Journal of the European Ceramic Society **5** (1989) 75–80

State of the Art

Stuijts Memorial Lecture 1989: The Mastery of Microstructure*

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(Received 16 August 1989, revised version received and accepted 28 August 1989)

Abstract

In the light of his experience in the successful development of ceramic microstructures to match specific electromagnetic applications, Professor Stuijts proposed a number of themes in 1972 as being ripe for progress in the general task of exploiting ceramics for advanced engineering use Three of these topics are considered with emphasis on the work that has been done and on current attitudes The three themes are powder preparation, the importance of technological factors, and the need for valid generalised pictures of complex processes

Three additional approaches that have proved fruitful in the intervening years in tackling the issues that were identified as being of concern are the use of mapping techniques, the use of computer modelling and the use of detailed microstructural characterisation notably in the form of electron microscopy

The combination of methods has allowed the development of a more systematic approach to ceramic processing along the lines that Professor Stuijts was seeking

Angesichts seiner Erfahrungen in der erfolgreichen Entwicklung keramischer Mikrostrukturen fur den gezielten Einsatz bei elektromagnetischen Anwendungen, hat Prof Stuijts 1972 eine Anzahl von Themen vorgeschlagen, die fur die Auswertung von Keramiken, speziell in Hinblick auf die Anwendung als Hochleistungswerkstoff, Fortschritte in generellen Fragen brachten Drei dieser Themen sind aus der bisher geleisteten, aber auch aus der noch laufenden Arbeit besonders hervorzuheben Die drei Themen sind die Pulverherstellung, die Bedeutung technologischer Faktoren und die Notwendigkeit fur allgemeingultige Schemata fur komplexe Prozesse

Drei weitere Vorgehensmethoden, die sich in den vergangenen Jahren bei der Losung der gestellten Aufgaben als erfolgreich erwiesen haben, sind die Anwendung von Mapping-Techniken, Computermodellen und detaillierten Charakterisierungsmethoden fur die Mikrostruktur, wobei hier speziell die Elektronenmikroskopie zu erwahnen ist

Die Kombination dieser Methoden ermöglichte die Entwicklung einer systematischeren Vorgehensweise bei der Keramikherstellung in der Richtung, die Prof Stuijts verfolgte

Grâce à son expérience acquise dans le développement de microstructures céramiques et à sa réussite à les adapter à des applications électromagnétiques particulières, le Professeur Stuijts a proposé en 1972 d'approfondir un certain nombre de thèmes pour faire progresser l'exploitation technologique des céramiques Trois de ces thèmes sont considérés en mettant l'accent sur le travail effectué jusqu'à présent et sur les attitudes actuelles Ce sont la préparation de la poudre, l'importance des facteurs technologiques et le besoin de rapporter des procédés complexes à des représentations généralisées simples

Trois approches supplémentaires se sont révélées fructueuses dans les années intermédiaires pour avoir su cerner les problèmes jugés préoccupants le recours aux techniques d'image élémentaires, l'utilisation de la modélisation informatique et l'utilisation de la caractérisation microstructurale détaillée notamment grâce à la microscopie électronique

La combination des méthodes a permis le développement d'une approche plus systématique de l'élaboration des céramiques dans la direction recherchée par le Professeur Stuijts

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*Given on the occasion of the First Meeting of the European Ceramic Society, 20 June 1989 in Maastricht, The Netherlands

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The properties and the fitness for use of ceramic materials are largely determined by microstructure The character, quantity and distribution of crystalline grains, porosity, intergranular phases and interfaces, must be chosen to provide the properties that are required in the application of the component, whether as building brick, cutting tool, electronic substrate or turbine blade The fabrication of the chosen microstructure is then the central concern of ceramic processing science

Professor Stuijts earned the respect and admiration of colleagues for his work over many years in the design and fabrication of microstructures Two aspects made his contribution especially persuasive The first was that, as someone working equally happily in the academic and industrial communities, he was able to bring ideas and understanding through to the point of successfully exploited products, notably in the magnetics area [1] The second was that he was able to use his knowledge of the science underlying the processes of microstructural change by developing, with colleagues at the Philips Research Laboratories, fabrication methods which actually produced the desired microstructures [2] This systematic linking of ceramic processing science and industrial materials development gave an excellent example of the path to follow in bringing ceramics to the full range of applications for which their properties make them suited

In surveying the subject in 1972, Professor Stuijts[1] selected a number of themes as 'issues of emerging importance' in relation to processing (Table 1) The intervening years have served to underline the quality of his prediction, and it is instructive to review the progress that has been made in following the proposed directions Three examples can be taken for closer consideration

The most dramatic improvement in recent years in the general level and reliability of ceramic processing is to be attributed to the great advances made in *powder preparation* Largely through the increased emphasis on chemical preparation methods,[3] but also through the continuing refinement of mechanical methods, there is now a range of commercially available powders offering a degree of convenience (e.g. reduced sintering temperature, increased shaping flexibility) and consistency that brings corresponding advantages in the properties and reliability of the final sintered products

Table 1. Issues (1972)

Powder quality
Compact characterisation
Technologically important factors
Sound models experiments and materials
Grain boundary mobility
Generalising statements on sintering phenomena
Liquid phase sintering

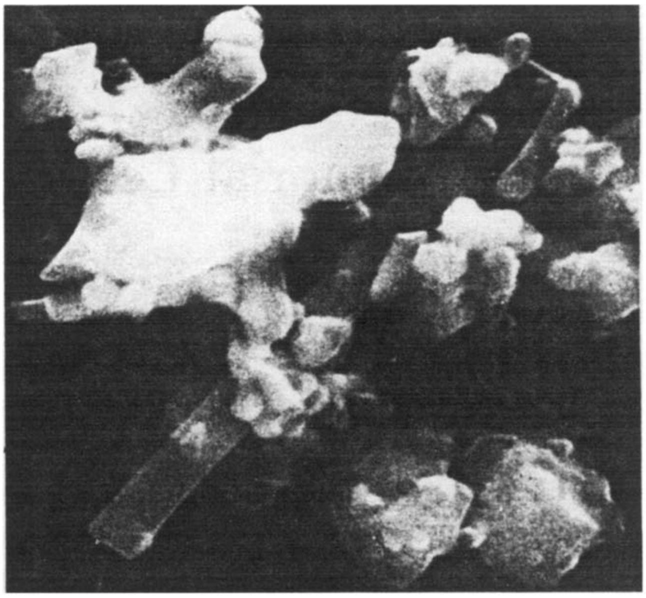

Fig. 1. A silicon nitride powder containing variations of particle shape and size sufficient to result in severe packing differences (accidents) in pressed components

If, as with ceramic microstructures in general, progress is seen as the simultaneous simplification of structure (accidental variations are avoided) coupled with increasing complexity (deliberate design features are incorporated), then the improvements in powder preparation have thus far been predominantly concerned with the simplification step. Chemical methods offer the opportunity of avoiding structural accident (Fig 1) and of attaining[4] uniform, homogeneous powder arrays (Fig 2) The step of introducing deliberate complexity (Table 2), as, for example, in coated powders, where sintering additives are introduced not homogeneously but rather in concentrated form at the particle surface where the activity is required, remains for the most part to be exploited It is a particularly promising direction for future development

The recognition of *technological factors*, i e. of factors in addition to the physical or chemical nature of the material itself, has grown in recent years and has contributed to the measured advance of ceramics into such critical engineering applications as heat engine components When innovation is based on materials substitution into a mature technology, as in the case of diesel engines, then cost considerations are likely to make the task difficult. As seen in Table 3, the motor vehicle application sets exacting targets[5] for materials which must be

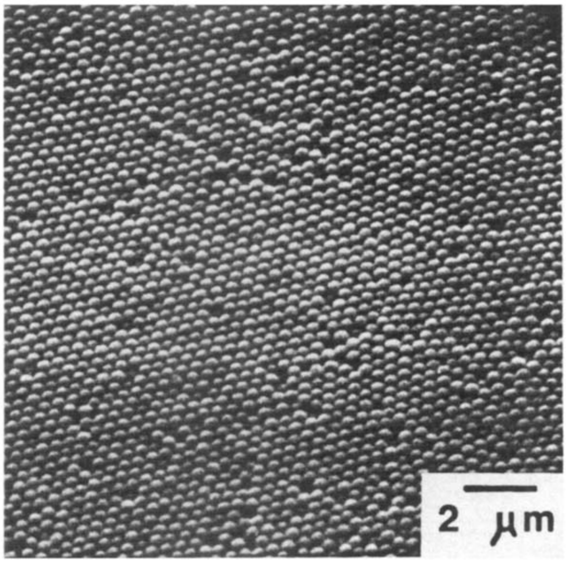

Fig. 2 Monodisperse SiO_2 powder[4] shows the extent to which particle shape and size can be controlled by chemical processing routes

Table 3. Approximate costs of 1 g of material used in different applications

Motor vehicles	DM	0.01
Aircraft	DM	1.00
Spacecraft	DM	100.00

carefully designed and reliably fabricated. The cost margins for aircraft use are more generous, but the requirement for reliability then becomes absolute. Such considerations emphasise the importance of the motor vehicle gas turbine as a means of bringing ceramics into large volume use in engines, here, again, technological factors, such as the relative performance of turbines in respect to emissions levels, can be expected to be among the decisive factors.

A problem that was seen by Professor Stuijts as vital in ensuring the practical value of processing science was the need to present advances in knowledge in terms of *generalised statements on phenomena*, which could then be used beyond the narrower confines of the conditions set by a particular group of experiments. This need has been fully recognised in connection with sintering processes, a fruitful development has been the emphasis on the two major patterns of microstructural change that can occur during sintering, namely on densification and on coarsening (grain growth), and on the selection of processing conditions to influence the relative rates of these processes with the ambition of producing desired microstructures. The current debates on back-stresses during the sintering of inhomogeneous[6–10] or composite microstructures, or on the rôle of grain growth in modifying the thermodynamic driving force for densification,[11,12] are directly concerned with such generalised statements.

Table 2. Development directions for ceramic powders

Character	*Conventional target*	*Development*
Size	0.1–1.0 μm	Finer sizes
Size distribution	Monodisperse	Size variation
Shape	Spherical	Shape variation
Chemical homogeneity	High	Coatings
Crystallinity		Monocrystal
Density	High	
Cost	Low	

In contrast, the value of precise atomistic descriptions of mechanisms has become increasingly questioned. The rôle of impurities, the change in mechanism expected for changing conditions of temperature, grain size or dopant level, and the ambiguity of interpretation associated with kinetic measurements, have all tended to undermine belief in the value of exact identification of the defect chemical species involved in rate controlling phenomena. The very complexity of the magnesia rôle in the sintering of alumina[13–17] has, for example, reduced expectations that arguments by analogy will prove fruitful in respect to additive effects on the development of microstructure. As noted by Descartes,[18] complexity is to be distrusted (Fig. 3), and the merely probable is to be rejected. The search for understanding has, in the light of the ever increasing richness of detail,[19] proved elusive (Fig. 4).

Three factors have, however, been found helpful in the search for the generalised statements required for the systematic development of microstructures. The first is the use of *maps* as summaries of accumulated experience.[20] Kinetic expressions for process rates by different mechanisms allow identification of the ranges of processing conditions, e.g. temperature, applied pressure (hipping), or grain size, within which given mechanisms are preferred, the results are then represented schematically in diagrams or maps. These are then compared with experiments in an iterative and accumulative manner until a growing degree of confidence can be placed in the result. Such maps can summarise process mechanisms,[20] microstructural conditions[21] or advocated process techniques.[12] As a means of generalising experience, this approach has shown considerable value.

Voyant que la philosophie a ete cultivee par les plus excellents esprits qui aient vecu depuis plusieurs siecles, et que néanmoins il ne s'y trouve encore aucune chose dont on ne dispute, et considerant combien il peut y avoir de diverses opinions touchant une même matiere, qui soient soutenues par des gens doctes, sans qu'il y en puisse avoir jamais plus d'une seule qui soit vraie, je reputais presque pour faux tout ce qui n'etait que vraisemblable.

Although the best minds have turned to philosophy over the centuries, everything remains in dispute The learned hold so many explanations where only one can be really valid that I decided that all which was proposed as probable must be taken as false

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Car il me semblait que je pourrais rencontrer plus de verité dans les raisonnements que chacun fait touchant les affaires qui lui importent, et dont l'évenement le doit punir bientôt apres s'il a mal jugé, que dans ceux que fait un homme de lettres dans son cabinet touchant des spéculations qui ne produisent aucun effet, et qui ne lui sont d'autre consequence sinon que peut-être il en tirera d'autant plus de vanite qu'elles seront plus eloignées du sens commun, à cause qu'il aura dû employer d'autant plus d'esprit et d'artifice à tâcher de les rendre vraisemblables

Models are more likely to be valid where the matter is of direct concern to the author and where he is accountable for the consequences than where he is himself unaffected, in the latter instance he will draw the more credit the more his ideas are distant from common sense (he will need more wit and cunning to make them seem probable)

Fig. 3. Descartes, *Discours de la Methode* (1637)

A second factor is the increasing rôle of *computer modelling* This has now been exploited for the description of structures such as surfaces[22] or grain boundaries,[23] for the evaluation of the energies associated with defect formation and movement,[24] and, more recently, for the simulation of microstructural change itself either in densification[25] or in grain growth [26] The effectiveness of low energy boundaries in seeding abnormal grain growth has, for example, been strikingly shown,[26] supporting the early view of Professor Stuijts and colleagues[27] that boundaries associated with liquid phases are to be seen as an important element in initiating the process

The third factor has been the recognition of the primacy of microstructural examination in laying the basis for understanding, a view greatly reinforced by the dramatic contribution of *electron microscopy* There are notable examples, as in the transformation toughening of zirconia (Fig 5),[28] in the grain boundary engineering of silicon nitride,[30] and in current work with fibre/whisker composites,[31] where detailed observation of local physical or mechanical events has proved critical in the building of generalised models capable of contributing to predictive and systematic design

The combination of these factors has allowed the development of an excellent basis for the understanding of microstructural change, particularly where the detailed steps of *physical or mechanical changes* can themselves be directly observed The less persuasive aspect of current understanding lies in the interpretation of *chemical influences* Progress can be made by a combination of detailed characterisation of the influence of a particular additive coupled with a comparative survey of the influence of many different additives, the specific behaviour and the recognition of classes of additive behaviour can then provide a basis for prediction. This has been employed, for example, in describing interfacial segregation,[32] the control of grain boundary mobility,[33] and the promotion of densification [34] Such studies provide a basis for intelligent trial and error, as opposed to the random variety, in the selection of additives, this is a branch of the subject where major advance can be expected in coming years, with the greater levels of impurity control and instrumental chemical resolution that are becoming available

Habe nun, ach, Philosophie, Juristerei und Medizin
und leider auch Theologie
durchaus studiert mit heißem Bemuhn
da steh ich nun, ich armer Tor
und bin so klug als wie zuvor

I have studied every art
and stay as wise as at the start

Fig. 4. Goethe, *Faust* (1798)

In 1979, Professor Stuijts[35] noted that 'ceramic processing science had not yet led to the development of first principles capable of guiding materials development' The current position is that such first principles are now, for physical and mechanical factors, largely in place, for chemical factors there remains much progress to be made. A major task can now be seen, however, in *industrialising* the skills that have been won, i e in upscaling processing methods and matching them to applications with sufficient reliability and cost effectiveness so that the long-discussed potential of ceramics is fully realised In reaching the mastery of microstructure that such a development will require, much satisfaction can be

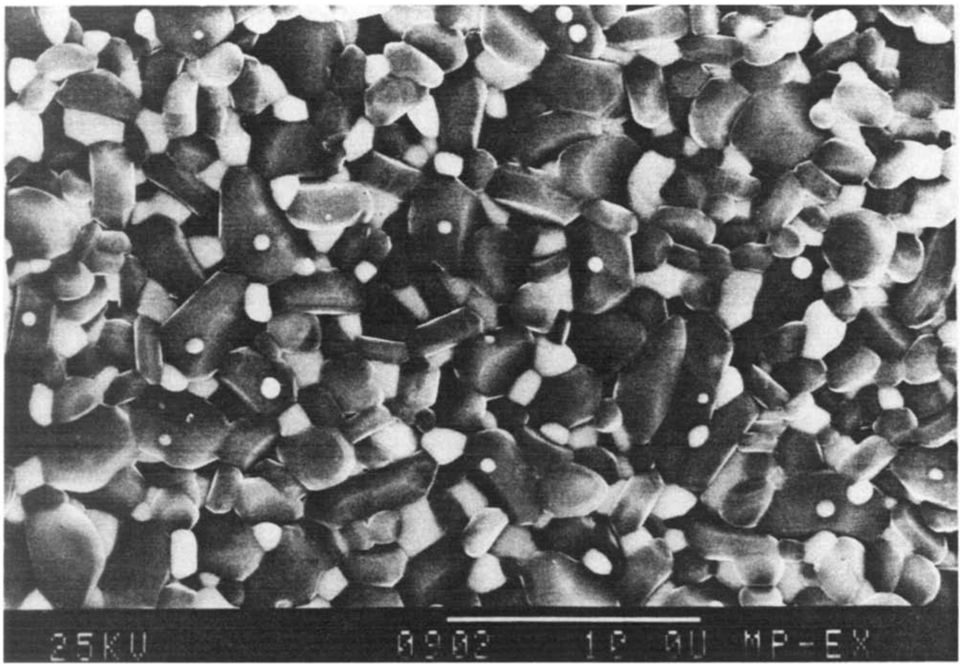

Fig. 5 Dramatic mechanical property improvements have resulted from the designed incorporation of zirconia inclusions into ceramic matrices, as shown here (SEM) for alumina (Courtesy N Claussen) The transmission electron microscope has proved crucial for the understanding of the local transformation events in the zirconia particles [29]

gained from the progress of recent years, it will remain true, however, as Professor Stuijts himself emphasised, that 'there is still enough room for imagination in materials development' [35]

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