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Journal

Journal of materials research, 67(4)

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Publication Date

1990-10-01

Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

Materials & Chemical Sciences Division

National Center for Electron Microscopy

Submitted to Journal of Materials Research

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October 1990

Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098.

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STRUCTURE AND COMPOSITION CHARACTERIZATION OF SUBMICRONIC

MULLITE WHISKERS

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Abstract

Two sets of submicronic mullite whiskers, which have potential applications for fiber reinforced composites or as a thermal insulator, have been characterized to be tetragonal or orthorhombic using x-ray and electron microscopy techniques. The whiskers decompose upon heating under vacuum by a continuous loss of silicon and reduction in oxygen content up to the limit for which pure aluminum metal and α -alumina are formed.

I. Introduction

The only crystalline phase in the aluminosilicate system with a large regime of temperature stability is mullite. The high refractoriness of mullite combined with low thermal expansion and conductivity, low creep rate, good chemical and thermal stability together with good toughness and strength make it an excellent candidate for structural ceramics and electronics applications. The chemical composition of mullite ranges usually from $3\text{Al}_2\text{O}_3:2\text{SiO}_2$ to $2\text{Al}_2\text{O}_3:1\text{SiO}_2$ (hereafter referred to as m:n ratio).

The structure of mullite was derived from sillimanite [1] and has been refined and studied in great detail [2,3,4]. The appropriate formula for mullite is $\text{Al}_2(\text{Al}+2x\text{Si}_{2-2x})\text{O}_{10-x}$. The number of oxygen vacancies per average unit cell x , occurring in the Oc sites, is directly related to the m:n ratio by the relation $x = (2m/n - 2)(2m/n + 1)$. Although the 3:2 mullite was found to be disordered [5], a higher alumina content leads to the ordering of oxygen vacancies, with the formation of an incommensurate modulation whose periodicity varies

continuously with composition [6,7,8]. Antiphase domain structures (APBs) result from this ordering [9], with superstructure reflections observable in the diffraction patterns [10,11,12]. APBs are either parallel to (100) [10,12] or, for mullites richer in alumina, almost parallel to (601) [11,12].

Mullite is orthorhombic (pbam space group). The lattice parameters a , b , and c vary almost linearly with composition. In fact, for increasing Al_2O_3 content (from m:n 3:2 to 5:2), a decreases from 0.77nm to 0.75nm while b increases from 0.75 to 0.77nm [13]. This shows the possibility of tetragonal mullite formation at the composition where a and b have the same value. In a few instances tetragonal mullite has been reported and the tetragonality was assumed to be process-dependent [14,15]. So far the solubility limit of mullite has been extended by rapid quenching from 70.6mole% up to 76mole% Al_2O_3 . However, the existence of silica-free α -alumina [16,17,18] with the structure similar to that of mullite shows the possibility of an extended solid solubility limit up to the corundum stability field.

In this work two sets of submicronic mullite whiskers have been studied by x-ray diffraction, and transmission, analytical, and high resolution electron microscopy. Produced as rigid felts, mullite whiskers have promise for application as a preform for fibre reinforced ceramic (or metal) matrix composites, or as a thermal insulator. Hence, the full characterization of structure, composition and thermal stability of these whiskers is of utmost importance.

II. Experimental Details

The first set of mullite whiskers (Mu-1) produced by reaction sintering of topaz [19] was obtained from the N.S.W.C., Silver Spring, MD. (courtesy of I. Talmy). The second set of mullite whiskers (Mu-2) were obtained from ancient Indian "wootz" crucibles used for iron melting (courtesy of T. L. Lowe). These whiskers were formed by the crystallization from the glass during the firing cycle of a mixture of clay and rice husk constituting the internal wall of a refractory vessel [20]. The Mu-2 whiskers were obtained

by HF-leaching of the undesired silica-rich glassy matrix. High temperature annealing was performed in molybdenum crucibles under a vacuum of 10^{-6} atm. X-ray diffraction (XRD) analysis was carried out using a Siemens θ - 2θ diffractogram. Transmission electron microscopy (TEM) and microanalysis were performed on finely crushed powders of the mullite whiskers, suspended on a holey carbon Cu-grid, using a JEOL-200CX analytical microscope equipped with x-ray detectors and a Kevex 8000 system for energy dispersive x-ray spectroscopy (EDS) quantitative microanalysis. High resolution electron microscopy was performed at the National Center for Electron Microscopy using the Atomic Resolution Microscope at an operating voltage of 800kV, and computer simulation was carried out at the Center using the CEMPAS image simulation package.

III. Results and Discussion

1) Analysis of the as-received whiskers

i) Morphology and Microstructure. A low magnification electron micrograph of the whiskers presented in Fig. 1 shows the large difference in size distribution between the two sets of mullites. The average width of the Mu-1 whiskers lies within a range of 200nm to 400nm whereas the Mu-2 whiskers are much finer with an average width of 10nm to 30nm. For both sets of mullite, either a cylindrical or faceted morphology was observed. The high resolution image along the [110] direction of a Mu-2 whisker presented in Fig. 2 (for two different values of defocus), shows the *c*-axis growth habit of the whisker which, in fact, is found to be a common characteristic of all the whiskers. The insets are the optical diffractograms and the computer simulated image for a 5nm thick, 3:2 mullite, in agreement with the measured composition, as discussed below.

ii) Structure. Besides the size distribution, both the structure and the chemical composition of the two sets of mullite differ from one another. On the XRD pattern of the Mu-1 presented in Fig. 3a, the absence of line splitting at the 120/210, 240/420, etc. reflections means that the length of the *a* and *b* axes are equal within these resolution limits.

Thus the Mu-1 whiskers have a tetragonal symmetry instead of the commonly observed orthorhombic structure as is the case for the Mu-2 whiskers (see Fig. 3b).

Tetragonal mullite has been observed earlier [14,15]. The formation of the tetragonal phase is predicted for a range of high alumina content mullites [13]. However, the reported compositions, as well as the compositions determined for the present whiskers (see below), do not fall within the predicted range. It was suggested [15] that tetragonality may either be due to the development of domain structures or twinning of orthorhombic structural units with alternation of a and b along the c -axis or due to a different atomic arrangement of the unit cell. The high resolution images obtained from the as-received Mu-1 tetragonal whiskers viewed along the a or b -axis did not show the existence of any domain structure nor twinning. In order to verify the second possibility, and to image possible atomic position or atomic arrangement changes within the unit cell, it is necessary to observe the structure with an electron beam parallel to the c -axis. Unfortunately, we have not yet been successful in obtaining a cross section of the whiskers which have the c -axis growth habit.

iii) Composition. The whisker morphology allows a direct measurement of thickness to be made in the STEM mode leading to the correction for the absorption factor during EDS chemical analysis. Given the large thickness range of Mu-1 whiskers, a systematic EDS-point analysis was carried out in order to elucidate the eventual size dependence of the whisker composition. Hence, the compositions were recorded for whiskers in size increments of 30nm to 50nm. Within each given range of thickness at least four measurements have been performed on four different whiskers. After collecting all the data, it was noticed that representative classes of composition versus thickness fell into a few large categories as reported in Table 1. Mu-1 has a composition of 3:2 (or very close) for either very thin whiskers of thickness $d < 100\text{nm}$ or very thick whiskers with $d > 500\text{nm}$. The middle size whiskers ranging between 100nm to 300nm vary in composition between 3:2 and 2:1, namely $m:n$ of 1.63 to 1.83. The error values reported

in parentheses are not due to systematical measurement errors, but represent the variation in composition found within each range of thickness for different mullites. The detected composition of the Mu-2 whiskers corresponds systematically to the 3:2 mullite with an occasional detection of titanium up to 1.2at%. Titanium is one of the clay's constituents and has some solid solubility in mullite [21], with the substitution of octahedral aluminum cations by titanium. It has been shown that Ti in solution shifts the composition of mullite towards 1.7:1 [22].

2. Analysis of the Annealed Whiskers

i) Composition. Annealing of the mullite whiskers under vacuum was performed at three different temperatures (1200, 1400 and 1500°C). The same global thermal behavior was observed with the phase transformation occurring at a faster rate with increasing temperature. Here the sequence of transformations occurring upon heating is presented. Fig. 4 summarizes the compositional changes during the annealing. As annealing proceeds, the silicon content of the whiskers decreases drastically until it reaches almost zero. The amount of silica loss was measured for different thicknesses using the same systematic EDS analysis as that used for the as-received whiskers.

For comparison, Table 1 provides the detected compositions of a large number of Mu-1 whiskers annealed for 2h at 1400°C. Besides the very thick whiskers, the loss of silica is rather large especially for whiskers less than 300nm thick. Within the range of 100 to 300nm thickness, compositions up to 79.6% mole Al_2O_3 which correspond to 88.6%Al were detected. This large aluminum-content mullite with $x = 0.658$ is beyond the highest aluminous mullite ever reported [13]. The obtained compositions of 97 to 100% aluminum were in fact from barely recognizable whiskers which, as will be discussed later, is due to the sintering of the whiskers.

Line scans were performed through the width of several cylindrical whiskers annealed 2h at 2400°C (the cylindrical morphology allows accurate thickness measurements to be made for the absorption correction) in order to register an eventual

variation in composition. A continuous decrease in silicon content was recorded when moving from the center toward the edges. The maximum variation in Si registered for a thickness of around 300nm was 1.5at%. For thicker whiskers, the variation in composition was not significant (<0.5at%).

(ii) Structure. The XRD pattern of Mu-1 whiskers annealed for 12h at 1500°C is presented in Fig. 5. Two major changes are observed. An obvious shoulder is formed on the left hand side of the 210/120 peak and a new set of lines has appeared which belongs to the hexagonal α -Al₂O₃ corundum.

The formation of the shoulder is either due to the existence of a large range of composition, and hence a variation in lattice parameter, or it may simply be due to the transformation of the metastable tetragonal to the stable orthorhombic phase upon annealing. It was possible to obtain a cross-section from an annealed whisker which allowed confirmation of the latter hypothesis to be made, namely that the observed shoulder on the 120/210 peak is due to the tetragonal to orthorhombic transformation. The high resolution image of this cross-section is presented in Fig. 6. Along the *c*-axis the orthorhombic nature of the whisker is clearly observed [5]. Furthermore, the faceted nature ((110) facets) of this whisker is well demonstrated.

iii) Morphology and Microstructure. During heat treatment, the morphology and the microstructure of the whiskers alter and go through different changes. Many whiskers develop small internal pores of various sizes. Pores as large as 30nm in size were observed. A pair of TEM micrographs of a typical internal cavity formed within an annealed Mu-1 whisker is presented in Fig. 7. The pair of images are taken respectively for overfocused and underfocused conditions in order to confirm that the contrast (black or white halo around the pore) is actually due to a cavity. The cavities are presumably Kirkendal voids left behind during the diffusion of aluminum and silicon through the lattice from the core towards the free surfaces. The formation of Kirkendal voids along with the fact that the concentration variation between the center and the edges of the whiskers is not

very large, indicates that the diffusion rates are of the same order of magnitude as the evaporation rate.

Another microstructural change noticed for alumina-rich mullites was twinning of the whiskers. In Fig. 8, a typical conventional micrograph of a group of alumina-rich twinned whiskers (obtained from the Mu-1 set annealed 6h at 1400°C) is presented. The high resolution image of a twinned whisker in Fig. 9 shows the formation of surface ledges oriented along high index planes. The whisker is 92nm thick and contains a large number of fine scale twins probably related to the change in volume which accompanies the transformation to a higher aluminous mullite. This result is in agreement with previous unit cell measurements [24]. For this direction, near the [121] zone axis of the mullite, the twin plane is inclined with respect to the beam direction. In fact the twinning occurs on the (001) planes and is better observed with the incident electron beam parallel for example to the *a* or *b* axis.

The (100) view of the lattice image shown in Fig. 10 contains the (001) microtwinning texture together with the APBs parallel to (601) planes. The existence of such APBs is the signature of oxygen vacancy ordering [10,11,12]. This suggests that not only does silicon diffuse out of the whiskers, but also that oxygen vacancies are formed during vacuum annealing. The fact that the APBs are along (601) is further evidence that this whisker is a highly aluminous mullite ($x > 0.5$ [12]) with oxygen vacancies formed at the Oc-sites. In fact, for a wide thickness range, under this defocus condition (close to Scherzer), the characteristic white dot pattern arranged in bands parallel to the APBs represent the projected positions of the Oc-site vacancy channels (parallel to the *b*-axis) in agreement with previous observations on bulk mullites [12]. The optical diffraction pattern (Fig. 10b) obtained from a twinned lamella shows the superlattice reflections formed as pairs of the $h01$ reciprocal lattice section along the $[601]^*$. The separation of split maxima is $S = 0.258a^*$ which corresponds to $x = 0.53$ if linear extrapolation of previous data is used [13]. In fact, in previously reported data, *S* decreases linearly from $S = 0.395$ to $S =$

0.288 when increasing x from $x = 0.25$ to $x = 0.47$, but then deviates from linearity for higher x values. However, S values as low as $S = 0.226a^*$ are found in the present work which, using the linear extrapolation, should correspond to $x = 0.6$ in agreement with the composition measured by EDS (Table 1). In terms of the stereochemical criteria, the highest detected aluminous mullite whiskers (with $x = 0.658$), corresponds to the theoretical upper limit of oxygen removal ($x = 2/3$) which is one-third of the original Oc-atoms [12]. For lower alumina content whiskers, in agreement with previous observations [10, 11, 12, 13], the superstructure spots occur parallel to a^* , at $1/2c^*$.

Besides the ordered oxygen vacancy superlattice, no extra superlattice reflections were observed indicating that there is no silicon vacancy ordering if silicon vacancies exist.

The final stage of the decomposition during high temperature annealing is the formation of alumina as was observed by x-ray diffraction. However, during TEM observation, no evidence of heterogeneous nucleation of any of the alumina polymorphs at the surface or within an annealed mullite whisker was found. The only observed form of alumina, as is shown in Fig. 11, was in the form of agglomerates of sintered whiskers with globular morphology. This material is the stable hexagonal α - Al_2O_3 , as verified by the x-ray data. The sintered powders were strain free and single crystal as shown by convergent beam diffractions patterns (see Fig. 11b). The fact that the sintered powders coexist with mullite whiskers may suggest that alumina forms during the exothermic sintering reaction which should occur concomitantly with the reduction of the remaining silica. The sintering reaction is accompanied by a local rise in temperature leading to fast grain growth (grain boundary migration) and hence the formation of single crystals.

It is emphasized that very few whiskers, with freshly nucleated silica-free phase, formed at their tips have been observed. Surprisingly the diffraction analysis of the precipitates correspond to pure aluminum metal. A TEM image of such a whisker is presented in Fig. 12. The aluminum is better observed in the dark field image (see Fig. 12b) formed with the $(220)\text{Al}$ reflection. The $[001]$ and $[111]$ zone axis of the aluminum

precipitates are presented respectively in Figs. 12c and d. The [111] aluminum is parallel to the [110] mullite and the (010) aluminum planes are parallel to the *c*-axis of mullite ($d_{202} = d_{002} = 0.146\text{nm}$). The formation of aluminum may be due to the existence of strong reducing elements coming from the evaporation of some reactive material which has previously contaminated the furnace or the crucible. However, strong reduction in oxygen has been reported in thin regions of oxides under high-vacuum using high resolution electron microscopy [25,26]. The electron beam induced surface reaction under vacuum owes to the formation of pure Sn metal in Sn_2O [27] which subsequently amorphizes and reoxidizes when irradiation continues. Thus it may not be excluded that under a strongly reducing atmosphere, aluminum metal forms at the tip of the whiskers, melting almost instantaneously (when considering the high annealing temperatures) but then subsequently reoxidizing by reaction with oxygen released from adjacent areas or from other whiskers. The end result is the formation of the stable form of alumina. However, these speculations need to be further investigated.

IV. Conclusions

Analytical and high resolution electron microscopy together with x-ray diffraction has allowed a full characterization to be made of two sets of submicronic mullite whiskers. One set of whiskers had the metastable tetragonal symmetry which transformed upon heating to the stable orthorhombic phase. The other set was orthorhombic. Further annealing of both sets under vacuum results in a continuous loss in silicon and finally the appearance of the $\alpha\text{-Al}_2\text{O}_3$ phase in the form of sintered powders. Mullite whiskers of composition as high as 79.6mole% Al_2O_3 were detected. The loss in silica is accompanied by a loss of oxygen. The appearance of antiphase boundaries and twinning indicate the occurrence of oxygen vacancy ordering. The detection of pure aluminum metal at the tips of single annealed mullite whiskers suggests the existence of a strong reducing atmosphere. An alternative path for α -alumina formation may be through the reoxidation of the aluminum.

Acknowledgements

This research was partially supported by the Naval Surface Warfare Center under contract No. NSWC 60921-90-C-0276, and we thank Dr. I. G. Talmy of NSWC for providing the whiskers. Helpful discussions with Professor A. W. Searcy and T. L. Lowe, who also provided whiskers, are appreciated. Dr. A. P. Tomsia collaborated on the heat treatments. The facilities at the National Center for Electron Microscopy used in this work are supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Science Division, U. S. Department of Energy under Contract No. DE-AC-03-76SF00098.

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Table 1. Compositional data deduced by EDS measurements from the As-Received (As.) Mu-1 whiskers and after annealing (Ann.) for 2h at 1400°C, where m:n is the alumina to silica ratio and x is the number of oxygen ions missing per unit cell, $x = [2(m/n) - 2]/[2(m/n) + 1]$.

	Thickness [nm]	Al [at%]	m/n	x	Al ₂ O ₃ [mole%]	Al ₂ O ₃ [wt%]
As Received	d < 100	75.0(±0.1)	1.5	0.251	60.06	70.48
Annealed	d<100	80.8(±0.1)	2.1	0.424	67.78	76.64
As-Received	100 < d < 300	77.5(±1.0)	1.72	0.325	63.27	73.09
Annealed	100 < d < 300	85.6(±3.0)	2.97	0.568	74.83	81.87
As-Received	300 < d < 500	77.5(±1.0)	1.72	0.325	63.27	73.09
Annealed	300 < d < 500	78.1(±1.0)	1.78	0.343	64.07	73.73
As-Received	500 < d	75.3(±0.2)	1.52	0.26	60.38	70.74
Annealed	500 < d	75.2(±0.2)	1.52	0.26	60.26	70.64

Figure Captions

- Fig. 1. Low magnification of (a) Mu-1 and (b) Mu-2 whiskers showing the large difference in thickness of the two sets of mullite whiskers.
- Fig. 2. High resolution and the simulated image of a Mu-2 whisker along the (110) zone axis at (a) $df = -60\text{nm}$ and (b) $df = -100\text{nm}$. The simulated thickness is 5nm for a 3:2 mullite.
- Fig. 3. X-ray diffraction patterns of (a) the tetragonal Mu-1 and (b) the orthorhombic Mu-2 whiskers.
- Fig. 4. The evolution of the composition upon annealing of the mullite whiskers under vacuum.
- Fig. 5. X-ray diffraction pattern of Mu-1 whiskers annealed for 12h at 1500°C showing the splitting of the 120/210 reflection and the new set of lines which belong to corundum ($\alpha\text{-Al}_2\text{O}_3$).
- Fig. 6. High resolution micrograph of the (001) cross section of a faceted mullite whisker after 2h annealing at 1400°C showing the orthorhombic structure.
- Fig. 7. (a) Overfocused and (b) underfocused micrograph showing the typical contrast for the cavity formed during annealing of the mullite whiskers.
- Fig. 8. TEM micrograph of alumina rich mullite whiskers subjected to severe twinning after 6h annealing at 1400°C .
- Fig. 9. High resolution image near the [121] zone axis of a twinned whisker showing the formation of surface ledges. The twin plane is inclined with respect to the beam direction.
- Fig. 10. High resolution image along the [010] zone axis of a microtwinning whisker, containing APBs inclined against (601). The twin direction is the c -axis. The insets are the optical diffractogram from a single twin lamella and from a large area containing few twin planes, showing the twinned superstructure spots and streaking along (001) due to the irregularly spaced (001) twin lamellae.

Fig. 11. (a) TEM micrograph of α -Al₂O₃ phase observed as sintered powders and (b) CBED of the hexagonal phase along the [0111] zone axis.

Fig. 12. (a) Bright field micrograph of a single annealed mullite whisker containing freshly nucleated pure aluminum metal phase at its tip; (b) the corresponding dark field micrograph imaged with the (220) reflection of the aluminum; (c) and (d) are respectively the (001) and the (111) zone axis of the aluminum precipitates. The (111) zone axis of the aluminum is parallel to the (110) zone axis of the mullite.

XBB 904-3023

Fig. 2a

Fig. 2b

XBB 904-3024

XBL 905-1984

Fig. 3

XBL 905-1892

Fig. 4

XBL 905-1893

Fig. 5

XBB 905-4324

Fig. 6

XBB 905-4320

Fig. 7

Fig. 8

XBB 905-4323

Fig. 9

XBB 905-4322

XBB 905-4321

Fig. 10

XBB 906-4584

Fig. 11

Fig. 12

XBB 906-4553

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