Kirk-Othmer Encyclopedia of Chemical Technology.

GRAPHITE, APPLICATIONS OF ARTIFICIAL

1. Aerospace and Nuclear Reactor Applications

Graphite with its exceptional strength and thermal stability at high temperatures is a prime candidate material for many aerospace and nuclear applications. Its properties, through process modifications, are tailorable to meet an array of design criteria for survival under extremely harsh environmental operations.

Graphite is a lightweight structural material that retains most of its mechanical strength even at temperatures in excess of 2200°C. It has a high resistance to thermal shock and exhibits good neutron interaction characteristics and stability under irradiation. The ease of machining and commercial availability of graphite are other desirable qualities for these applications; however, oxidation at high temperatures presents a problem requiring oxygen protective systems for any prolonged use.

Aerospace and nuclear reactor applications of graphite demand high reliability and reproducibility of properties, physical integrity of product, and product uniformity. The manufacturing processes require significant additional quality assurance steps that result in high cost.

1.1. Aerospace

Graphite applications in the aerospace industry include rocket nozzle components, nose cones, motor cases, leading edges, control vanes, blast tubes, exit cones, thermal insulation, and any other applications where a rapid temperature rise and unusually high operating temperatures are encountered. Graphite is one of the few materials that can reasonably meet the demands encountered under these conditions. Of particular importance in this type of application are the excellent thermal properties of graphite, eg, high thermal shock resistance, high thermal stress resistance, and a strength increase with temperature increase. In addition, its excellent machinability makes it possible to maintain the required close tolerances for the machining of precision components for aerospace vehicles.

The erosion of graphite in nozzle applications is a result of both chemical and mechanical factors. Changes in temperature, pressure, or fuel-oxidizing ratio markedly affect erosion rates. Graphite properties affecting its resistance to erosion include density, porosity, and pore size distribution (see Ablative materials).

The entrance cap, throat, and exit cone sections in a typical nozzle are frequently made or lined with conventional bulk graphite, especially in small nozzles where dimensional stability is extremely important, since a small change in dimension causes a relatively large change in performance. In other designs the throat may be made of conventional graphite with entrance cap and exit cone molded of carbon or graphite fibrous materials that serve as reinforcement for high temperature plastic materials. In larger nozzles, all three sections might be made of fiber-reinforced material because of high strength, light weight, and ease of construction of the composite materials.

Nose cones and leading edge components fabricated of graphite are used on both ballistic and glider-type reentry vehicles. Ballistic vehicles are exposed to short duration, extremely severe friction heating and oxidizing conditions upon reentry to the atmosphere. Glider-type reentry vehicles are exposed to less severe conditions for longer periods. Design technology and the ability to control properties of manufactured graphite favorably has increased its use in aerospace applications.

The severe operational environment of advanced high performance missiles and space vehicles imposes exacting requirements on structural components that must withstand temperatures of approximately 2000°C in oxidizing atmospheres. Extensive studies on coatings for the protection of graphite under these conditions has been carried out under U.S. Air Force funding (1). A thin coating of iridium metal (2) was found to be the only material that was totally satisfactory for the protection of graphite from oxidation at 2000°C.

1.2. Nuclear Applications

The strength of graphite at high temperatures and its behavior with respect to products of nuclear fission/fusion make it a suitable material for nuclear moderators and reflectors, materials of construction, and thermal columns in various reactors. Since its use in the first nuclear reactor, CP-1, constructed in 1942 at Stagg Field, University of Chicago, many thousands of metric tons of graphite have been used for this purpose. Uranium–graphite moderators were used in the Calder Hall reactors for power generation. The advanced gas-cooled reactors (AGR), the high temperature gas-cooled reactors (HTGR), the molten salt breeder reactors (MSBR), and liquid metal fueled reactors (LMFR) all use graphite moderators. The thermal stability, resistance to corrosion, and high thermal conductivity of graphite make it a most suitable moderator material for consideration in advanced design, high temperature, atomic energy efficient nuclear reactors.

Graphite is chosen for use in nuclear reactors because it is the most readily available material with good moderating properties and a low neutron capture cross section. Other features that make its use widespread are its low cost, stability at elevated temperatures in atmospheres free of oxygen and water vapor, good heat transfer characteristics, good mechanical and structural properties, and excellent machinability.

Neutron economy in graphite occurs because pure graphite has a neutron capture cross section of only $0.0032 \pm 0.002 \times 10^{-24}$ cm^2. Taking into account the density of reactor grade graphite (bulk density 1.71 g/cm^3), the bulk neutron absorption coefficient is 0.0003/cm. Thus a slow neutron may travel >32 m in graphite without capture.

The purity of reactor-grade graphite is controlled by raw material selection and subsequent processing and purification. High temperature purification is used for most applications; however, some moderator graphites applications require considerably higher purity levels such as halogen purification to remove extremely stable carbides, especially that of boron. The actual purity requirements are determined by the reactor design.

On exposure to high temperature radiation (3) over a long period of time, graphite undergoes dimensional changes. For example, graphite initially contracts on exposure to fast neutron doses, but the rate of contraction decreases with exposure until it reaches a minimum volume; further exposure causes volume expansion, with the rate of expansion increasing rapidly at neutron doses above 3×10^{22} neutrons/cm^2(>50 keV) in all bulk graphite tested to date. This behavior is caused by atomic displacements that take place when graphite is exposed to fast neutrons, resulting in anisotropic crystallite growth rates. The crystal expands in the c-axis direction and contracts in the α-axis directions. The bulk dimensional change depends on the geometrical summation of the individual crystallite changes and, hence, is dependent on the starting materials and the method of fabrication. The extent of radiation damage is also strongly dependent on the temperature of the graphite during irradiation. The severity of graphite radiation damage at high temperatures was underestimated since the magnitude of this temperature dependence was not recognized until about 1965.

Figure 1 shows the volume change in a conventional nuclear graphite during irradiations at various temperatures of relatively high fluxes. Figure 2 shows the length change in an isotropic nuclear graphite during irradiations at various temperatures at relatively high fluxes. The actual changes in dimensions are, of course, different from grade to grade and depend largely on the degree of anisotropy present in the graphite (3).

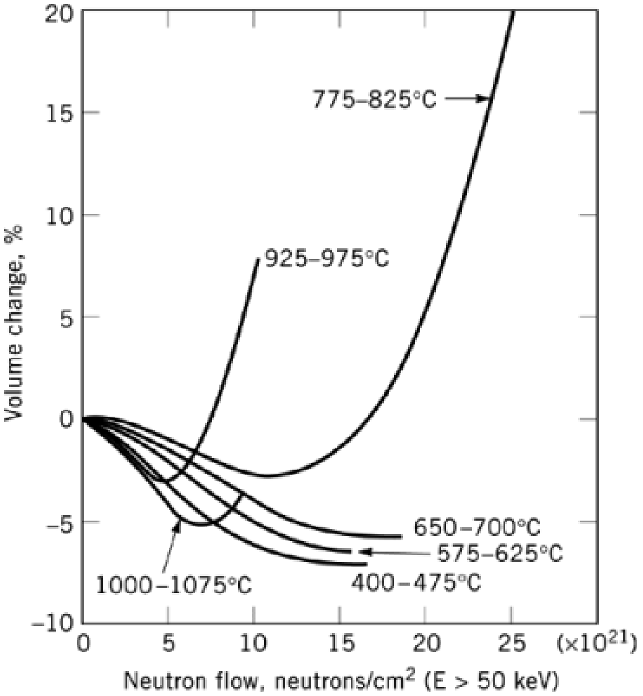

Fig. 1. Volume change in anisotropic graphite during General Electric Test Reactor (GETR) irradiations. Courtesy of Oak Ridge National Laboratory, managed by Martin Marietta Energy Systems, Inc. for the U.S. Department of Energy under Contract No. DE-AC05-840R21400.

Table 1. Properties of Nuclear Graphites

Property	Anisotropic graphite[a]	Isotropic graphite[b]
density, g/cm^3	1.71	1.86
resistance, $\mu\Omega$·cm	735	1000
tensile strength, kPa[c]	9,930	46,172
coefficient of thermal expansion (CTE), 10^{-6}/°C		
with-grain	2.2	5.3
against-grain	3.8	5.3
anisotropy ratio (CTE ratio)	1.73	1.0
total ash, ppm	740	400
boron content, ppm	0.4	0.3

[a] Ref. 4.
[b] Ref. 5.
[c] To convert kPa to psi, multiply by 0.145.

Table 1 (4, 5) lists some useful properties of several graphites used for moderators or reflectors in nuclear reactors. Reactor designers have taken advantage of graphite's properties in applying the material to other than moderator and reflector components, usually in conjunction with some other material.

Combined as an admixture with some forms of boron or other high neutron absorbing elements, graphite offers advantages as a neutron shield, control rod, or secondary shutdown material of high temperature

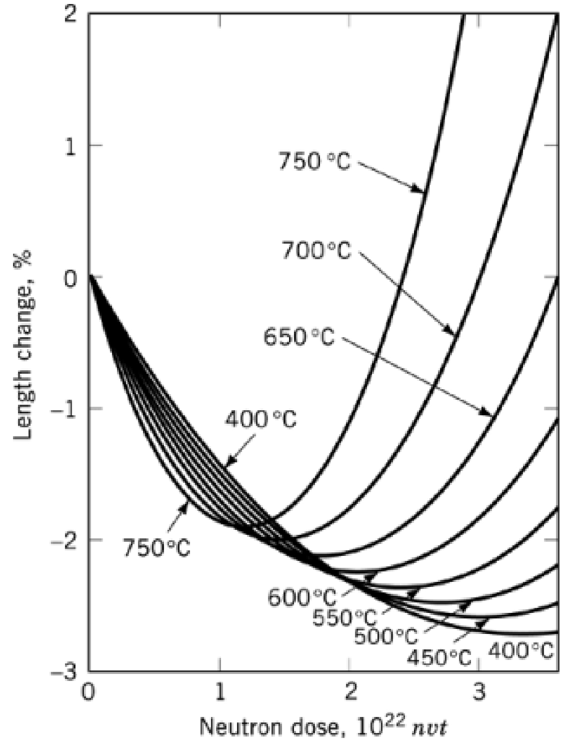

Fig. 2. Radiation-induced dimensional changes in isotropic graphite at various temperatures. $nvt = \text{neutron(density)} \cdot \text{velocity} \cdot \text{time}$ .Courtesy of Oak Ridge National Laboratory, managed by Martin Marietta Energy Systems, Inc. For the U.S. Department of Energy under Contract No. DE-AC05-840R21400.

stability, without danger of meltdown. In fast reactors, where high energy neutrons reach the shield region, the presence of carbon atoms slows these neutrons down to energies where the probability of capture in the neutron absorber greatly increases. Graphite also serves as a stable matrix for the neutron absorber because it is able to withstand neutron and localized alpha recoil damage, offering protection against gross shield degradation.

Bulk graphites are also used in the HTGR concept to support and surround the active fuel core. These components tend to be large, complex-shaped blocks and have been produced from commercial grades of molded or extruded graphites.

In combination with compounds of uranium or thorium, graphite offers advantages as a matrix for fissile or fertile reactor fuel in thermal reactors. In this instance, the graphite serves a dual purpose, as a moderator and as a stable disbursing phase for fuel. Its stability under irradiation and at high temperature aids in minimizing fuel degradation and permits longer useful fuel life. Because of its excellent thermal properties and mechanical integrity, graphite offers an excellent heat-transfer medium for heat removal and also resists thermal shock.

2. Chemical Applications

Carbon and graphite exhibit excellent resistance to the corrosive actions of acids, alkalies, and organic and inorganic compounds, an attribute that has fostered the use of graphite in process equipment where corrosion is a problem. Other than in the chemical process industries, graphite is used extensively in the steel, food, petroleum, pharmaceutical, and metal finishing industries. The high thermal conductivity and thermal stability of graphite have made it a useful material in heat exchangers and high temperature gas-spray coolers.

Manufactured carbon and graphite exhibit varying degrees of porosity depending on its method of preparation. Equipment fabricated from these materials must be operated essentially at atmospheric pressure; otherwise, some degree of leakage must be tolerated. Carbon used as a liner for tanks and vessels for the handling of highly corrosive inorganic acids such as hydrofluoric, nitric, phosphoric, sulfuric, and hydrochloric (6) is backed up by an impervious membrane of lead (7) or plastic to prevent seepage through the lining. The carbon lining protects the impermeable membrane material from adverse temperature and abrasion effects. Carbon linings have provided indefinite life with a minimum of maintenance.

2.1. Self-Supporting Structures

Self-supporting structures of carbon and graphite are used in a variety of ways. Water-cooled graphite towers serve as chambers for the burning of phosphorus in air. The high thermal conductivity of graphite allows rapid heat transfer to a water film on the outside of the tower, thereby maintaining inside wall temperature below 500°C, the oxidation temperature threshold of graphite. Phosphorus combustion chambers six meters in diameter by eleven meters in height (8) have been built using cemented graphite block construction.

The resistance of graphite to thermal shock, its stability at high temperatures, and its resistance to corrosion permit its use as self-supporting vessels to contain reactions at elevated temperatures (800–1700°C), eg, self-supporting reaction vessels for the direct chlorination of metal and alkaline-earth oxides. The vulnerability of cemented joints in these applications requires close tolerance (±0.10 mm) machining, a feat easily accomplished on graphite with conventional metal machining equipment.

Carbon Raschig-ring tower packing is available in sizes of 10–77 mm diameter. Bubble-cap trays, up to 3 m diameter for hydrochloric–organic stripping towers, and packing support structures, up to 5.5 m diameter for scrubbing towers in pulp and paper mill liquor recovery processes, have been installed. Because none of these components requires complex machining or a high degree of imperviousness, carbon rather than graphite is often used in these applications because of its lower cost.

2.2. Impervious Graphite

For applications where fluids under pressure must be retained, impregnated materials are available (6). Imperviousness is attained by blocking the pores of the graphite or carbon material with thermosetting resins such as phenolics, furans, and epoxies. Because the resin pickup is relatively small (usually 12–15 wt %), the physical properties exhibited by the original graphite or carbon material are retained. However, the flexural and compressive strengths are usually doubled. Graphite is also made impervious in a vacuum impregnation process.

Because carbon is difficult to machine, very little impervious carbon equipment is made. However, impervious graphite has been accepted as a standard material of construction by the chemical process industry for the fabrication of process equipment, such as heat exchangers, pumps, valves, towers, pipe, and fittings (9, 10).

Many types of impervious graphite shell and tube, cascade, and immersion heat exchangers are in service throughout the world (11). The most common is the shell and tube design where an impervious graphite tube bundle with fixed and floating covers is employed in combination with a steel shell. Whenever parts must be joined, such as the tube to the tube sheet in a shell and tube heat exchanger, very thin resin cement joints are used. These resin cements have the same corrosion-resistant characteristics as the resins used to impregnate the graphite. Because of the high thermal conductivity of graphite, heat exchangers fabricated of impervious graphite have thermal efficiencies equal to metal heat exchangers of equivalent heat-transfer area.

Heat exchangers up to 1.8 m diameter with areas up to 1300 m^2 are commercially available with operating pressures to 690 kPa (100 psi) and temperatures up to 170°C (12–14) (see Heat exchange technology).

Impervious graphite shells and tubes are used in numerous applications for transferring thermal energy, for example, boiling, cooling, or condensing. Large units are used extensively for cooling–condensing wet sulfur dioxide gas in sulfuric acid production plants that burn sludge acid (7).

Graphite heat exchangers are also used for evaporation of phosphoric acid and rayon spin bath solution; cooling electrolytic copper cell liquor; heating pickle liquor used for descaling sheet steel; boiling, heating, cooling, and absorbing hydrochloric acid and hydrogen chloride; and in many heating and cooling applications involving chlorinated hydrocarbons and sulfuric acid.

Impervious graphite heat exchangers machined from solid blocks are also available (15, 16). The solid block construction is less susceptible to damage by mechanical shock, such as steam and water hammer, than are shell and tube exchangers. Block exchangers are limited in size and cost from 50–100% more than shell and tube units on an equivalent area basis.

Impervious graphite centrifugal pumps, pipe fittings, and valves were developed because most chemical processes require the movement of liquids. Graphite pipe and fittings in sizes ranging from 25 to 635 mm ID are used to convey corrosive fluids.

Towers, entrainment separators, thermowells, and rupture disks are fabricated of impervious graphite material. Many equipment items are available from stock. Special equipment can be custom-designed and built, and both standard and special items can be integrated to handle a complete process step. Systems for the absorption of hydrogen chloride in water to produce hydrochloric acid use impervious graphite equipment throughout. Usually, absorption is done in a falling-film absorber (17), a special design adaption of the shell and tube heat exchanger. This approach to absorption of hydrogen chloride (18) was developed and expanded in the United States and is now accepted as the standard.

Stripping hydrogen chloride (15–21) from aqueous hydrochloric acid and the subsequent production of anhydrous hydrogen chloride can be efficiently and economically achieved with a series of impervious graphite shell and tube heat exchangers that operate as falling-film reboiler, water and brine-cooled condensers, and bottoms acid cooler. In plants with available chlorine and hydrogen, the production of hydrogen chloride in any form or concentration can be achieved in a system that combines the burning of hydrogen in chlorine in a water-cooled graphite combustion chamber; absorption is carried out in an impervious graphite falling-film absorber, and a train of impervious graphite exchangers is used for stripping and drying (22).

2.3. Low Permeability Graphite

Most resin-impregnated impervious graphite materials have a maximum operating temperature limit of 170°C because of resin breakdown above this temperature. Certain special grades with a temperature limitation of 200°C are on the market (23). The chemical industry has developed high temperature processes (370°C and above) where equipment corrosion is a serious problem. Graphite equipment could solve the corrosion problem, but complete fluid containment is usually needed. To meet this need, graphite manufacturers have developed low permeability graphite materials where permeability is reduced by deposition of carbon and graphite in the pores of the base material (23). This material is not limited in its operating temperature, except in oxidizing conditions, and it is used to fabricate high temperature interchanger ejectors, fused salt cells, fused salt piping systems, and electric resistance heaters.

2.4. Porous Graphite

Several grades of low density, porous carbon and graphite are commercially available. A controlled combination of high permeability and porosity characterizes these materials. Average pore diameters for typical grades are

0.03–0.12 mm with a total porosity of 48%. Porous graphite is manufactured by graphitizing the amorphous material.

Porous carbon and graphite are used in filtration of hydrogen fluoride streams, caustic solutions, and molten sodium cyanide; in diffusion of chlorine into molten aluminum to produce aluminum chloride; and in aeration of waste sulfite liquors from pulp and paper manufacture and sewage streams.

3. Mechanical Applications

Carbon–graphite possesses lubricity, strength, dimensional stability, thermal stability, and ease of machining, a combination of properties that has led to its use in a wide variety of mechanical applications for supporting rotating or sliding loads in contact. Its principal applications are in bearings, seals, and vanes, which are in sliding contact with a partner material. Mechanical applications of ccarbon–graphite include face, ring, and circumferential seals for gases and fluids both corrosive and noncorrosive; carbon cages for roller and ball bearings, carbon sleeve bearings and bushings, carbon thrust bearings or washers, and combination sleeve/thrust bearings; packing rings for steam and water valve shafts and packing rings for compressor tail rods; and nonlubricated compressor parts such as piston rings, wear rings, segments, scuffer shoes, shaft tail-rod packing rings, pistons, and piston skirts. Miscellaneous applications include flat-plate slider parts for supporting machinery and facilitating sliding movement under load; and rotor vanes and metering device parts.

Carbon–graphite materials employed for mechanical applications are prepared by mixing selected sizes and types of carbon and graphite with binder materials such as pitches and resins. The mixtures are formed into compacts and baked to temperatures of ca 1000–3000°C. Specific raw materials and processing techniques are employed to obtain desired properties for the finished carbon–graphite materials (24).

The successful application of carbon–graphite as a sliding contact depends on the proper use of additives and impregnants in the carbon–graphite materials. Carbon–graphite, long considered to be self-lubricating, depends on the presence of adsorbed films of water vapor and/or oxygen for its low friction and low wear properties. This adsorbed boundary layer is soon lost when the operation is conducted at high altitude, high temperature, or in cold, dry air. A substitute boundary layer can be formed by incorporating additives such as metallic sulfides, oxides, and halides, and impregnants such as thermoplastic and thermosetting resins. Additives and impregnants also serve to improve oxidation resistance, provide impermeability to high pressure gases and liquids, and even permit operation under high vaccum conditions (25), a primary requirement of equipment used for exploring outer space.

Carbon–graphite materials do not gall or weld even when rubbed under excessive load and speed. Early carbon materials contained metal fillers to provide strength and high thermal conductivity, but these desirable properties can now be obtained in true carbon–graphite materials that completely eliminate the galling tendency and other disadvantages of metals.

4. Electrode Applications

With the exception of carbon use in the manufacture of aluminum, the largest use of carbon and graphite is as electrodes in electric-arc furnaces. In general, the use of graphite electrodes is restricted to open-arc furnaces of the type used in steel production; whereas, carbon electrodes are employed in submerged-arc furnaces used in phosphorus, ferroalloy, and calcium carbide.

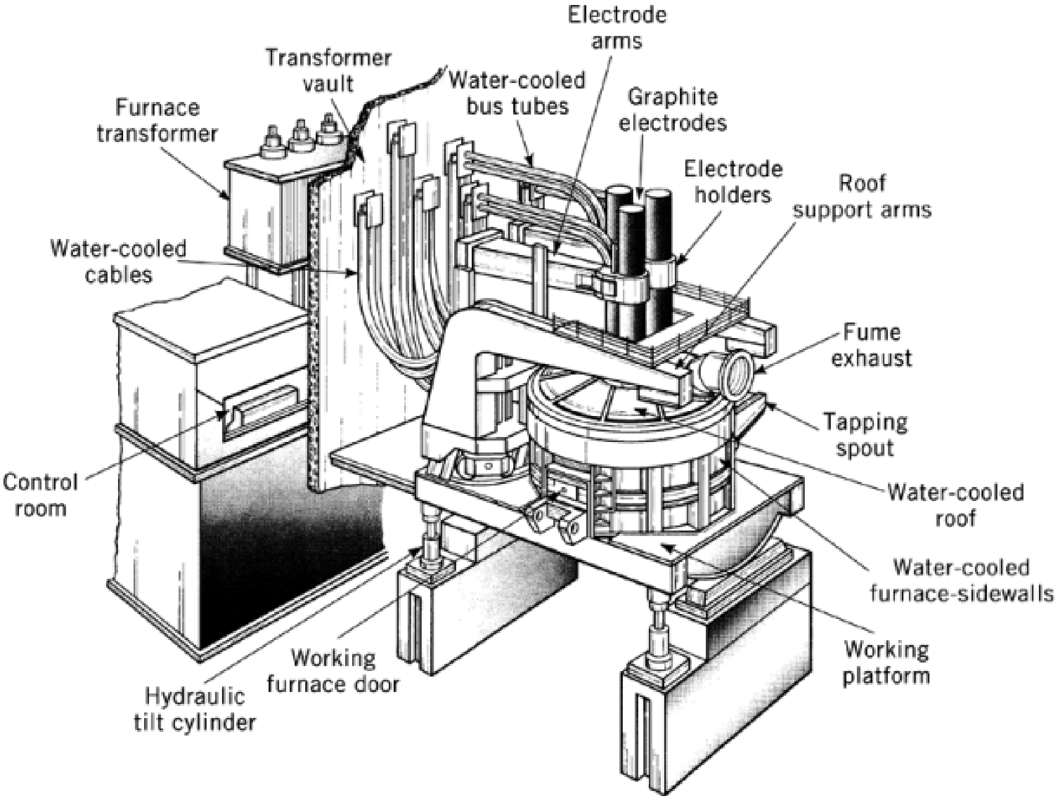

Fig. 3. Schematic of an electric-arc furnace. Courtesy of UCAR Carbon Technology Corp.

4.1. Graphite Electrodes

Graphite electrodes are commercially produced in many sizes ranging from 32 mm diameter by 610 mm length to 700 mm diameter by 2800 mm length, each diameter generally being available in two or three lengths. Such electrodes are used in open-arc furnaces for the manufacture of steel (26), iron and steel castings, brass, bronze, copper and its alloys, nickel and its alloys, magnesium, lead, tin, fused cast refractories, fused refractory grain, mineral wool insulation (27), and the treatment of toxic wastes (28). By far the largest use of graphite electrodes is in the manufacture of steel and, as a consequence, the growth of graphite production has been closely related to the growth in electric furnace steel production. Figure 3 is a schematic of a typical three-phase alternating current open-arc furnace, and shows the position of the three graphite electrode columns in this type furnace. A small but growing number of arc furnaces now use direct current and a single-electrode column positioned in the center of the furnace. In either case, steel is produced by filling the cylindrical shell with ferrous scrap, direct reduced iron, or occasionally, molten pig iron, then melting and refining the metallic charge with the intense heat derived from the electric arc generated at the tips of the electrodes.

Prior to the mid-1940s, the arc furnace was used almost exclusively for the production of low tonnage, high quality steels such as stainless and alloy steels. Since then, its use has been extended to production of the more common high tonnage steel grades, including sheet steels (see Steel). Domestic growth of arc furnace steel production has been dramatic, rising from 6% of total steel production in 1950, to 20% in 1975, and to 36% in 1990. Over 210 million metric tons of steel were produced in 1990 in electric-arc furnaces worldwide (29), approximately 26% of total world steel production, and these furnaces consumed over 800,000 tons of graphite electrodes (see Furnaces, electric-arc furnaces).

Graphite electrodes are consumed in the melting process. For iron and steel production, the average consumption is ca 2–5 kg/t, depending on the quality of the charge material, the quality of the electrodes, and numerous factors related to the productivity and operation of the arc furnace (30). A combination of these factors has resulted in a reduction of about 40% in specific electrode consumption over the past 15 years. Electrode consumption can be classified into three broad categories: tip consumption, sidewall consumption, and breakage. Roughly half of the observed consumption occurs at the electrode tip where the intensely hot and rapidly moving arc spot produces both vaporization of the graphite and some ejection of small graphite particles. In addition, the electrode tip is eroded by contact with the liquid steel and slag. For a given electrode size, the rate of incremental tip consumption increases proportionally to the power and the square of the operating current (31). As a consequence, modern high power arc furnaces operate with high voltage, low current arcs in order to minimize electrode consumption. The periphery or sidewall of the hot electrode is slowly consumed by reaction with oxidizing atmospheres both inside and outside the furnace, resulting in tapering of the electrode toward the arc tip. Sidewall consumption is time dependent and is lowest for high productivity furnaces. Certain fume removal systems and the use of oxygen in the furnace for assisting melting and refining may increase sidewall consumption. Since sidewall consumption may account for 40% or more of total electrode consumption, extensive efforts have been made to reduce this component of consumption. Oxidation retardants, electrode coatings, and water-spray rings have been moderately successful in minimizing sidewall oxidation of the portion of the electrode outside the furnace. However, the extreme thermal and chemical environment inside the furnace has prevented to date similar reduction in sidewall consumption of electrodes once inside the furnace. A third form of consumption consists primarily of electrode breakage resulting from excessive movement of large masses of scrap during melting or the presence of nonconductors in the charge. Although such breakage generally accounts for less than 5% of net electrode consumption, excessive thermal shock, improper joining practices, excessive electrode column vibration, incorrect phase rotation, and inappropriate power programs can magnify this form of electrode consumption (30). Although attempts have been made to correlate electrode consumption with relatively small changes in electrode properties, the significant reductions in electrode consumption achieved make it clear that charge quality, furnace productivity, and furnace operating practices exert a more profound influence on electrode performance. Most notable is the established inverse relationship between furnace productivity and electrode consumption.

As the electrode is consumed from the tip, the periodic addition of electrodes to the top of the columns becomes necessary. In most domestic steel plants, electrode additions are made on top of the furnace without removing the electrode columns. In certain countries, however, the electrode columns are removed from the furnace, and electrode additions are made in an assembly station adjacent to the furnace. A typical electrode column contains two full-length electrodes plus a portion of a third electrode that is partially consumed.

Two configurations are in common use today for joining graphite electrodes. Both use tapered components with a thread pitch of either three or four threads per inch (2.54 cm). Electrodes of up to about 300 mm diameter frequently have one end machined with an external tapered male thread, whereas the other end contains an internally threaded female tapered socket. Two electrodes are joined by screwing the male end of one electrode into the female socket of the second electrode and applying the desired tightening torque. The more common system of joining electrodes is illustrated in Figure 4. In this case, an externally threaded, double-tapered connecting pin is used in conjunction with electrodes which have internally threaded tapered sockets in both ends. Prior to joint assembly, the connecting pin is screwed into the lower socket of the electrode to be added. This electrode is then raised, and the assembly screwed into the open socket of the other electrode on the furnace and torqued tight. Most electrode manufacturers now preset the connecting pin in one socket of each electrode prior to shipment in order to minimize potential electrode joining difficulties. National Electrical Manufacturers Association standards have been established for electrode sizes and for electrode socket and

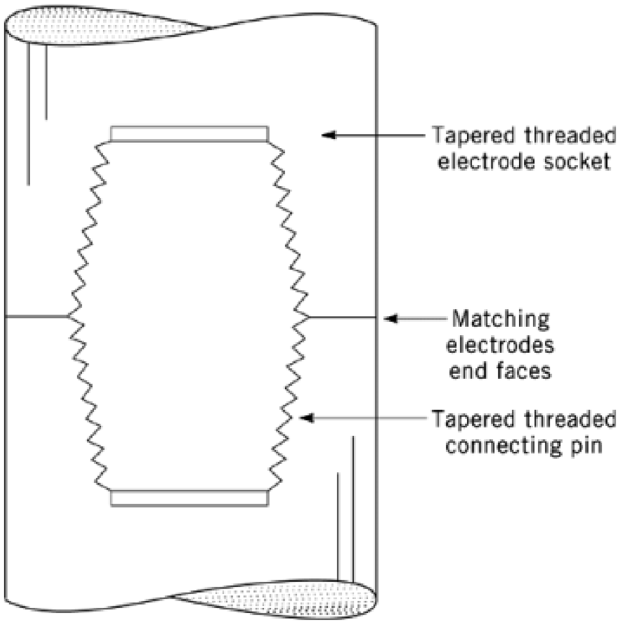

Fig. 4. Illustration of the use of a double-tapered connecting pin to join electrodes.

connecting pin configurations, sizes, and tolerances (32) in order to assure the interchangeability of electrodes and connecting pins from different manufacturers.

Graphite electrodes are produced in two broad-grade classifications, regular-grade and premium-grade. Typical room temperature properties of these grades are given in Table 2. The principal differences between the two grades are that the premium-grade is made from a superpremium needle coke and is pitch-impregnated prior to graphitization. The premium-grade electrode is used where very high performance is required, such as in the ultrahigh powered arc furnaces. The current-carrying capacity of an electrode column depends on many characteristics of the furnace operation as well as the characteristics of the electrode and electrode joint. Over the years, significant progress has been achieved in improving the current-carrying capacity of electrode columns. For example, the 510 mm diameter electrode first introduced in 1938 was designed to carry 26,000 A; this same size electrode carried 45,000 A by 1961, 55,000 A by 1975, and $>60,000$ A by 1990. Such improvements stem primarily from improved raw materials and process technology advancements that are not fully reflected in changes in electrode properties.

In service, graphite electrodes operate at up to 2500 K and are subject to large thermal and mechanical stresses and extreme thermal shock. Graphite is unique in its ability to function in this extreme environment. The relatively low electrical resistance along the length of the electrode minimizes the power loss owing to resistance heating and helps keep the electrode temperature as low as possible. This characteristic is most important in ultrahigh power furnaces where the approximately 30% lower electrode resistivity of premium-grade electrodes is usually essential for successful operation. A high value of the thermal shock parameter is also important (Table 2); this parameter is improved by high strength and high thermal conductivity combined with low elastic modulus and low coefficient of thermal expansion. The ability of graphite electrodes to withstand thermal shock has been increased significantly in the past decade as a consequence of enhancement in the elastic modulus and coefficient of thermal expansion resulting from improved raw materials and advanced manufacturing technology.

Table 2. Typical Room Temperature Properties of Regular- and Premium-Grade Graphite Electrodes and Connecting Pins

Property[a]	Regular-grade electrodes	Premium-grade electrodes	Premium-grade connecting pins
bulk density, g/cm^3	1.60	1.70	1.80
resistivity, $\mu\Omega$·m	7.3	5.5	4.2
flexural strength, kPa[b]			
wg[b]	6,900	9,100	20,000
cg	5,800	7,000	12,500
elastic modulus, GPa[b]			
wg	5.3	7.6	17
cg	3.5	5.0	6.9
coefficient of thermal expansion (CTE), 10^{-6}/°C			
wg	0.60	0.40	0.30
cg	1.40	1.10	2.20
thermal conductivity, W/(m·K)[c]			
wg	134	168	180
cg	67	101	110
thermal shock parameter[d]			
wg	290×10^3	490×10^3	700×10^3
cg	80×10^3	130×10^3	90×10^3

[a] wg = with − grain; cg = cross − grain.

[b] To convert Pa to psi, multiply by 1.45×10^{-4}.

[c] Measured.

[d] Thermal shock parameter = $\frac{\text{thermal conductivity} \times \text{strength}}{\text{CTE} \times \text{elastic modulus}}$

The joints between electrodes are an extremely important part of the electrode system, both from the standpoint of resisting the mechanical forces of scrap caves and of carrying high electrical current density without localized overheating. To assure high mechanical and electrical performance of joints, electrode manufacturers carefully control the axial and radial thermal expansion characteristics of both the electrodes and the connecting pins, as well as the machining tolerances, in order to achieve the desired level of thermal tightening of the joint as the electrode column is heated in service. Such joints should possess high strength, especially in flexure, and possess very low electrical contact resistance. Careful assembly and proper torque are vital to good performance of electrode joints (30).

4.2. Carbon Electrodes

Carbon electrodes are rigid carbonaceous shapes deployed in electric furnaces. They are the final link in the chain of conductors from the energy source to the reaction zone of an electrically heated vessel. The gap bridged by the electrode is that between the contact plates that transmit current to the electrode and the discharge area at the arc end of the electrode.

Two types of carbon electrodes are in widespread use. Prebaked carbon electrodes (Fig. 5) are those made from a mixture of carbonaceous particles and a coal-tar pitch binder. The electrode is formed by extrusion or molding from a heated plasticlike mix and subsequently baked. Final bake temperature is sufficient to carbonize the binder, ie, about 850°C. At this temperature the binder is set, all volatiles have left, and a significant portion of the product shrinkage has occurred.

Self-baked carbon electrodes are those whose shapes are formed *in situ* (33). The carbonaceous mixture is placed into a hollow tube-shaped metal casing. The upper end receives the unbaked mixture as a solid block, small particles, or warm plastic paste. The casing contains inwardly-projecting longitudinal perforated fins

Fig. 5. Prebaked electrodes. Courtesy of UCAR Carbon Technology Corp.

that become surrounded by baked carbon as the casing is incrementally moved downward and through the contact plates. Casing and carbon are consumed in this furnace.

Several systems are under development that will reduce or negate casing movement with the carbon (34–36). These will be more acceptable to furnace processes where product contamination by the casing material is undesirable.

Two types of furnaces use carbon electrodes. In the open-arc the raw materials fed into the furnace do not contact the electrode. An arc is maintained between the electrode and the charged material or the liquid bath that results from melting the charge. A furnace producing molten steel from a charge of solid metallic scrap is an example of an open-arc. The development of graphite electrodes has practically eliminated carbon electrodes from most open-arc units. The graphite can conduct much more current per unit of cross-sectional area and use of carbon in modern high powered open-arc systems is not a practical choice. Carbon is still the electrode of choice for certain open-arc furnaces such as those that produce refractories and those utilized in slag cleaning facilities.

Submerged-arc furnaces are the other type of equipment using carbon electrodes. In these, the charge materials are in contact with the electrodes. Some of the energy imparted from the electrode may be in the form of an arc to the charge or to the bath. It also may flow between electrodes through a conductive charge. Submerged-arc furnaces are quiet because the sound generated by the arc is attenuated by the covering burden of charge material.

A variety of products are made in submerged-arc furnaces. Among them are various alloys and compounds. Each uses a particular type or grade of carbon electrode to hold production costs at the lowest possible level. Graphite electrodes could be and are used in some submerged-arc furnaces. Such a choice is the result of special conditions that warrant use of the more expensive graphite in lieu of carbon.

Ferroalloy and carbide producers are the largest volume users of carbon electrodes. These are the self-baking type and ferrosilicon is the dominant product. Prebaked cathodes are used to produce silicon where a low iron content in the metal is required. A typical open top submerged-arc furnace is shown in Figure 6. Two silicon grades dominate the market; chemical-grade for producing silicones, and metallurgical-grade for alloying with aluminum. Another important use of prebakes is in the production of thermal phosphorus to be used for the manufacture of high purity phosphoric acid. These furnaces are sealed. A typical phosphorus furnace is shown in Figure 7.

Several grades of carbon electrodes are available. The characteristics of each result from the raw materials and processes used in manufacturing. The generic descriptions and primary constituents are as follows:

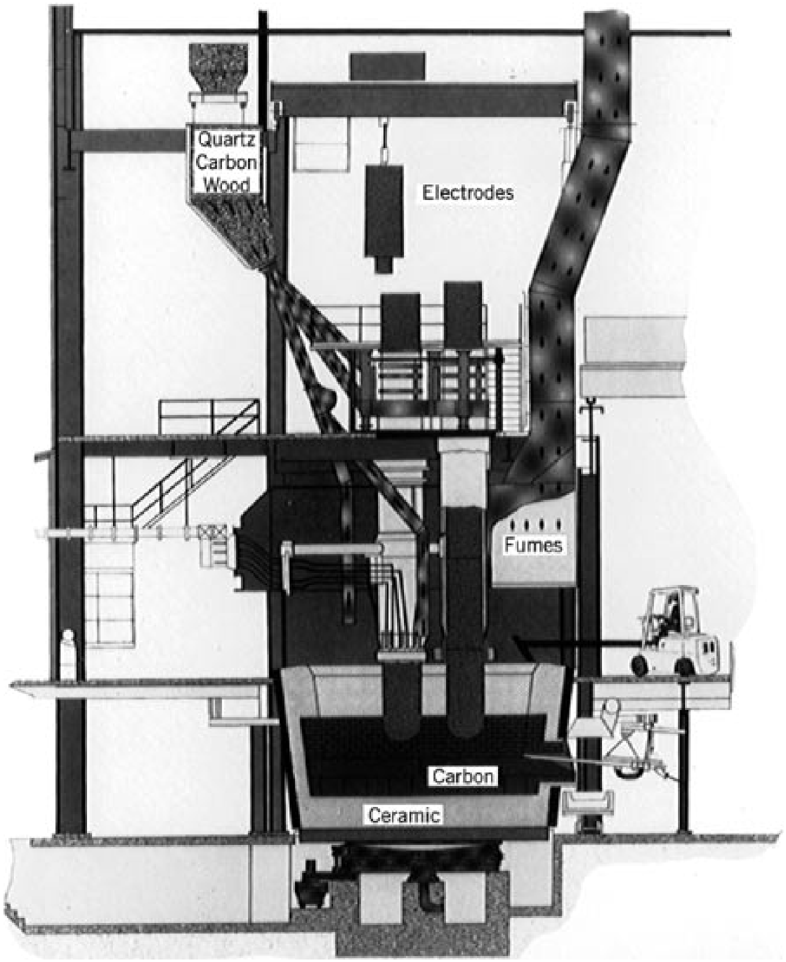

Fig. 6. Typical silicon metal furnace. Courtesy of UCAR Carbon Technology Corp.

Coal electrodes	The primary constituent is calcined anthracite coal particles.
Coke electrodes	The primary constituent is calcined petroleum coke particles.
Semigraphite electrodes	The primary constituent is sized graphite particles.
High purity electrodes	The final product has a low ash and/or low iron content achieved by raw material selection or special processing.

All carbon electrodes are amorphous. They are formed from a mixture of particles, fillers, and a binder, and they are baked to about 850°C. This is not high enough to cause the development of a crystalline structure as would occur if the temperature was elevated to that needed for graphitization.

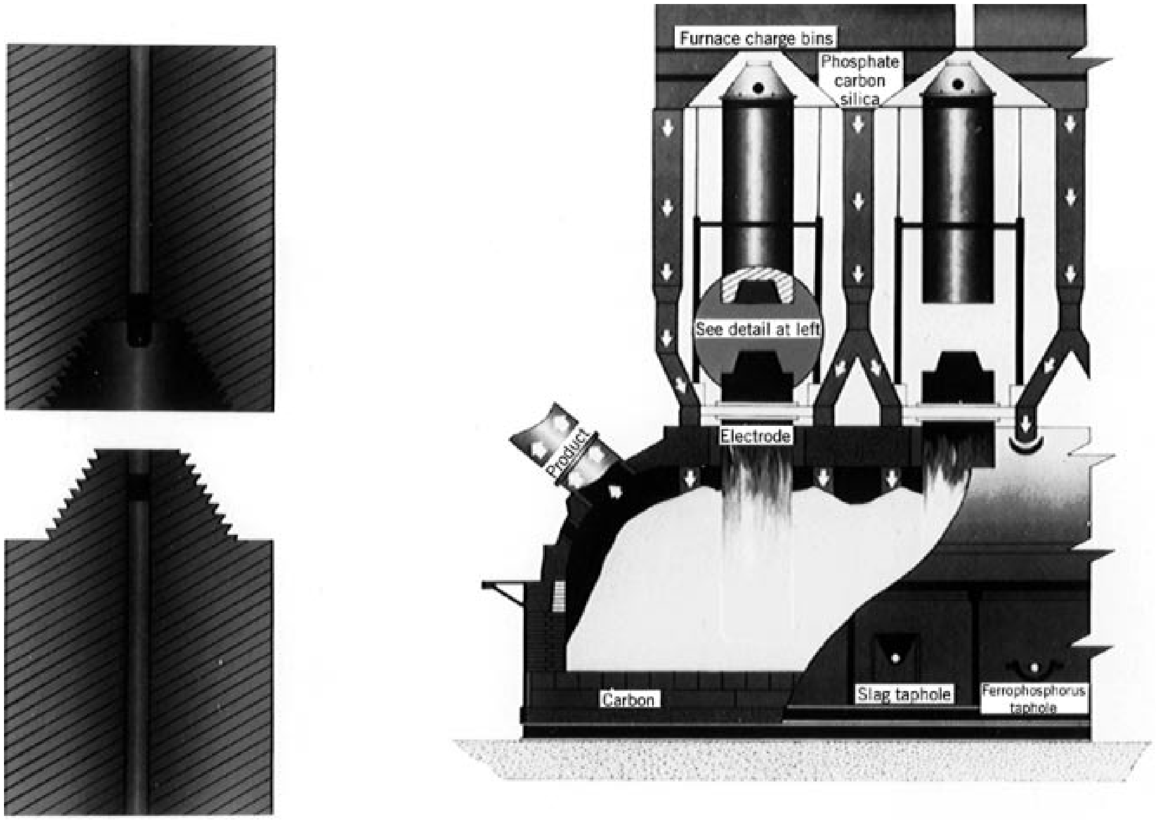

Fig. 7. Typical phosphorus furnace. Courtesy of UCAR Carbon Technology Corp.

Prebaked carbon electrodes are manufactured in all diameters up through 1500 mm. Some prebakes are produced as quadriforms to suit specific furnaces. Self-baking electrodes are in service through 2134 mm diameter. Electrode lengths are as needed for particular applications. Rounds are available in lengths up to 2794 mm and quadriforms as long as 3556 mm. Self-baked electrodes are continuous.

Production of carbon electrodes is a capital-intensive business. Two suppliers dominate the prebaked market. Carbon paste producers are more numerous and tend to serve local markets. There is no international standard for the threaded joints on carbon electrodes. Manufacturers of straight pin carbon electrodes have followed the physical specifications adopted for graphite electrodes (37). Unified standards do not exist for pinless joints resulting in limited interchangeability among brands. Electrode diameters are offered in both English and metric sizes with no restrictions on new or unique diameters.

The physical properties of carbon electrodes are determined by the raw materials and processes used in their manufacturer. There are no universal grade designations and the published properties are quite broad. Table 3 shows ranges for some of the common commercially available grades.

The density, flexural strength, and electrical conductivity increase with greater amounts of graphite particles in the mix. Flexural strength and Young's modulus move together. Ash increases in proportion to the amount of coal in the mix. Higher graphite content increases the electrode cost. Users select that electrode which gives the lowest bottom line cost per unit of production. The stated properties cannot be equated to performance. The user needs to resolve the overall performance in actual production to determine the best grade for the specific application.

Table 3. Ranges of Physical Properties of Typical Carbon Electrode Grades

Property	Range
apparent density, g/mL	1.50–1.68
specific resistance, $\mu\Omega\cdot$m	21–50
flexural strength, MPa[a]	3.1–6.9
Young's modulus, GPa	4.1–8.8
ash content, wt %	0.6–8.0

[a] To convert MPa to psi, multiply by 145.

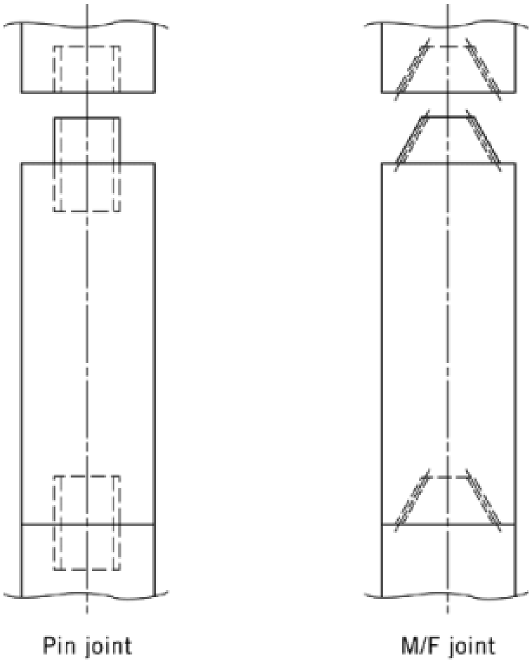

Fig. 8. Prebaked electrode joints. Courtesy of UCAR Carbon Technology Corp.

All electrodes pass through the contact pads of the electrode holder and terminate some distance above the furnace hearth. As it is consumed, the electrode is slipped downward. Self-baked electrodes are continuous and the slipping rate can be only as rapid as baking allows. Prebaked rounds are connected axially one to another with threaded features (Fig. 8). A carbonaceous connecting pin, usually made of graphite, is threaded into sockets in the ends of two axially aligned electrodes. Forming a male (external) thread on one electrode permits its connection into a threaded female (internal) socket on the other electrode. Quadriform electrodes are firmly connected to the source of power and are suspended, keeping the electrode vertical in the furnace. When the usable length of the quadriform is consumed, the remainder of the electrode is removed and replaced with full-length stock. One form of the noncircular electrode is called a packet. A few packet furnaces are still in operation.

There is no limit to the possible configurations of electric furnaces. They exist as single-phase or polyphase, a-c or d-c, and one to six or more electrodes. The furnace cross sections can be round, heart-shaped, oval, rectangular, or variations of each. Some furnaces, especially those for silicon production, have a rotating hearth and shell.

Carbon electrodes are the normal choice for the link in the connection chain to deliver power to the arc tip. Graphite may be used in special applications, but the higher cost of graphite favors the use of carbon electrodes. Carbon possesses properties ideal to its application as an electrode. These properties include no softening point, no melting point, electrical conductivity, strength increases with increasing temperature, resistivity drops as temperature increases, available in the size and purity desired, and cost effectiveness.

Pin-type electrode connections are engineered to perform across a wide temperature range. Thermal tightening is a feature that causes increasing unit contact pressure on the end faces and the contacting thread flanks. Axial CTE of the carbon is greater than the axial CTE of the pin. Transverse CTEs are controlled to either add to or not detract from the axial tightening effect. The magnitude of the thermal tightening is often such that permanent tensile elongation occurs in the pin and permanent compressive deformation occurs at the end faces. As a result, the joint will open when it is cooled sufficiently, because the elastic limits of the pin and electrodes had been exceeded.

The mechanisms of electrode consumption are dictated by the environment in which the electrode operates. Oxidation occurs on those surfaces exposed to oxygen when they are above the oxidation threshold temperature. Erosion removes carbon as it is abraded through its contact with the charge material or from movement of the electrode in the burden. The electrode is constantly repositioned in the axial direction as necessary to maintain a desired arc length. Part of the tip losses result from sublimation. At the tip there is particle loss resulting from the detonation effect of an alternating current arc. A portion of the electrode enters the electro-thermal reduction process. The electrode becomes a reductant if the charge mixture is carbon “short,” ie, less than stoichiometric. The loss of large or massive amounts follows column breakup from cracks and splits in the structure or from a full break across the electrode cross section.

Self-baking carbon electrode columns have longitudinal fissures. This results from melting or oxidation of the casing fins that extend inwardly from the casing. These fissures act as crack stoppers, but they also generate additional exposed area for oxidation. Joints in prebaked electrode columns that do not remain tight become open at the end faces. Socket splitting and cross cracks usually occur. If these joints survive to the arc region, the splits and cracks can propagate and cause chunking of the carbon. These rather large pieces are undesirable for several reasons. They can upset the reaction in progress at the arc tip, they are a cause of inefficient electrode utilization, and they may foul tap holes thereby impeding proper furnace operation.

The demand for self-baking carbon electrodes is keyed to the steel industry since it is the principal consumer of ferroalloys. Demand for prebaked electrodes is predicted to grow through the year 2000 at an annual rate of about 5%. The demand for chemical-grade silicon is the driving force for this growth. Production of metallurgical-grade has a positive but lower growth rate than that for chemical-grade silicon. Usage in production of elemental thermal phosphorus is predicted to be stable since improvements in the wet acid process can supply the increasing demand for high purity phosphoric acid.

Although carbon electrode production has been regarded as a mature business, the steady growth in demand and the need for improved electrodes has prompted ongoing development efforts in these areas: (*1*) cost containment through raw material substitutions and process improvements; (*2*) higher purity electrodes for those processes such as silicon production; (*3*) improvements in thermal shock resistance to enhance electrode performance; and (*4*) better joining systems for prebakes.

4.3. Anode Applications

Graphite has been used as the primary material for electrolysis of brine (aqueous) and fused-salt electrolytes, both as anode and cathode. Technological advances, however, have resulted in a dimensionally stable anode (DSA) consisting of precious metal oxides deposited on a titanium substrate that has replaced graphite as the primary anode (38–41) (see Alkali and chlorine products).

The cell is the basis of all electrolysis. The anode admits current into the electrolyte and the cathode serves as a means of exit for the electrical current. The electrical flow provides a definition for electrolysis: the flow of current from the anode through the electrolyte and out of the cell through the cathode with ensuing decomposition of the electrolyte, with products being formed at the electrodes.

Graphite properties conducive to successful electrolytic application include high electrical conductivity, high degree of insolubility and operation at low voltage, high purity, low initial cost, easily machinable, and few limitations as to size and shape.

The service life of graphite anodes largely depends on three factors: (*1*) electrochemical attack by nascent oxygen, resulting from oxidation at the surface; (*2*) chemical attack through the chemical reaction of cell products; and (*3*) mechanical loss of material as a result of (*1*) and (*2*). Although the eventual service life is limited by these three factors, it is not uncommon for graphite anodes to provide 300–700 days of service life. Anode operating life is also influenced by a number of cell operating variables, such as cell temperature, brine flow rate, brine concentration, and the current density imposed on the anode.

The two basic types of graphite anodes used are plain and impregnated. Impregnation prevents anolyte penetration of the graphite pores and resultant corrosion from within. For impregnated anodes, base graphites with initial porosities of 15 to 30% are given a vacuum-pressure impregnation usually with an oil, such as linseed, to fill or coat the accessible pores. For some low temperature electrolysis applications, resin treatments such as phenolics are used. These applications are well below the temperature limitations of these resins. Proper impregnation provides a 25 to 50% increase in anode life over unimpregnated graphite.

4.4. Chemical Production

Electrolytic production of chemicals is conducted either by solution (water) electrolysis or fused-salt electrolysis. Fluorine, chlorine, chlorate, and manganese dioxide are liberated from water solutions; magnesium and sodium are generated from molten salt solutions.

4.4.1. Fluorine

This application uses carbon plates as the anode in a fluorine salt solution. Since the ordered crystal structure of graphite results in short life, carbon is the preferred anode material (see Fluorine).

4.4.2. Chlorine

Most processors have converted from graphite to metal anodes. The two basic designs were diaphragm cells, which used graphite plates as anodes, and mercury cells in which a layer of mercury acted as the cathode with intricately machined graphite blocks as the anode (42).

4.4.3. Chlorate

Conversion to metal anodes has also taken place in this process. Sodium hydroxide, which is formed at the cathode, reacts to form the sodium chlorate product (see Alkali and chlorine products).

4.4.4. Manganese Dioxide

Graphite plates used as anodes in this process are coated with MnO_2 during electrolysis. The anodes are removed from the solution periodically and the MnO_2 is removed by mechanical methods. Graphite can also be used as the cathode material. Titanium is used as anode materials where high quality MnO_2 is desired.

4.4.5. Magnesium

This molten salt electrolysis process is the current principal method of magnesium production. The graphite anodes can be either round or rectangular in nature (see Magnesium and magnesium alloys).

4.4.6. Sodium

In this process, sodium is produced from molten salt. Individual cells are made up of a number of large round anodes (25–50 cm diameter) and steel pipe cathodes (see Sodium and sodium alloys).

4.5. Cathodic Protection

Another application for graphite anodes is for cathodic protection. All metal structures placed on or underground are subject to corrosion by galvanic action. Current flow, either localized or general, results in oxidation, ie, rusting, of steel. Graphite anodes are used for impressed current protection and a current is induced in the circuit counter to the galvanic current. Since the polarity is reversed, the steel does not corrode. The graphite is normally impregnated with linseed oil resin impregnates to enhance life and an electrical connection is made inside the anode with a copper wire that is ultimately connected to steel to be protected. Life of the anode can range from 3–30 years in cathodic protection applications (43, 44).

The low cost, light weight, and excellent electrical conductivity of graphite anodes have made this impressed current protection system valuable for cathodic protection of pipelines, storage vessels, process equipment, and also for well casings both on- and offshore.

5. Metallurgical Applications

Because of their unique combination of physical and chemical properties, manufactured carbons and graphites are widely used in several forms in high temperature processing of metals, ceramics, glass, and fused quartz. A variety of commercial grades is available with properties tailored to best meet the needs of particular applications (45). Industrial carbons and graphites are available in a broad range of shapes and sizes.

5.1. Structural Graphite Shapes

In many metallurgical and other high temperature applications, manufactured graphite is used because it neither melts nor fuses to many common metals or ceramics, exhibits increasing strength with temperature, has high thermal shock resistance, is nonwarping, has low expansion, and possesses high thermal conductivity. However, because of its tendency to oxidize at temperatures above 750 K, prolonged exposure at higher temperatures frequently necessitates use of a nonoxidizing atmosphere. In addition, prolonged contact both with liquid steel and with liquid metals that rapidly form carbides should be avoided.

Some of the more common applications for structural graphite shapes are (*1*) hot-pressing molds and dies (46) for beryllium at 1370 K and 6.9 MPa (1000 psi); diamond-impregnated drill bits and sawtooth segments at 1250 K and 13.8 MPa (2000 psi); tungsten and other refractory metals and alloys up to 2370 K and 6.9 MPa (1000 psi); and boron nitride and boron carbide up to 2060 K; (*2*) molds for metal casting steel railroad car wheels made by the controlled-pressure pouring process (47); steel slabs and billets made by the controlled-pressure pouring process (48); continuous casting of copper and its alloys, aluminum and its alloys, bearing materials; zinc and its alloys; and gray iron (49, 50); centrifugal casting of brasses, bronzes, steels, and refractory metals (51); nickel anodes; welding rods and thermite welding molds; shapes of refractory metals (Ti, Zr, Mo, Nb, and W) and carbides; and shapes of gray, ductile and malleable irons (52); (*3*) foundry accessories including: mold chill plates, core rods, and riser rods; crucible skimmer floats; plunging bells for magnesium additions to ductile iron and desulfurization of blast-furnace hot metal (53, 54); stirring rods for nonferrous metals; and railroad brake shoe inserts; (*4*) injection tubes and nozzles for purifying molten aluminum (55) and other nonferrous metals, desulfurization of blast furnace and foundry iron with calcium carbide or magnesium, and carbon raising of foundry iron with graphite powders; (*5*) aluminum extrusion components including dies, guides from die openings, run-out table boards, and cooling-rack inserts; (*6*) rolls for handling metal sheets are used in certain processes because they are self-lubricating and reduce surface marring; (*7*) immersion thermocouple protection tubes for nonferrous metals; (*8*) welding electrodes for welding, gouging, and cutting iron and steel, particularly with the aid of an air blast (56); (*9*) crucibles, either induction or resistance heated, for producing tungsten carbide, beryllium fluoride and beryllium, titanium and zirconium fluoride, semiconductor crystals germanium and silicon, and for laboratory chemical analysis equipment; (*10*) ceramic and glass production including: inserts for glass bottle takeout holders, casting molds for fused-cast refractories of alumina, magnesium, and chrome–magnesite composition up to 2650 K (57); mold susceptors for fabricating fused magnesia crucibles; susceptors, electrical resistor elements, fusion crucibles, molds and dies for the production of fused quartz (58); linings for float-glass plate production; and linings for hydrofluoric acid tanks for glass etching; (*11*) boats, trays, and plates for sintering clutch plates, brake disks, and cemented carbides and for the manufacture of semiconductor material and transistors; (*12*) furnace jigs for brazing honeycomb panels, automotive ignition points and arms, automotive radiator cores, transistor junction assemblies, and glass-to-metal seals; and (*13*) tooling for forming high temperature composite resin system aircraft parts (59).

5.2. Electric Heating Elements

Machined graphite shapes are widely used as susceptors and resistor elements to produce temperatures up to 3300 K in applications utilizing nonoxidizing atmospheres. The advantages of graphite in this type of application include its very low vapor pressure (lower than molybdenum), high black body emissivity, high thermal shock resistance, and increasing strength at elevated temperatures with no increase in brittleness. Graphites covering a broad range of electrical resistivity are available and can be easily machined into complex shapes at lower cost than refractory metal elements. Flexible graphite cloth is also used widely as a heating element since its low thermal mass permits rapid heating and cooling cycles. Typical applications include molten-iron or steel-holding furnaces, continuous casting tundishes, liquid–steel degassing units, chemical reaction chambers, quartz-fusion apparatus, zinc-vaporization chambers, sintering furnaces, vapor deposition units (qv) (60, 61), and single-crystal silicon ingot growing furnaces (Czochralski method). In the furnaces that use vacuum or inert gas atmospheres, porous carbon or graphite, flexible carbon or graphite felts, and rigid fibrous graphite thermal insulation materials are extensively used.

5.3. Carbon and Graphite Powder and Particles

Manufactured graphite powders and particles are used extensively in metallurgical, chemical, and electrochemical applications where the uniformity of physical and chemical characteristics, high purity, and rapid solubility in certain molten metals are important factors (62). The many grades of carbon and graphite powders and particles are classified on the basis of fineness and purity. Applications for these materials include facings for foundry molds and steel ingot molds; additives to molten iron to control carbon level and chill characteristics; covering material for molten nonferrous metals and salt baths to prevent oxidation; additives to sintered materials to control carbon level and frictional characteristics; additions in oil, grease-, and oil-less bearings; oil well drill bit lubricant; and as charge-carbon and slag foaming agents in steel made in electric arc furnaces. The electrical and thermal conducting characteristics of carbon and graphite powders and particles account for their use as additives in dry cell batteries, paint, thermoset polybutadiene composites (63), ground anode backfill, concrete and corrosion-resistant sulfur concrete.

6. Refractory Applications

Various forms of carbon, semigraphite, and graphite materials have found wide application in the metals industry, particularly in connection with the production of iron, aluminum, and ferroalloys. Carbon has been used as a refractory material since 1850, though full commercial acceptance and subsequent rapid increase in use has occurred only since 1945.

6.1. Carbon as a Blast Furnace Refractory

The first commercial use of carbon as a refractory for a blast furnace lining took place in France in 1872, followed in 1892 by a carbon block hearth in a blast furnace of the Maryland Steel Co. at Sparrows Point, Maryland. After a period of abated interest the excellent results obtained with several carbon hearths in England, Germany, and the United States during the late 1930s and early 1940s, renewed enthusiasm for the material. Although initially used only for the hearth bottom of blast furnaces, carbon, semigraphite, semigraphitized carbon, and graphite refractories have been successfully applied to hearth walls, tuyere zones, boshes, and even the lower to midstack of modern, intensely cooled, high performance blast furnaces around the world (64). More than 400 individual carbon or graphite blast furnace linings have been installed in North America through mid-1990. Additionally, carbon has also been used extensively for iron trough, iron runner, and slag runner safety linings, especially when external cooling is employed to extend the life of the ceramic working linings.

Carbonaceous and graphitic materials possess important characteristics that make them ideal blast furnace refractories: (*1*) they do not soften or lose strength at high operating temperatures of approximately 1150–1200°C; (*2*) they resist attack by molten slag and iron; (*3*) their relatively high thermal conductivity, when combined with adequate cooling and proper design concepts, promotes the formation of solidified coatings of slag and iron on their hot face. These coatings prevent erosion from the molten materials and process gases, promoting long life (65); (*4*) they possess excellent resistance to thermal shock, preventing spalling and cracking which interrupts heat transfer to the cooling system and exposes more refractory surface area to chemical attack; (*5*) a positive, low coefficient of thermal expansion provides dimensional stability and tightening of joints in the multipiece linings. However, because of their relatively low threshold temperature for oxidation from steam, carbon dioxide, or air, care must be taken to limit their exposure to these elements and maintain proper cooling at all times, to minimize damage from these temperature-dependent reactions (66, 67).

The prime requirement of any carbonaceous material used in the blast furnace hearth wall or bottom is to contain liquid iron and slag safely within the crucible, throughout extended periods of continuous operation, often up to 15 years.

This requirement is most readily achieved if the lining design concepts employed and the carbonaceous or graphitic materials utilized with these concepts, combine to provide a refractory mass free from cracking caused by mechanical and thermal stress (68). Additionally, the refractory materials must exhibit thermal conductivities that are high enough to permit the formation of solidifying layers of iron and slag on their hot faces and permeabilities that are low enough to prevent the impregnation of the refractories by alkalies and other process contaminants (69). It is also helpful if the refractory materials themselves are resistant to attack from alkalies by virtue of the inclusion of various additives during their manufacture (68). Proper cooling of the materials also contributes to their longevity.

For practical reasons, the blast furnace hearth is divided into two principal zones: the bottom and the sidewalls. Each of these zones exhibits unique problems and wear mechanisms. The largest refractory mass is contained within the hearth bottom. The outside diameters of these bottoms can exceed 16 or 17 m and their depth is dependent on whether underhearth cooling is utilized. When cooling is not employed, this refractory depth usually is determined by mathematical models; these predict a stabilization isotherm location which defines the limit of dissolution of the carbon by iron. Often, this depth exceeds 3 m of carbon. However, because the stabilization isotherm location is also a function of furnace diameter, often times thermal equilibrium cannot be achieved without some form of underhearth cooling.

This cooling can be accomplished by utilizing water, oil, induced or forced draft air, or passively, with a thick layer of high thermal conductivity graphite. The use of underhearth cooling not only allows a thermal equilibrium to be achieved but also permits a shallower depth of refractory to be used, reducing lining cost. The cooling system can employ a pipe system or airtight steel plenums (70).

The main mechanisms of hearth bottom wear are high heat load, chemical attack, erosion from molten liquids, mechanical and thermal stress, and penetration because of ferrostatic and process pressure. A variety of special purpose carbons have been developed to minimize or eliminate the damage caused by these wear mechanisms.

In North America the individual blocks of carbon used in the hearth bottom have exceeded 6 m in length. In Europe and Asia these blocks are much shorter because of manufacturing capabilities. The longer bottom blocks permit the spanning of the hearth diameter with only two pieces, which prevents flotation of the carbon by the denser molten iron. This is because the bearing provided by the dead load of the hearth walls, which rest on the ends of the carbon block "beams," anchors the bottom blocks and prevents flotation. If smaller blocks are utilized with two or more joints across the bottom, special reverse taper mating surfaces or interlocking techniques are required to prevent block flotation.

Occasionally the carbon bottom blocks in the lower most course are arranged vertically in "soldier block" fashion. The theory is that this arrangement permits the greatest heat-transfer capability of the material because of its anisotropy. Because these carbon blocks are usually extruded or pressed, there can be a 10% higher thermal conductivity in the direction perpendicular to the pressing direction and this practice takes advantage of this carbon characteristic. However, the vertical blocks in this configuration are more susceptible to differential temperature-induced stress and cracking. These designs are most prevalent in Russia and other Eastern European countries utilizing their technology.

The latest design philosophy is to utilize various types of ceramic refractories as the working lining on top of the carbon bottom. This relegates the carbon material in the bottom to a cooling function instead of a crucible function, since the molten liquids are contained completely within the ceramic layers. The high conductivity of the carbon or graphite used as the cooling layer permits thermal equilibrium to be achieved while the liquid iron is still within the ceramic. One European ironmaker has dispensed with all carbon in the hearth bottom and instead utilizes a graphite-cooled ceramic bottom, promoting this concept worldwide.

The hearth wall zone of the blast furnace presents different design problems and exhibits different main mechanisms of wear. Because these linings must contain molten iron, water cooling must be maintained on the external furnace jacket, either utilizing flooded water jackets or spray showers or by the use of water-cooled, cast-iron stave coolers located within the vessel at the cold face of the refractories. Heat removal, therefore, must travel through the wall thickness to the cooling element. Thus the design concepts employed as well as the properties of the refractory utilized have a direct bearing on the success of the hearth wall.

In North America, a special, high conductivity, low permeability, "hot-pressed" carbon brick is utilized almost exclusively for hearth walls. Because of their relatively small size and special, heat setting resin cement, and because the brick is installed tightly against the cooled jacket or stave, differential thermal expansion can be accommodated without refractory cracking and effective cooling can be maintained. Additionally, the wall thickness is generally smaller than 1 m, which promotes the easy formation of a protective skull of frozen materials on its hot face. Thus hearth wall problems and breakouts because of carbon wall refractory failure are virtually nonexistent.

Elsewhere, large block carbons are utilized as wall material, generally with thicknesses in the range of 1.5–2.5 m. However, the single-thickness blocks have a tendency to crack and spall because of high mechanical and thermal stress and lack of expansion provisions. To combat this problem, various exotic carbons have been developed to resist hot metal penetration and increase thermal conductivities, but it should be noted that these measures do not solve the cause of the cracking, which is a lack of provisions to accommodate differential expansion.

Another problem inherent to large block designs is the fact that ramming materials must be utilized to fill the annulus that forms between the circular shell or staves and the large cross-section carbon block. Usually this annulus is 75–150 mm thick. The ramming materials used to fill this annulus always possess thermal conductivities that are severely affected by shrinkage of the ram, improper rammed density, deterioration over time because of contamination from leaking cooling water or the process environment. As a result, this rammed gap acts as a thermal barrier to effective heat transfer to the cooling system. The result is a higher carbon wall temperature with consequent accelerated wall wear.

This wear is caused primarily from high thermal and mechanical stress, chemical attack, attack by iron and slag, oxidation, and severe thermal shock. Thus the design of the hearth wall and the concepts employed are just as important as the carbon or graphite materials chosen for the refractory material. Despite their benefits and properties, no carbon or graphite material can overcome the problems of an improper hearth wall design concept.

In the early 1960s, the use of carbon and more recently, semigraphite and graphite as bosh refractories, has found increased acceptance and resulted in longer furnace campaign life. As is the case in the hearth wall, long life is dependent on efficient cooling of the carbonaceous or graphitic material. Originally, all carbon materials were cooled on their cold face, either by external shell spray cooling or external water jackets. Later, cast-iron, water-cooled staves were successfully used to cool the cold face of the bosh wall. However, all of these cooling systems required that the heat travel completely through the wall to reach the cooling element. These cooling methods were employed as a result of the high risk of water leaks because of poor quality coolers. These leaks could prove disastrous because the leaking water and consequential steam would badly oxidize the surrounding carbon. However, dramatic improvements in copper cooling plate technology and casting practice and sophisticated water leak detection systems have combined to provide low risk, high efficiency, inserted copper cooler systems for carbonaceous or graphitic linings. This results in lower refractory temperatures and, thus, lower chemical attack from alkalies, which are temperature-dependent reactions with carbon (71).

These advances in cooling technology have also provided an opportunity to extend the thermal shock and chemical resistance of graphitic materials above the bosh, into the blast furnace stack. It has been recognized by many blast furnace operators around the world that the most destructive wear mechanism in the upper bosh and lower to midstack level is thermal shock. It is also universally recognized that the only refractory material that can withstand the magnitude of the temperature peaks normally occurring in these blast furnace zones are graphitic in nature. Therefore, the combination of densely spaced, modern copper coolers with high quality semigraphite or graphite refractories has proven to be a successful extender of blast furnace campaign life (72). Several furnaces have recently been relined utilizing these concepts in Holland, Canada, Italy, and the United States; several others are planned in England and Central America. As a result, the percentage of carbonaceous and graphitic materials utilized in the blast furnace is increasing tremendously and this trend should continue into the late 1990s.

6.2. Refractories for Cupolas

In many ways, the use of carbon cupola linings has paralleled the application of carbon in the blast furnace. Carbon brick and block are used to line the cupola well (73) or crucible. When properly installed and cooled carbon linings last for many months or even years of intermittent operation. Their resistance to molten iron and both acid and basic slags provides not only insurance against breakouts but also operational flexibility to produce different iron grades without the necessity of changing refractories. Carbon is also widely used for the tap hole blocks, breast blocks, slagging troughs, and dams.

6.3. Refractories for Electric Reduction Furnaces

Carbon hearth linings are used in submerged-arc, electric-reduction furnaces producing phosphorus, calcium carbide, all grades of ferrosilicon, high carbon ferrochromium, ferrovanadium, and ferromolybdenum. Carbon is also used in the production of beryllium oxide and beryllium copper where temperatures up to 2273 K are required.

The principles pertaining to carbon blast furnace hearths apply as well to submerged-arc furnace hearths. In some processes, such as in d-c arc furnaces, the electrical conductance of carbon is a most important factor. The long life of carbon linings in these applications is attributable to carbon's exceptional resistance to corrosive slags and metals at very high temperatures.

6.4. Refractories in the Aluminum Industry

Carbon materials are used in the Hall-Heroult primary aluminum cell as anodes, cathodes, and sidewalls because of the need to withstand the corrosive action of the molten fluorides used in the process (see Aluminum and aluminum alloys). Production of one metric ton of molten aluminum requires about 500 kg of anode carbon and 7.5–10 kg of cathode blocks which is the largest industry usage of carbon materials. Aluminum smelters generally have an on-site carbon plant for anode production. Anode technology is focused on raw materials (petroleum coke and coal-tar pitch), processing techniques, and rodding practices (74).

Prebaked cathode blocks used today are electrically calcined anthracite coal, semigraphite, semigraphitized, or graphite composition (75). Desired cathode operating characteristics include resistance to sodium attack, high operating strength, low porosity, high thermal shock resistance, and low electrical resistance. The use of graphitic prebaked cathode blocks is a balance among performance, power, life, and economics within the cell; coal-based carbon is still the predominant cathode block composition. Cathode technology developments include coatings of titanium diboride (76) for voltage reduction and cemented collector bar assembly practices (77).

Sidewall blocks are monolithic rammed carbon or prebaked carbon and graphite blocks. Thermal conductivity to maintain a ledge against the molten metal is the most significant sidewall operating parameter. Some pots utilize both carbon and graphite sidewall blocks to maximize performance in specific sidewall areas. Thermal shock resistance and strength are also desired sidewall properties. Low temperature resin cast carbon, ie, cold-cast materials, can also be used for corner block locations. Carbonaceous cements and pastes are used for joint integrity and thermal balance.

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