more fully in a future article.) Included among the conditions thought to be of autoimmune origin are rheumatoid arthritis, systemic lupus erythematosus (SLE), certain anemias, late-developing diabetes, and multiple sclerosis.

Systemic lupus erythematosus afflicts an estimated 400,000 to 500,000 people; about 80 percent of whom are women, and usually develops between the ages of 20 and 40. It can be mild but in its severe form the disease is both debilitating and life-threatening.

Investigators studying SLE frequently use the New Zealand Black (NZB) mouse. Between the ages of 5 and 9 months, mice of this strain spontaneously develop an autoimmune disease that closely resembles human SLE. The animals have LE cells (a type of white blood cell characteristic of SLE), antibodies to nucleic acids, hemolytic anemia, and immune complex glomerulonephritis, an inflammation of the kidney caused by deposition of antibody-antigen complexes. About 10 percent of NZB mice also develop a malignant lymphoma analogous to chronic lymphocytic leukemia.

According to Norman Talal, Michael Dauphinee, and their colleagues at the University of California Medical Center, NZB mice progressively lose T cell functions. Even before autoimmunity is clinically detectable, their T cells are developmentally and functionally abnormal. Bach found that thymosin-like activity in serum declines at an abnormally early age in NZB mice; it is negligible by the time they are 2 months old. This implies that the thymosin deficiency causes abnormal T cell development and, consequently, autoimmunity.

Humans with active SLE may also be deficient in thymosin. Edgar Cathcart and Morton Scheinberg of Boston University Hospital showed that lymphocytes from patients with active SLE formed fewer T rosettes in vitro than did those from healthy controls or from patients with rheumatoid arthritis or inactive SLE. The SLE patients had more of what the investigators call "null" cells, that is, cells with neither B nor T marker antigens. Treating the lymphocytes in vitro with thymosin fraction 5 increased the number of T rosettes formed and decreased the number of null cells.

Talal hypothesized that administration of thymosin fraction 5 to NZB mice might correct their T cell deficiencies and prevent the onset of autoimmunity. It did restore to normal one of the aberrant developmental responses of NZB thymocytes; however, thymosin delayed, but did not prevent, the development of autoimmune disease in the animals. Talal said that additional factors probably play a role in the etiology of autoimmunity in NZB mice. These could include genetic disposition and viruses, both of which have been implicated in the etiologies of several autoimmune conditions. Other investigators have shown that NZB thymocytes are infected with murine leukemia virus. Viral infection could destroy the cells or cause them to function abnormally.

Another possibility is that a hormone or hormones not present in fraction 5 may be involved. This preparation did not correct all of Heather's immunodeficiencies either. Other investigators have isolated thymic hormones that are involved in T cell differentiation. For example, Gideon Goldstein,

(*Continued on page 1217*)

Ceramics: Brittle Materials for High Temperature Structures

Two of the world's most pressing problems—the increasing cost and decreasing availability of fossil fuels and the pollution of the environment resulting from their use—could be partially solved if the operating temperature of a heat engine were raised. More useful work for the same amount of fuel consumed would be obtained because of the engine's increased thermal efficiency, and fewer pollution-causing emissions would be released because of the more complete combustion permitted by richer fuel to air ratios.

Unfortunately, even the most exotic high temperature alloys (super alloys containing large amounts of nickel, cobalt, or chromium) cannot withstand temperatures in excess of about 1100°C. However, as a result of advances made over the past 15 years, refractory ceramic materials based on silicon nitride (Si_3N_4) and silicon carbide (SiC) are being considered for use as high temperature structural materials in places where previously only metals would have been used. Most current research is oriented toward ceramic gas turbines for vehicles, stationary power generators, and remotely piloted aircraft.

As compared with metals, ceramics are more resistant to high temperatures, oxidation, corrosion, and erosion, and are quite strong, but they are quite brittle. In particular, when rapid temperature changes occur, ceramics tend to fracture under the resulting thermal stresses, a condition known as thermal shock. Moreover, the virtual absence of plastic deformation that can relieve stress concentrations in brittle materials means that the stresses must be accurately known everywhere in a part, so that design is a formidable problem. As a result, interest in ceramics as structural materials has been a cyclical phenomenon since about 1900.

Interest in silicon carbide and silicon nitride derives from their having a combination of moderate-to-low linear thermal expansion coefficients and high-to-moderate thermal conductivities respectively. These ceramics are thus less susceptible to thermal shock than other such materials. But because both compounds decompose by sublimation at high temperatures, parts cannot be fabricated by casting, thus necessitating recourse to powder metallurgical methods, such as sintering or hot-pressing.

In the mid-1950's, workers at the Admiralty Materials Laboratory in the United Kingdom discovered that relatively dense silicon nitride with good properties could be made by reacting silicon powder in a nitrogen atmosphere at high temperature. The first step in this so-called reaction bonding or reaction sintering process is the compacting of the silicon powder into the desired shape. An organic binder or plasticizing agent (which is later burned off) is mixed with the silicon powder to hold it together and to permit the formation of complex shapes through use of processes such as injection molding, which is common in the plastic industry. This "green" body is heated first to 1350°C and then to 1450°C to bring about the nitridation reaction.

Silicon nitride parts made in this way are about 70 to 85 percent of the maximum possible density and maintain their mechanical strength up to about

1400°C. Pores larger than 10 micrometers seem to act as flaws which can serve as the origin of cracks that lead to fracture.

A somewhat similar process has since been developed for silicon carbide by researchers at the Atomic Energy Authority laboratory at Springfield in the United Kingdom. A mixture of silicon carbide powder and carbon powder is formed compactly to the desired shape. An organic binder may or may not be used. The compacted powder is heated to 1600°C to 1700°C, and molten silicon metal is allowed to infiltrate the powder. The silicon and carbon react to form silicon carbide, which binds the powder particles together.

Silicon carbide made in this manner can reach 100 percent of the theoretical density, but with the disadvantage that some unreacted silicon remains. The silicon melts at about 1410°C thus limiting the use of reaction bonded silicon carbide to temperatures much lower than this.

In the meantime, at the end of the 1950's, workers at The Plessey Company, Ltd., also in the United Kingdom, found that adding small amounts of magnesia (MgO) permitted dense silicon nitride to be made by hot-pressing. Hot-pressing involves simultaneous application of uniaxial pressure (250 atmospheres) and heat (1700°C).

Use of magnesia (about 5 percent by weight) for hot-pressed silicon nitride was further improved by the British company Joseph Lucas Ltd.; by 1970 theirs was the standard way to produce this material. Improvements by the Lucas company basically involved obtaining small grain sizes (the ceramic is crystalline, with the size of the crystalline regions being of the order of 1 micrometer) with a resulting low porosity and high strength.

Hot-pressed silicon nitride has mechanical properties that are superior to those of the reaction bonded material at room temperature. For example, the bending strength is about three times higher. Above about 1200°C, however, the properties degrade. One problem is the onset of creep. Creep is the plastic deformation of a material under a load lower than that normally required to cause the material to fail and can cause significant dimensional charges in parts. A decrease in strength also occurs at high temperatures and may be attributable to the existence of small cracks that grow slowly before reaching a large enough size for fracture to occur.

Work at several laboratories has resulted in a model that ascribes the degradation of mechanical strength at high temperatures to the formation, during hot-pressing, of a glassy magnesium silicate (Mg_2SiO_4) phase between the particles of silicon nitride. This glassy grain boundary phase is thought to soften at elevated temperatures so that the grains can slide past one another, giving rise both to the creep phenomenon and the slow crack growth. The model is still regarded as controversial.

A hot-pressed version of silicon carbide has also been developed with alumina (Al_2O_3) as the densification aid. Hot-pressed silicon carbide is similarly afflicted with a putative vitreous grain boundary phase that limits its service to temperatures below about 1200°C to 1400°C. However, as compared with the silicon nitride, silicon carbide is considerably less susceptible to creep, the rate of creep being 10^3 to 10^4 times lower.

A Peculiar Fibrous Microstructure

Hot-pressed silicon nitride is considerably tougher than silicon carbide. Toughness is different from strength and relates to the resistance to catastrophic crack growth (fracture) under an applied load. According to F. F. Lange of the Westinghouse Research Laboratories, Pittsburgh, Pennsylvania, the toughness of silicon nitride is likely due to its peculiar fibrous microstructure. Silicon carbide consists of more symmetrical grains. This explanation awaits quantitative verification, however, and is not universally accepted by other investigators.

Hot-pressed ceramics and reaction bonded versions actually represent complementary materials. Although the hot-pressed material is denser and stronger than the reaction bonded, hot-pressing is limited to fabricating simple shapes. Moreover, there is considerable shrinkage (at least 50 percent by volume) during hot-pressing. Both considerations result in the need for expensive diamond grinding to bring the hot-pressed parts into final shape. Reaction bonded ceramics, however, can be fabricated into complex shapes relatively inexpensively.

In the last 4 years, a number of improvements in the properties of silicon nitride and silicon carbide have been made. Lange and his co-workers discovered, for example, that impurities play an especially important role in the high temperature properties of hot-pressed ceramics, apparently by affecting the viscosity of the glassy grain boundary silicate phase. Calcium is the worst offender, having deleterious effects at concentrations as low as a few hundred parts per million. Commercial vendors of silicon nitride and silicon carbide now market powders of high purity.

Although hot-pressed silicon nitride made from high purity powder exhibits increased resistance to creep at high temperatures, the properties of the magnesium silicate glass still limit the performance. George Gazza of the Army Materials and Mechanics Research Center (AMMRC), Watertown, Massachusetts, thus began looking for better densification additives than magnesia. After an extensive search, Gazza decided to try yttria (Y_2O_3) because yttrium silicate ought to be more refractory than magnesium silicate.

As it turned out, additions of 10 to 20 percent (by weight) of yttria to silicon nitride powder were found to increase the creep resistance of hot-pressed silicon nitride about 1000 times to nearly that of hot-pressed silicon carbide at 1400°C. Some researchers now think that the mechanism for enhanced high temperature properties is related to the formation of a crystalline yttrium oxynitride phase (or phases) that result when the yttria and the silicon nitride react. Moreover, researchers have found that calcium may not be as detrimental to the properties of hot-pressed silicon nitride when yttria is added.

Roy Rice and his associates at the Naval Research Laboratory have tried using zirconia (ZrO_2) as a densification aid and found more modest beneficial effects on the high temperature properties of silicon nitride. For example, scientists at the Avco Corporation, Lowell, Massachusetts, and the Aerospace Research Laboratories, Wright-Patterson Air Force Base, Ohio, made measurements indicating that the stress-rupture properties and oxidation resistance increased tenfold.

Sintering is similar to hot-pressing in that high temperatures are used to bring about densification of a powder body and binding of the particles together, but it is unlike hot-pressing in that no pressure is applied during the heat treatment. Sintering has not been successfully applied to the making of dense silicon carbide or silicon nitride until recently.

Svante Prochazka of the General Electric Research and Development

Center, Schenectady, New York, has found that additions of less than 1 percent (by weight) of boron and carbon to silicon carbide powder can permit the sintering of dense silicon carbide parts. Although the boron apparently does segregate at the grain boundaries, there is no glassy grain boundary phase. As a result, the high temperature strength of the General Electric silicon carbide is retained up to 1600°C, and creep is undetectable at 1480°C. Although there is some shrinkage during the sintering process, the expensive diamond grinding required for hot-pressed ceramics can be avoided.

Silicon nitride can be sintered to nearly full density as well, if about 50 percent (by weight) alumina is "added" as a sintering aid. The resulting ceramic is actually a family of solid solutions of silicon nitride and alumina and has been given the name "sialon" (Si-Al-O-N). The properties of these ceramics have been studied by K. H. Jack and his colleagues at the University of Newcastle upon Tyne in the United Kingdom and by Y. Oyama and his associates at the Toyota Research Laboratories in Japan, as well as in several laboratories in the United States. The motivation for their study is to be able to combine the desirable properties of hot-pressed silicon nitride with the inexpensiveness and ease of fabrication possible with the sintering process. Despite much enthusiasm early on, no sialons have yet been made with properties comparable to good hot-pressed silicon nitride.

The beneficial aspects of these advances do not, as a rule, change the intrinsically brittle nature of ceramics, nor do they increase the impact strength or fracture toughness. There have, however, been a number of approaches to ameliorating the susceptibility of silicon nitride and especially silicon carbide to fracture.

Since many of the cracks (actually microcracks many of which have dimensions of the order of micrometers) are on the surface of the ceramic part, Henry Kirchner and his co-workers at Ceramic Finishing Company, State College, Pennsylvania, have been trying various surface treatments designed to increase the stress required to cause a surface crack to grow during impact. One approach involves the use of compressive surface stresses, which would tend to cancel stresses occurring during impact or other load. Compressive stresses can be induced by various thermal or chemical means. The bonding of energy absorbing surface layers to the ceramic is another approach being studied.

Composite materials provide an alternative means of altering the properties of ceramics by combining the good properties of the component materials and not the bad. For example, researchers have studied the possibility of dispersing chucks or filaments of silicon carbide as a second phase within silicon nitride in the hopes of combining the toughness of silicon nitride with the superior high temperature creep resistance of silicon carbide.

J. J. Brennan of the United Aircraft Research Laboratories, East Hartford, Connecticut, has been embedding ductile tantalum wires in hot-pressed silicon nitride. At present, silicon nitride made in this way appears to have somewhat lower strength than silicon nitride made without the tantalum wires, and oxidation of the wires can be a problem at high temperatures. However, the resistance to fracture during impact tests is considerably improved, increasing by a factor of 30 in the temperature range from room temperature to 1300°C.

A Silicon Carbide Composite

W. B. Hillig and his co-workers at General Electric have made a silicon carbide composite material. In the GE composite, carbon fibers are shaped appropriately, and the fibers are then infiltrated with molten silicon, which reacts with the carbon, somewhat as in reaction bonded silicon carbide. The result is a composite of silicon carbide fibers in a metallic silicon matrix. This fully dense material can have a range of ductility corresponding to a silicon carbide content of from 20 to 80 percent. The melting point of silicon sets the upper temperature limit for the use of this material.

Silicon carbide and silicon nitride can be prepared by chemical vapor deposition. Unfortunately, because of the difficulties and expense of making large and complex shapes with uniformly good properties, this method has not been widely considered.

R. J. Diefendorf and his associates at Rensselaer Polytechnic Institute have made vapor deposited silicon carbide with about twice the strength of silicon carbide made in other ways. In part, the superior high temperature strength results from the lack of any additives whatsoever, since no densifying step occurs in chemical vapor deposition. Instead a mixture of hydrogen and methyltrichlorosilane (CH_3Cl_3Si) is passed through a furnace where silicon carbide is deposited on the wall of a deposition tube.

When complex shapes are being made, there can be a problem with excess carbon at the grain boundaries. Thus simple shapes in theory are not so difficult to make by chemical vapor deposition, but complex shapes are much harder. Because of this and because of cost considerations, Diefendorf suggests that, if perfected, chemically vapor deposited silicon carbide may be primarily a competitor of hot-pressed or sintered ceramic, but not reaction bonded.

One of the major difficulties in designing with ceramics, in addition to their inherent brittleness, is a statistical nature in their properties which is related to the distribution of imperfections from which cracks leading to fracture can grow. Any given group of ostensibly identical parts may fail at widely differing loads. One concept that has evolved is the strength-probability-time (SPT) diagram. Developed by R. W. Davidge and his associates at the Atomic Energy Research Establishment, Harwell, United Kingdom, the SPT diagram gives the engineer a statistical probability that a part will survive a specified time under a given stress load at a given temperature.

A. G. Evans (now at the Rockwell International Science Center, Thousand Oaks, California) and S. M. Wiederhorn of the National Bureau of Standards, Gaithersburg, Maryland, have taken a different approach by combining fracture measurements interpreted in terms of the slow crack growth model discussed earlier with proof testing in order to calculate minimum failure times for parts that survive the proof test.

The minimum time to fail under a given load can be found from a measured crack velocity and initial crack size. Unfortunately, the cracks are so small that existing nondestructive testing procedures cannot reliably detect them. In a proof test, a load sufficient to cause some samples to fail is applied. The proof stress can be related to the minimum size crack that could result in such a failure and hence to the maximum initial crack size that could be in the remaining samples.

In a second article, the performance of silicon nitride and silicon carbide when used in actual or simulated gas turbine environments will be examined.

—Arthur L. Robinson