

## ARAŞTIRMA MAKALESİ/RESEARCH ARTICLE

### HIGH TEMPERATURE THERMAL STABILITY OF MULTI-CATION DOPED $\alpha$ -SIALON CERAMICS CONTAINING YTTERBIUM

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#### ABSTRACT

Recently, it has been found that the phase composition and microstructure of  $\alpha$ - $\beta$  sialon ceramics is greatly affected by heat treatment procedures when rare earth oxides are used as sintering additives. The  $\alpha$ -sialon phase is only stable at high ( $\geq 1650^\circ\text{C}$ ) temperatures and transforms to rare earth rich intergranular phases plus  $\beta$ -sialon at lower temperatures. The present paper describes similar studies carried out on multi-cation doped  $\alpha$ -sialon ceramics where ytterbium as the sole sintering additive and present with other (Sr-, La-, Ce-, Nd-) cations, the resulting  $\alpha$ -sialon products are fully resistant towards  $\alpha \leftrightarrow \beta$  transformation when heat treated at  $1450^\circ\text{C}$ . Clearly the  $\alpha \leftrightarrow \beta$  sialon transformation temperature for ytterbium  $\alpha$ -sialons is below  $1450^\circ\text{C}$ , the conventional heat-treatment temperature used for most of the current research on rare earth densified  $\alpha$ -sialons.

**Keywords:**  $\alpha$ -sialon, multi-cation dopants, ytterbium, thermal stability.

### YİTERBİYUM İÇEREN ÇOK KATYONLU $\alpha$ -SIALON SERAMİKLERİNİN YÜKSEK SICAKLIK KARARLILIĞI

#### ÖZ

Nadir element oksit katkılı  $\alpha$ - $\beta$  sialon seramiklerinde oluşan faz kompozisyonlarının ve mikroyapılarının sinterleme sonrası yapılan ısı işlemlerden önemli ölçüde etkilendiği yakın zamanda yapılan çalışmalarda bulunmuştur. Bu çalışmalarda  $\alpha$ -sialon fazının sadece  $1650^\circ\text{C}$ 'nin üzerindeki sıcaklıklarda kararlı olduğu, daha düşük sıcaklıklarda ise  $\beta$ -sialon ve nadir element içeren tane sınırları fazlarına dönüştüğü gözlemlenmiştir. Bu çalışmada ytterbiyum temel katyon olmak üzere çok katyon içeren (Sr-, La-, Ce-, Nd-)  $\alpha$ -sialon seramikleri üzerine sinterleme sonrası yapılan ısı işlem sonuçları tartışılmıştır.  $1450^\circ\text{C}$ 'de yapılan ısı işlem sonuçları Yb- katyonlarını içeren  $\alpha$ -sialon seramiklerinin  $\alpha \leftrightarrow \beta$  sialon dönüşümüne uğramadığını ve bu katyonları içeren sistemlerde  $\alpha \leftrightarrow \beta$  sialon dönüşüm sıcaklığının  $1450^\circ\text{C}$ 'nin çok altında olabileceğini göstermiştir.

**Anahtar Kelimeler:**  $\alpha$ -sialon, çok katyon katkılı, ytterbiyum, termal kararlılık.

#### 1. INTRODUCTION

$\text{Si}_3\text{N}_4$  based materials are one of the most extensively studied structural ceramics for high temperature applications. It is generally assumed in silicon nitride processing that the product assemblage at firing temperatures is preserved on cooling down to room temperature, but with minor changes to secondary phases e.g. liquid crystallisation or conversion into a glass.

This is based on the assumption that the high temperature products are equilibrium phases under the given experimental conditions, and phases that are stable at high temperatures, are normally stable at room temperature. However, this is not a universal truth, and systems do exist in which phase transformations occur on cooling. These transformations are generally not easy to observe because no characterisation work is carried out at high temperatures.

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The  $\alpha$ -sialon phase, isostructural with  $\alpha$ -Si<sub>3</sub>N<sub>4</sub>, contains two large isolated interstices where some metal ions can be accommodated. The general formula of  $\alpha$ -sialon is represented as  $M_xSi_{12-m-n}Al_{m+n}O_nN_{16-n}$ , where M is one of the cations Li, Mg, Ca, Y and most rare earths (excluding La- and Eu-), m is the number of Si-N bonds in  $\alpha$ -Si<sub>3</sub>N<sub>4</sub> replaced by Al-N; n is the number of Si-N bonds in  $\alpha$ -Si<sub>3</sub>N<sub>4</sub> replaced by Al-O; and x is equal to m divided by the valency of the M cation.  $\alpha$ -SiAlON ceramics offer two advantages over  $\beta$ -SiAlON and  $\beta$ -silicon nitride (i) the metal oxide present in the starting mixture promotes the formation of liquid phase which in turn enhances the densification of the material. Moreover, at sintering temperatures these metal cations can be incorporated into  $\alpha$ -SiAlON structure and hence these type of ceramics provide the possibility of producing single-phase SiAlON ceramics with minimum grain boundary phase; (ii) the material processed as explained in (i) shows increased hardness. Therefore,  $\alpha$ -sialon ceramics have received considerable interest over the last 20 years.

Recent studies on rare earth densified  $\alpha$ -sialon ceramics have shown that the resulting  $\alpha$ -sialon product, present either as a single phase or in conjunction with  $\beta$ -sialon, is unstable when heat-treated at lower (1350-1600°C) temperatures, and transforms to a mixture of  $\beta$ -sialon plus other crystalline or liquid metal sialon phases (Mandal et al, 1993). This provides a convenient mechanism for controlling the mechanical properties of the final material. However, this transformation can only be used beneficially in applications where the maximum service temperature is below the transformation temperature of the grain boundary glass (1000°C). High temperature properties, especially oxidation and creep resistance, significantly deteriorate above this temperature because of the residual glass.

Our previous work and research in other laboratories has concentrated on achieving an understanding of the mechanism of  $\alpha \leftrightarrow \beta$  sialon transformation (Mandal and Thompson, 1995; Shen et al, 1996; Zhao et al, 1996; Mandal et al, 1997; Mandal and Thompson, 1997; Falk et al, 1997). A combination of X-ray and microstructural observations on sintered and heat-treated rare earth densified mixed  $\alpha$ - $\beta$  sialon and  $\alpha$ -sialon ceramic composites has shown that the amount and viscosity of liquid phase; type of sintering additive; heat treatment temperature and time; type of crystalline grain boundary phases; presence of  $\beta$ -sialon grains are all important factors which affect the transformation. However, a full understanding of the transformation mechanism is still not available.

In the present work,  $\alpha$ -sialon compositions have been densified by capsul free Sinter/HIPing using

multi-cations with ytterbium as a principal cation. The resulting materials have been heat treated at 1450°C for up to 1 week to observe the  $\alpha \rightarrow \beta$  sialon transformation and also the crystallisation behaviour. The effect of multi-cations on the  $\alpha \rightarrow \beta$  sialon transformation is discussed.

## 2. EXPERIMENTAL

The overall compositions of the prepared starting powder mixtures corresponded to single-phase  $\alpha$ -sialon with  $m=1.25$  and  $n=1.15$ . Starting powder mixtures were prepared by using Si<sub>3</sub>N<sub>4</sub>, AlN, Al<sub>2</sub>O<sub>3</sub> and M<sub>x</sub>O<sub>y</sub> (M=Sr, La, Ce, Nd, Yb or mixtures). The raw materials used were silicon nitride (UBE-E10), aluminium nitride (HC Starck-Berlin, Grade C), aluminum oxide (Alcoa, Grade A16SG), M<sub>x</sub>O<sub>y</sub> (99.9%, Alfa Rare Earth Products). When calculating the compositions, 1.4% O and 1.6% O (the manufacturer's specifications) present on the surfaces of Si<sub>3</sub>N<sub>4</sub> and AlN respectively were taken in account. Powder mixtures were prepared by wet milling under ethanol in a planetary ball mill. The milled slurries were dried in a rotary evaporation dryer, resulting in weak agglomerates which easily could be passed through a 125  $\mu$ m sieve. Specimens were compacted into pellets by pressing uniaxially and then isostatically under 600 MPa. The green pellets were capsul free Sinter/HIPed in ASEA QIH6 HIP-unit at 1800°C (2 hours at 1 MPa and 1 hour at 50 MPa argon gas pressure). Heat treatment has been carried out for all samples in an molybdenum heating element furnace (ASTRO) at 1450°C for up to 168 hours (1 week) under nitrogen atmosphere. Product phases were characterised by X-ray diffraction (XRD) using a diffractometer. After application of carbon coating, polished surfaces of sintered samples were examined in SEM by using back scattered mode with EDX facilities.

**Table 1. Densities and XRD results of Sintered and Heat Treated Samples.**

Sintering Additive	Density (g/cm <sup>3</sup> )	XRD Results (as sintered)	XRD Results (as heat-treated)
Yb <sub>2</sub> O <sub>3</sub> -Nd <sub>2</sub> O <sub>3</sub>	3.439	$\alpha$ -SiAlON, 21R	$\alpha$ -SiAlON, 21R, M'
Yb <sub>2</sub> O <sub>3</sub> -CeO <sub>2</sub>	3.451	$\alpha$ -SiAlON, 21R	$\alpha$ -SiAlON, 21R, JEM
Yb <sub>2</sub> O <sub>3</sub> -La <sub>2</sub> O <sub>3</sub>	3.443	$\alpha$ -SiAlON, 21R	$\alpha$ -SiAlON, 21R, N
Yb <sub>2</sub> O <sub>3</sub> -SrO	3.388	$\alpha$ -SiAlON, 21R	$\alpha$ -SiAlON, 21R, S

M' =  $\approx$ Nd<sub>2</sub>Si<sub>2</sub>AlO<sub>4</sub>N<sub>3</sub>, JEM =  $\approx$ CeSi<sub>5</sub>Al<sub>2</sub>O<sub>1</sub>N<sub>9</sub>,

S =  $\approx$ SrO1.3Al<sub>2</sub>O<sub>3</sub>0.7Si<sub>2</sub>N<sub>2</sub>O and N = La<sub>3</sub>Si<sub>8</sub>O<sub>4</sub>N<sub>11</sub>

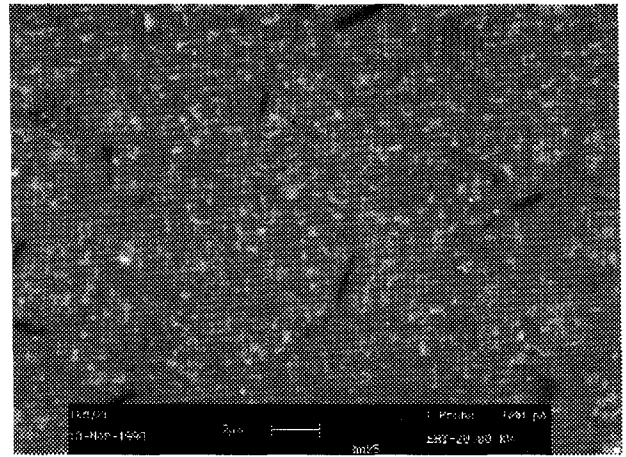
### 3. RESULTS

The composition  $m=1.25$ ,  $n=1.15$  was selected for this study because this is close to the minimum oxygen limit of  $\alpha$ -sialon compositions for easy investigation (i.e. excluding the use of metal nitrides as starting compounds because of their expense and sensitivity to hydrolysis). Capsul-free post-HIPing were applied to samples containing close porosity after pressureless sintering as there was no effect of over pressure on the samples containing open porosity. All the samples were sintered to theoretical densities by Sinter/HIPing. Table 1 gives densities of various samples.

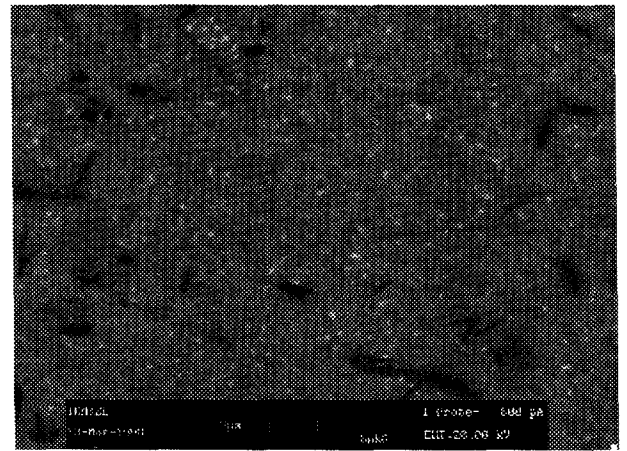
X-ray examination of the sintered samples showed that the cooling rate was sufficiently high to prevent crystallization of any grain boundary glass. The results for the sintered and heat treated samples are given in Table 1. In all samples,  $\alpha$ -sialon was the predominant phase after sintering and minor amount of 21R polytypoid phase was also appeared in all samples.

In spite of the mean cation size of Yb-Sr (1.00 Å), Yb-La (0.97 Å), Yb-Ce (0.95 Å), which are quite large to stabilize  $\alpha$ -sialon (from literature), they seem to produce pure  $\alpha$ -sialon. This can be explained in terms of large  $\alpha$ -sialon stability region for Yb, which has the smallest rare earth cation (0.864 Å) and the  $\alpha$ -sialon stability increases with decreasing the cation size.

A typical back-scattered SEM images of the samples densified with  $\text{Nd}_2\text{O}_3\text{-Yb}_2\text{O}_3$ , and  $\text{La}_2\text{O}_3\text{-Yb}_2\text{O}_3$  are given in Figure 1. The micrographs very clearly distinguish between the various phases, because the contrast is due to the mean atomic number: 21R grains (which contain no cations as sintering additives) are black and more needlelike; the  $\alpha$ -sialon grains (which contain a small amount of cations that are sintering additives) are gray and more equiaxed; and the cation-rich crystalline or glassy phases appear fine-grained and white, because of the high Z-cation content. The microstructures were homogeneous and consisted of equiaxed  $\alpha$ -sialon grains. Elongated  $\alpha$ -sialon grains also were observed in the sample that contained  $\text{La}_2\text{O}_3\text{-Yb}_2\text{O}_3$ . In the case of  $\text{Nd}_2\text{O}_3\text{-Yb}_2\text{O}_3$  sample, most of the cations (Nd- and Yb-) are incorporated into  $\alpha$ -sialon structure and therefore only very few grain boundary phases are visible. EDX spectra for the corresponding samples are given in Figure 2. The analysis of  $\alpha$ -sialon grains in different parts of the specimen showed very similar results that Nd-, Ce-, La-, and Sr- cations can be stabilized into  $\alpha$ -sialon grains with Yb-. The microstructures and the EDX spectras of Ce-, La- and Sr- doped samples are presented and discussed in detail elsewhere (Mandal and Hoffmann, 1999).



(a)

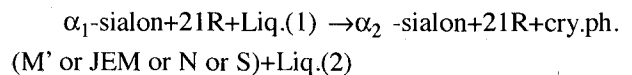


(b)

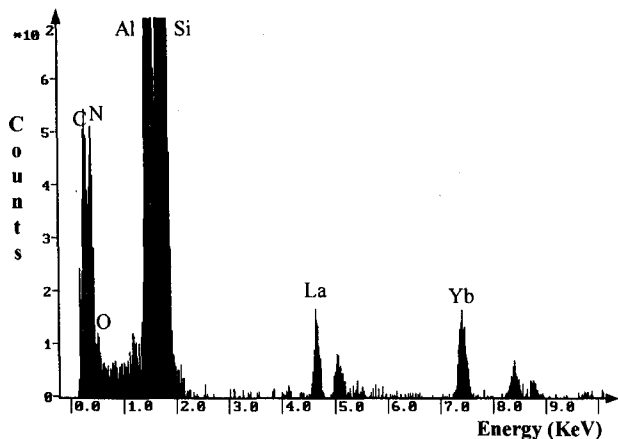
**FIGURE 1. Back-scattered SEM micrographs of the (a)  $\text{La}_2\text{O}_3\text{-Yb}_2\text{O}_3$ , (b)  $\text{Nd}_2\text{O}_3\text{-Yb}_2\text{O}_3$  samples after sintering.**

Subsequent prolonged heat treatments of samples were carried out at 1450°C in nitrogen atmosphere for 168 hours (1 week) in order to study the effects of heat treatments on the  $\alpha \leftrightarrow \beta$  sialon transformation and also the crystallisation behaviour.

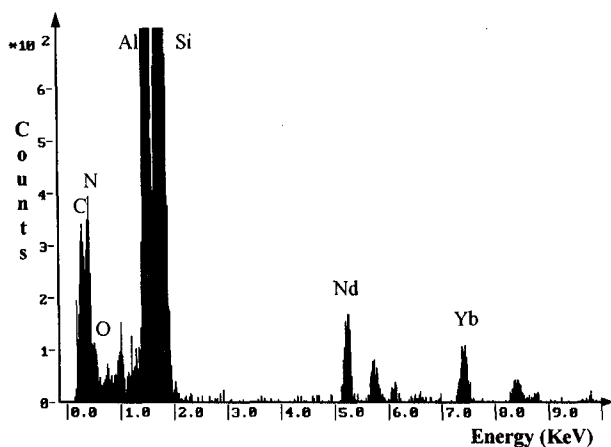
In the heat treated samples, suprisingly, there were no  $\alpha \leftrightarrow \beta$  sialon transformation was observed and therefore  $\alpha$ -sialon was the only matrix phase observed after 168 hours (1 week) of heat treatment. In addition,  $M'$  ( $\approx \text{Nd}_2\text{Si}_2\text{AlO}_4\text{N}_3$ ), JEM ( $\approx \text{CeSi}_5\text{Al}_2\text{O}_{11}\text{N}_9$ ), S- ( $\approx \text{SrO}_{1.3}\text{Al}_2\text{O}_3\text{O}_7\text{Si}_2\text{N}_2\text{O}$ ) and N- ( $\text{La}_3\text{Si}_8\text{O}_4\text{N}_{11}$ ), phases were also observed, in increasing amounts along with a decrease in the  $\alpha$ -sialon unit cell dimensions as the heat treatment time increased, for Nd-, Ce-, La- and Sr- doped samples respectively. The suggested equation for this reaction is:



EDX spectra for the samples after 168 hours heat treatment also proved that Nd-, Ce, La- and Sr-cations still remain inside the  $\alpha$ -sialon grains.



(a)



(b)

FIGURE 2. EDX spectra of the  $\alpha$ -sialon regions of (a)  $\text{La}_2\text{O}_3$ - $\text{Yb}_2\text{O}_3$ , (b)  $\text{Nd}_2\text{O}_3$ - $\text{Yb}_2\text{O}_3$  samples after sintering.

It is highly plausible that  $\text{Sr}^{2+}$  ( $r=1.12 \text{ \AA}$ ) and  $\text{La}^{3+}$  ( $r=1.06 \text{ \AA}$ ) do not form an  $\alpha$ -sialon on its own and although  $\text{Ce}^{3+}$  ( $r=1.03 \text{ \AA}$ ) and  $\text{Nd}^{3+}$  ( $r=0.99 \text{ \AA}$ ) form an  $\alpha$ -sialon which is only stable at high temperatures (above  $1650^\circ\text{C}$ ) and transforms to  $\beta$ -sialon at lower temperatures ( $1300$ - $1600^\circ\text{C}$ ) but all these large cations can produce a very stable  $\alpha$ -sialon phase with Yb.

Many of the results obtained in the present study are not consistent with conclusions previously deduced for rare earth stabilised  $\alpha$ -sialon materials. It is clear that ytterbium based  $\alpha$ -sialons in which ytterbium is present jointly with others, do not undergo  $\alpha \rightarrow \beta$  sialon transformation when heat-treated at  $1450^\circ\text{C}$ . This conclusion confirms the improved thermal stability of ytterbium  $\alpha$ -sialons compared with other rare earth analogues, and probably also correlates with the increased size of the single-phase  $\alpha$ -sialon phase-field in the ytterbium sialon system.

A more convenient way of understanding the present results is in terms of  $\alpha \leftrightarrow \beta$  transformation tempera-

ture (Mandal et al, 1999). This has not been defined precisely in previous studies, even though work on an  $m=1.5$ ,  $n=1.5$  Sm  $\alpha$ -sialon composition showed this to be  $\approx 1600^\circ\text{C}$ . Previous results are consistent in showing that the transformation temperature decreases with increasing atomic number in the rare earth  $\alpha$ -sialon series, and in fact certain Yb  $\alpha$ -sialon compositions are stable with respect to transformation. The present study shows that for all the ytterbium based  $\alpha$ -sialons studied, the transformation temperature is below  $1450^\circ\text{C}$ .

#### 4. CONCLUSIONS

$\alpha \leftrightarrow \beta$  sialon transformation has been explored in a wide range of multi-cation  $\alpha$ -sialon ceramics in which ytterbium is the principal cation. In all cases  $\alpha \rightarrow \beta$  transformation was not observed at  $1450^\circ\text{C}$ , and ytterbium acted as a stabiliser to inhibit transformation in  $\alpha$ -sialon compositions which otherwise would have readily exhibited transformation. It is significant that even small amounts of ytterbium result in significant stabilization in these mixed cation materials. Furthermore, the  $\alpha \leftrightarrow \beta$  sialon transformation temperature for these materials must lie below the lowest heat-treatment temperature used in the present study ( $1450^\circ\text{C}$ ), and further work is needed below this temperature to see whether transformation can be induced under these conditions.

#### ACKNOWLEDGEMENT

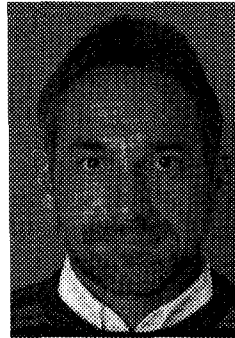
One of the authors (H.M.) would like to acknowledge financial assistance from the Alexander von Humboldt Foundation (AvH, Germany) during the course of this work.

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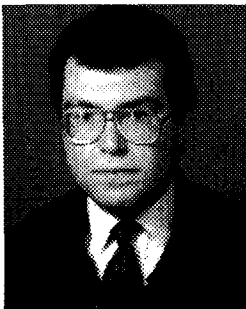
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1998. He is an active publisher, having contributed some 70 papers on the subject of silicon nitride and related nitrogen ceramics. Dr. Mandal's research activities focus on the improvement the refractoriness of nitrogen ceramics, and significant breakthroughs have included above-eutectic heat-treatment, vacuum heat-treatment to totally remove grain-boundary glass, and  $\alpha \leftrightarrow \beta$  sialon transformation to tailor properties of sialon ceramics.



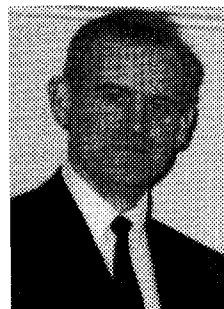
**Michael J. Hoffmann** is a professor for technical ceramics at the University of Karlsruhe and the leader of the Institute of Ceramics in Mechanical Engineering. He received his MS degree in technical mineralogy from the Technical University of Darmstadt and his PhD degree from the University of Stuttgart. He worked for more than

10 years at the Max-Planck-Institute for Metals Research in Stuttgart before he moved to Karlsruhe in 1995. Hoffmann's research activities focus on grain growth phenomena, microstructural development and microstructure-property relationships of silicon nitride and silicon carbide ceramics. He also studied piezoelectric ceramics based on lead zirconate titanate (PZT) and thermal barrier coatings. Hoffmann is the author or co-author of more than 80 technical publications.



**Hasan Mandal** obtained a B.Sc in Metallurgical Engineering from Middle East Technical University in 1987, and carried out research in the Materials Division at the University of Newcastle (1989-1992) obtaining his Ph.D in 1992 for a thesis entitled "Heat Treatment of Sialon Ceramics". He then

started post-doctoral work with Professor Derek P. Thomson at the University of Newcastle, and was part of the successful Newcastle team. He joined the Department of Ceramic Engineering at Anadolu University as an assistant professor in 1994 and become an associate professor in 1996. He appointed as the Head of Department of Ceramic Engineering in 1999. Since 1996, Dr. Mandal has had an active involvement with the Turkish Ceramic Society, holding posts of Member of Board of Directors. He has also played an active role in the European Ceramic Society, being a member of the Executive Council, and more recently Scientific Programme Director of the ECerS VIII meeting which will be held in 2003 in İstanbul. Throughout his career he was awarded numerous awards, including M.E.T.U Prof.Dr. Mustafa N.PARLAR, Educational and Research Foundation, Research Award in 1996, TÜBİTAK (Scientific and Research Council of Turkey), Research Award in



**Derek Thompson** was educated at Christ's College, Cambridge, obtaining a 1st class honours degree in Natural Sciences in 1967. He continued with research studies at Cambridge, receiving his M.A. and then Ph.D in 1972 for a mineralogical thesis entitled "a crystallographic study of hōgbo-mite and related minerals". He

then started post-doctoral work with Professor Ken Jack at the University of Newcastle upon Tyne, and was part of the successful Newcastle team that discovered sialons, initially in the  $\beta$ - and subsequently in the  $\alpha$ -forms. After the retirement of Professor Jack in 1984, Derek took over the leadership of the Nitrogen Ceramics research group, and continued work on sialons, extending this work to include the role of nitrogen in other ceramics (e.g. zirconia and silicon carbide). During the last 15 years, the main thrust of the group has been in improving the refractoriness of nitrogen ceramics, and significant breakthroughs have included above-eu-

tectic heat-treatment, vacuum heat-treatment to totally remove grain-boundary glass, and  $\alpha \leftrightarrow \beta$  sialon transformation to tailor properties of sialon ceramics. He moved up to a Readership in 1990 and then a chair in 1995, both in Engineering Ceramics. Since 1980, Derek has had an active involvement with the UK Institute of Ceramics (now part of the Institute of Materials), holding posts of secretary and later Chairman of the Ceramic Science Committee, and being a member of the Ceramics Industry Division Board, and also the main Council of the Institute of Materials. He has also played an active role in the European Ceramic Society, being a member of the Executive Council, and more recently Programme Director of the ECerS VI meeting held in 1999 in Brighton. He is an active publisher, having contributed some 200 papers on the subject of silicon nitride and related nitrogen ceramics.

Silver medal and Prize (Inst. of Metals) in 1989, and the Centennial Award (Ceram. Soc. Japan) in 1991. His initial work was in the field of metal nitrides, in which he carried out complete structural characterisations of compounds in the iron-nitrogen system, other transition metal - nitrogen systems and also more complex ternary nitride systems. He started working on silicon nitride in the mid 1950s and this led to the discovery of sialon ceramics in 1970-1. He carried out detailed development work on sialons during the 1970s (including work on sialon glasses) which led to the commercial exploitation of these materials in the early 1980s. Since his retirement, he has kept an active interest in the further development of sialon ceramics, especially in the capacity of Consultant for the Cookson group and he has also pursued his interests in metallic nitrides with studies of the magnetic properties of  $\text{Fe}_{16}\text{N}_2$ .



**Kenneth H. Jack** obtained a B.Sc in Chemistry from Durham University in 1939, and an M.Sc in 1944, and carried out research in the Cavendish Laboratory at the University of Cambridge (1947-9) obtaining his Ph.D in 1950. He was a lecturer in Chemistry at Durham University during the periods 1941-45, 1949-52 and

1953-57, the intervening periods being spent as a senior scientific officer at the British Iron & Steel Association (1945-9), and a research engineer at the Westinghouse Corporation (Pittsburgh) in 1952-3. He was then Research Director at Thermal Syndicate from 1957-64, before being appointed Professor of Applied Crystal Chemistry at Newcastle University in 1964, becoming Director of the Wolfson Research Group for High Strength Materials in 1970. He obtained a D.Sc in Materials Science from the University of Cambridge in 1978, and became a fellow of the Royal Society in 1980. He retired in 1984, becoming a Leverhulme Emeritus fellow in 1985-7 and a consultant for the Cookson Group from 1986-94, retaining an active interest in his fields of study and participating in many national and international conferences. Throughout his career he was invited to present many prestigious lectures, and was also awarded numerous medals, including the Saviile Shaw medal (SCI) in 1944, the Sir George Bielby memorial award (IOM, RIC, SCI) in 1951, the Kroll medal (Metals Soc.) in 1979, the Prince of Wales award for Industrial Innovation and Production in 1984 - joint with Dr R.J. Lumby), the Armourers & Brasiers Co. award (RS) in 1988, the World Materials Congress Award (ASM International) in 1988, the A.A.Griffith