 and is practically always present at a level of 10 ot 30 ppm in the starting composition. This amount, when conserved, is sufficient to form a monolayer in the grain boundaries. CaO is also a component of the seal and may be transported over the inner surface in the sealing operation, so that its presence cannot be entirely avoided. It has been observed that PCAs of different origin or with different sintering history (vacuum vs. hydrogen) respond by different rates of sodium attack [49]. Confirmation is needed to determine if such observations can be connected to subtle changes in chemistry.

Beta alumina of different morphology has been oberved at grain boundaries in the walls of aged arc tubes or after over wattage operation and is believed to be a consequence of another mode of attack by sodium. Isolated precipitates form in their early stages at grain junctions and are detectable only by TEM (Figure 3). In more advanced stages massive precipitation can take place and form a network of this phase [35]. It is sometimes observable as "white spots" by the naked eye and is accompanied by microcrakcing that ultimately leads to sodium leakage and lamp failure. The process is usually also accompanied by outer envelope darkening which has been attributed to a deposit of alumina, aluminum, and sodium [43, 50]. This reaction is also accelerated by increased wall temperature, sodium vapor pressure and dramatically by a field imposed usually by a fungsten wire wound around the arc tube and connected to an extra leadin. Under these latter circumstances, lamp failure may occur in a fraction of an hour [44, 51] and is accompanied by formation of dark spots.

The formation of beta alumina requires a source of oxygen to proceed. It was speculated that the oxygen could be provided by thermal dissociation of alumina in the grain boundaries near the outer surface of the arc tube. Aluminum ions would discharge at the surface and collect on the outer envelope while oxygen ions would diffuse as defects along grain boundaries inward becouse of chemical potential gradient and/or electric field, react with sodium ions that diffuse in opposite direction through alumina grain boundaries and form beta alumina in situ. This process could be rate limited by diffusion of either species i.e. oxygen or sodium, while the actual transport rates could be impurity-controlled. The model would easily account for the observed effect of the imposed electric field. Aluminum was indeed detected along with alumina and sodium in the deposit on the darkened outer lamp envelopes. A substantial volume change associated with the formation of beta alumina induces microcracking and/or pore formation and accelerates the degradation catastrofically leading to massive sodium leakage.

The above simple model however has not been corroborated by recent observations. (H. Sockel, University Erlangen-Nurenberg, personal communication). Results of oxygen diffusion studies suggest that oxygen diffusion through beta allumina forming at grain boundaries on the inside wall and propagating as thin wedges into the body of the tube, is controlling the rate of lamp degradation. Consequently extrinsic internal oxygen sources as analysed by Luthra (43), should be looked at very closely.

4. ALTERNATIVE ARC ENVELOPE MATERIALS

Single crystal sapphire was considered as lamp material when tubing became available following the invention of edge-defined crystal growth in the early sixties. Many experimental lamps were built and studied, but high price and other problems discouraged their use. Crack propagation in sapphire was frequently enhanced by low angle grain boundaries present in the tubes. Sealing was compounded by thermal expansion anisotropy, and overheating of the seals was observed to be caused by radiation being piped in by the clear sapphire wall. In addition transmittance was sometimes reduced because of molybdenum metal dispersed in the crystal during pulling.

The chemical resitance of oxides to attack by sodium is given to the first appoximation, by their free energies of formation. In Figure 5 we have plotted data for ΔG of three candidate materials, MgAl₂O₃,

 $\rm Y_2O_3, ~and ~Al_5\rm Y_2O_{12}$ (YAG) over the temperature range of interest [52, 53]. ΔG of all three exceeds ΔG of Al₂O₃ i.e., are more stable compounds than alumina and should be able to operate at higher temperatures and sodium vapor pressures. Yttria, for instance, should be stable at sodium vapor pressure of 1.1 Mpa at 1200° C [54]. Because all three of these materials are cubic, they all have the potential of becoming transparent and of yielding higher transmittance at finer grain sizes where optically anisotropic alumina scatters light caused by birefringence. Indeed, transparent polycrystalline magnesium aluminate and yttria have been fabricated into domes with superb light transmittance [55]. An additional advantage of yttria, pointed out by Waymouth [56], is its cutoff further into the IR spectrum, which brings about lower IR emission and consequently higher wall temperatures than alumina under identical conditions. This advantage could result in a higher efficiency lamp.

Mechanical and optical properties of these substances are found in Table I.

In the preparation of higly translucent polycrystalline tubing from these materials various additions were used to promote sintering and control grain growth. With binary oxides this task is even more difficult because in addition to securing very high purity of the starting powders, one must also control stoichiometry that is compounded by different volatilities of the oxide components. The problem of impurities may be somewhat alleviated in YAG because of its garnet structure which can accommodate an appreciable amount of a variety of cations. Mechanical properties, particularly strength and fracture toughness, would be expected to be somewhat lower in these materials than in alumina because of lower elastic moduli (see Table I). This aspect of these properties has been indeed obsered; however, these properties are microstructure-sensitive and may be therefore modified and optimized by refining the grains and other fabrication variables. Such appears to be the case with $Al_5Y_2O_{12}$ where an MOR value of 410 MPa was reported for a 4-um grain size translucent body [57], i.e., substantially more than MOR of a typical arc tube alumina.

A very informative recent review of polycrystalline ceramics for optical application was done by Rhodes [58]. Although testing of these materials has been in progress for some time information on lamp performance is scant, certainly in part because of the need to optimize every system. Differences in thermal expansion may require modifications of the seal and metal composition or a change in design.

Polycrystalline magnesium aluminate spinel showed rapid attack by sodlium vapor on in-lamp testing and massive formation of intergranular beta



Fig.5. Free energy of formation of arc tube materials from elements per gatom of oxygen.

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Properties	of Ceramics	for HPS Lar	np Arc Tubes	
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Property and Unit	Al_2O_3	Y_2O_3	MgAl ₂ O ₄	$\mathrm{Y}_{3}\mathrm{Al}_{5}\mathrm{O}_{12}$
Crystal structure	Н	С	С	С
Density, gm/cc	3.99	5.13	3.59	4.56
Melting point, °C	2050	2456	2135	1970
Elastic modulus, GPa	405	164	277	290
Thermal Expansion Coef \times 10-6/K at 1000° C	9.0	8.9	9.8	9.0
Thermal expansion anisotropy \times 10.6/K	0.9	0.0	0.0	0_0
Thermal conductivity at 300K W/cm/K	0.39	0.27	0.25	0.13
Hardness, GPa	19.6	7.2	16.1	15 0
Bend strength, MPa	300	150	180	410
Fracture toughness, MPam ^{-1/2}	3.5	1.7	1.8	1.7-3.1
Refractive index, n	1.77	1.93	1.71	1.84
$\Delta n/n, \%$	0.45	0.0	0.0	0.0
Dielectric constant	9.6	11.25	8.6	11.7
I.R. cutoff, µm	6.4	8.7	6.5	6.1

alumina, $Na_2O.5Al_2O_3$ [33]. Whether or not this action was the consequence of intrinsic chemical properties, such as structural relationship of spinel and beta alumina, or an interaction with dopants used to improve sintering of the spinel (Ca and LiF) is uncertain. Single crystals of spinel are remarkably resistant to attack by sodium [59]. The performance of spinel was nevertheless disappointing, and further testing was abandoned.

Many compositions were investigated in the development of transparent yttria ceramic. ZrO₂, ThO₂, HfO₂, Al₂O₃, SrO and MgO [60, 61, 62, 63, 64] were reported as dopants assisting densification, but pure, undoped Y_2O_3 was also successfully sintered to transparency [54]. An elegant principle of grain growth control was explored by Rhodes [65]. His composition, a Y_2O_3 -La₂O₃ solid solution, offers an easier and more effective control through a transient solid second phase formation that precipitates at the sintering temperature, exerts control of grain growth, and dissolves subsequently at lower temperature on annealing. This process takes advantage of a unique phase relation in this system.

Additions of ZrO2 and ThO2, and perhaps other oxides to yttria, cannot be used for lamp envelopes becaause they increase the oxygen partial pressure at which Y_2O_3 turns black – the result of formation of charged oxygen defects [60, 66]. In pure Y_2O_3 this oxygen pressure is low enough so that under conditions of HPS lamp operation no darkening occurs.

Most of the reported yttria materials were coarsegrained ceramics with grains 100 μ m and more and with relatively low strength. A process for sintering of pure Y_2O_3 to a fine grained, strong, and transparent body is still under development. This challenge has been obviously met with yttrium aluminum gamet. or YAG, the most recent entry to the family of materials explored for arc envelopes [67, 68]. Transparent YAG was prepared from presynthesized stimum alu minate with additions of 500 to 1000 ppm of SiO₂. MgO, or both, as sintering aids and/or grain growth control. Its sintering was done in vacuum or in hydrogen and gave a single phase material except for small alumina rich precipitates observable by TFM, which perhaps assisted in keeping the grains small, 3 to 1 μm [69]. Strength depended on sintering conditions and reached an impressive 640 MPa (MOR) Along with satisfactory fracture toughness, 3 $MPam^{-1/2}$ and excellent sodium vapor resistance, this material has certainly real potential.

The use of more chemically stable ceramic materials for arc tubes in order to make possible better and more efficient lamps memoral logical approach However, when or if there will be HFS lamps with arc tubes of alternative ceramics will depend on the credibility these materials will earn from engineers designing new lamps, on the costs of these new ceramics and on the progress in the development of other competing lamp system.

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Fig. 1. Pore cluster at the core of a grain in translucent PCA. Thin section, bright field.







Fig. 2. Beta alumina formed at the inside wall of an HPS are tube at about one-third of its length in an overwattage test (a,b) Bundles of Na₂O.11Al₂O₃ crystals (c) Section showing penetration of the crystals into the PCA body.



Fig. 3. Beta alumina formation at grain boundaries of an experimental HPS lamp (a) Early stage of formation of an isolated $Na_2 O.11 Al_2 O_3$ formation. Notice pores associated with the particle. TEM, bright field (b) Massive formation of the aluminate phase in a failed lamp. Bright field.