1 History and Bibliography of Diffusion

If a droplet of ink is placed without stirring at the bottom of a bottle filled with water, the colour will slowly spread through the bottle. At first, it will be concentrated near the bottom. After a few days, it will penetrate upwards a few centimeters. After several days, the solution will be coloured homogeneously. The process responsible for the movement of the coloured material is diffusion. Diffusion is caused by the BROWNian motion of atoms or molecules that leads to complete mixing. In gases, diffusion progresses at a rate of centimeters per second; in liquids, its rate is typically fractions of millimeters per second; in solids, diffusion is a fairly slow process and the rate of diffusion decreases strongly with decreasing temperature: near the melting temperature of a metal a typical rate is about one micrometer per second; near half of the melting temperature it is only of the order of nanometers per second.

The science of diffusion in solids had its beginnings in the 19th century, although the blacksmiths and metal artisans of antiquity already used the phenomenon to make such objects as swords of steel, gilded copper or bronce wares. Diffusion science is based on several corner stones. The most important ones are: (i) The continuum theory of diffusion originated from work of the German scientist *Adolf Fick*, who was inspired by elegant experiments on diffusion in gases and of salt in water performed by *Thomas Graham* in Scotland. (ii) The BROWNian motion was detected by the Scotish botanist *Robert Brown*. He observed small particles suspended in water migrating in an erratic fashion. This phenomenon was interpreted many decades later by *Albert Einstein*. He realised that the 'dance' described by *Brown* was a random walk driven by the collisions between particles and the water molecules. His theory provided the statistical cornerstone of diffusion and bridged the gap between mechanics and thermodynamics. It was verified in beautiful experiments by the French Nobel laureate *Jean Baptiste Perrin*. (iii) The atomistics of solid-state diffusion had to wait for the birth date of solid-state physics heralded by the experiments of *Max von Laue*. Equally important was the perception of the Russian and German scientists *Jakov Frenkel* and *Walter Schottky* that point defects play an important rôle for properties of crystalline substances, most notably for those controlling diffusion and the many properties that stem from it.

This chapter is not meant to be a systematic history of diffusion science. It is devoted in its first section to some major landmarks and eminent people in the field. The second section contains information about bibliography of diffusion in textbooks, monographs, conference proceedings, and data collections.

1.1 Pioneers and Landmarks of Diffusion

Establishment of the diffusion law: Experimental studies of diffusion were probably performed for the first time by *Thomas Graham* (1805–1869). Graham was born in Glasgow. His father was a successful textile manufacturer. He wished his son to enter the Church of Scotland. Defying his father's wishes he studied natural sciences, developed a strong interest in chemistry and became professor of chemistry in 1830 at the Andersonian Institute (now Strathclyde University) in Glasgow. Later he became professor of chemistry at several colleges including the Royal College of Science and Technology and the University of London in 1837. Graham helped to found the Chemical Society of London and became its first president. In 1854 Graham succeeded Sir John Herschel as Master of the Mint in London following the tradition – established by Sir Isaac Newton – of distinguished scientists occupying the post.

Graham is one of the founders of physical chemistry and he discovered the medical method of 'dialysis'. He initiated the quantitative study of diffusion in gases, largely conducted in the years of 1828 to 1833 [1, 2]. In one of his articles he explicitly stated what we now call Graham's law: *'The diffusion or spontaneous intermixture of two gases is effected by an interchange in position of indefinitely minute volumes of the gases, which volumes are not of equal magnitude, being, in the case of each gas, inversely proportional to the square root of the density of that gas.'* The crucial point about Graham's work on diffusion in gases was that it could be understood by the kinetic theory of gases developed by Maxwell and Clausius shortly after the middle of the 19th century. Graham's law can be attributed to the equipartition of kinetic energies between molecules with different molecular masses. In this way diffusion was connected with the thermal motion of atoms or molecules, and the idea of the mean free path entered science. Graham also extended his studies to diffusion of salts in liquids [3] and to the uptake of hydrogen in metals. He showed that diffusion in liquids was at least several thousand times slower than in gases.

The next major advance in the field of diffusion came from the work done by *Adolf Eugen Fick* (1829–1901). He was born in Kassel, Germany, as the youngest of five siblings. His father, a civil engineer, was a superintendent of buildings. During his secondary schooling, Adolf Fick was delighted by mathematics, especially by the work of Fourier and Poisson. He entered the University of Marburg with the intention to specialise in mathematics, but switched to medicine on the advice of an elder brother, a professor of anatomy. He got his doctorate with a thesis on *'Visual Errors due to Astigmatism'*. He spent the years from 1852 to 1668 at the University of Zürich, Switzerland, in various positions. After sixteen years in Zürich he accepted a chair in physiology in Würzburg, Germany.

Graham's work on diffusion of salts in water stimulated Fick to develop a mathematical framework to describe the phenomena of diffusion using the analogy between Fourier's law of thermal conduction and diffusion [4, 5]. Fick signed his papers on diffusion as 'Demonstrator of Anatomy, Zürich'. They were published in high-ranking journals. His approach was a phenomenological one and uses a continuum description. Nowadays, we would call his theory a 'linear response' approach. Fick is even better known in medicine. He published a well-rounded monography on *'Medical Physics'* [6] and a textbook on *'The Anatomy of Sense Organs'*. He became an outstanding person in the small group of nineteenth century physiologists who applied concepts and methods of physics to the study of living organisms, and thereby laid the foundations of modern physiology. Fick's vital contribution to the field of diffusion was to define the diffusion coefficient and to measure it for diffusion of salt in water. Mathematical solutions of Fick's equations began with the nineteenth century luminaries *Jozef Stephan* [7] and *Franz Neumann*, who were among the first to recognise the significance of boundary conditions for solutions of the diffusion equation.

Roberts-Austen – discovery of solid-state diffusion: *William Chandler Roberts-Austen* (1843–1902) graduated from the Royal School of Mines, London, in 1865 and became personal assistant to Graham at the Mint. After Graham's death in 1869 Roberts-Austen became 'Chemist and Assayer of the Mint', a position he occupied until his death. He was appointed professor of metallurgy at the Royal School of Mines in 1880 and was knighted in 1899 by Queen Victoria. He was a man of wide interests, with charm, and an understanding of people, which made him very popular. He conducted studies on the effects of impurities on the physical properties of pure metals and alloys and became a world authority on the technical aspects of minting coins. His work had many practical and industrial applications. Austenite – a non-magnetic solution of carbon in iron – is named after Sir Roberts-Austen.

He records his devotion to diffusion research as follows [8]: *'... My long connection with Graham's researches made it almost a duty to attempt to extend his work on liquid diffusion to metals.'* Roberts-Austen perfected the technique for measuring high temperatures adopting Le Chatelier's platinum-based thermocouples and studied the diffusion of gold, platinum, and rhodium in liquid lead; of gold, silver, and lead in liquid tin; and of gold in bismuth. These solvents were selected because of their relatively low melting temperatures. The solidified samples were sectioned and the diffused species determined in each section using the high precision assaying techniques developed for use in the Mint. Typically six or seven sections were taken and diffusion coefficients determined. Even more importantly, Roberts-Austen applied these techniques to the study of gold diffusion in solid lead as well. It is interesting to observe that the values of the diffusion coefficient of gold in lead reported by him are very close to those determined by modern techniques using radioactive isotopes. The choice of the system gold in lead was really fortunate. Nowadays, we know that the diffusion of noble metals in lead is exceptionally fast in comparison to most other diffusion processes in solids.

Arrhenius law of solid-state diffusion: The most surprising omission in Roberts-Austen's work is any discussion of the temperature dependence of the diffusion coefficient. Historically, the temperature dependence of reaction rates, diffusivities etc., now generally referred to as 'Arrhenius law', is named after the Swedish scientist *Svante August Arrhenius* (1859–1927). Arrhenius got a doctorship in chemistry in Uppsala, Sweden, in 1884 with a thesis about *electrolytic dissociation.* He was awarded a travel fellowship which enabled him to work with Ostwald in Riga, now Latvia, and with Kohlrausch in Würzburg, Germany. He also cooperated with Boltzmann in Graz, Austria, in 1887 and with Van't Hoff in Amsterdam, The Netherlands, in 1888. He was appointed for a chair in chemistry at the University of Stockholm in 1891. He abandoned this position in 1905 to become director of the Nobel Institute of Physical Chemistry. Arrhenius was awarded the 1903 Nobel prize in chemistry *for his theory of electrolytic dissociation.* It appears that the Arrhenius law for chemical reactions was proposed by the Dutch scientist *Jacobus Hendrik van't Hoff* (1852–1921), the first Nobel laureate in chemistry (1901). The suggestion that the diffusivity in solids should obey that law was apparently made by *Dushman and Langmuir* in 1922 [9].

Von Hevesy – the first measurements of self-diffusion: The idea of self-diffusion was already introduced by Maxwell, when treating the rate of diffusion of gases. The first attempts to measure self-diffusion in condensed matter were those of *Georg Karl von Hevesy* (1885–1966), who studied self-diffusion in liquid [10] and in solid lead [11] by using a natural radioactive isotope of lead. Von Hevesy had a fascinating scientific career. He was born in Budapest, Austria-Hungary, and studied at the Universities of Budapest, Berlin and Freiburg. He did research work in physical chemistry at the ETH in Zürich, with Fritz Haber in Karlsruhe, with Ernest Rutherford in Manchester, and with Fritz Paneth in Vienna. After World War I he teached for six months at the University of Budapest, and from 1920 to 1926 he worked with Niels Bohr at the University of Copenhagen. Together with the Dutch physicist Dirk Coster he discovered the new element 'hafnium' among the oreg of zirconium. He was professor at the University of Freiburg, Germany, from 1926 to 1934. During his eight years in Freiburg he initiated work with radiotracers in solids and in animal tissues. Fleeing from the Nazis in Germany, he moved to the Niels Bohr Institute in Copenhagen in 1934 and from there to Stockholm. In 1944 the Swedish Royal Academy of Sciences awarded him the Nobel prize in Chemistry of the year 1943 for *'. . . his work on the use of isotopes as tracers in the study of chemical processes.'* He became a Swedish citizen and was appointed professor of organic chemistry at the University of Stockholm in 1959. Von Hevesy, who married Pia Riis, daughter of a Danish ship owner, had four children, died in Freiburg, Germany. Von Hevesy is also the founder of radioistope applications in nuclear medicine. For example, the hospital of the author's University has a station called 'von Hevesy station'. Wolfgang Seith, who collaborated with von Hevesy in Freiburg, was appointed as the first professor in physical chemistry at the author's University in Münster, Germany.

Brownian motion: The phenomenon of irregular motion of small particles suspended in a liquid had been known for a long time. It had been discovered by the Scottish scientist *Robert Brown* (1773–1858). Brown was the son of an episcopalian priest. He studied medicine at Edinburgh University, but did not obtain his degree. At the age of twenty-one he enlisted in a newly raised Scottish regiment. At that time he already knew that his true interests lay not in medicine but in botany, and he already had acquired some reputation as a botanist. On a visit to London in 1798 to recruit for his regiment, he met the botanist Sir Joseph Banks, president of the Royal Society, who recommended Brown to the Admiralty for the post of a naturalist aboard a ship. The ship was to embark on a surveying voyage at the coasts of Australia. Brown made extensive plant collections in Australia and it took him about 5 years to classify approximately 3900 species he had gathered, almost all of which were new to science. By that time, Robert Brown was already a renowned botanist. Much later *Charles Darwin* referred to him as *'...princeps botanicorum'*. In addition to collecting and classifying, Brown made several important discoveries. Perhaps the most celebrated one by biologists is his discovery that plant cells have a nucleus.

Robert Brown is best known in science for his description of the random movement of small particles in liquid suspension, first described in a pamphlet entitled *'A brief account of microscopical observations in the months June, July and August 1827 on the particles contained in pollen ... '*, which was originally intended for private circulation, but was reprinted in the archival literature shortly after its appearance [12]. Brown investigated the way in which pollen acted during impregnation. A plant he studied under the microscope was *Clarkia pulchella*, a wildflower found in the Pacific Northwest of the United States. The pollen of this plant contains granules varying from about five to six micrometers in linear dimension. It is these granules, not the whole pollen grains, upon which Brown made his observation. He wrote *'... While examining the form of these particles immersed in water, I observed many of them very evidently in motion ... These motions were such as to satisfy me, after frequently repeated observation, that they arose neither from currents in the fluid, nor from its gradual evaporation, but belonged to the particle itself'*. The inherent, incessant motion of small particles is nowadays called BROWNian motion in honour of Robert Brown.

Einstein's and Smoluchowski's theory of Brownian motion: In the period between 1829 and about 1900 not much progress was made in the understanding of BROWNian motion, although developments in the theory of heat and kinetic theory stimulated new experiments and conjectures. It is striking that the founders and developers of kinetic theory, Maxwell, Boltzmann, and Clausius, never published anything on BROWNian motion. The reason for the lack in progress was that the major studies of that period focused on the particle velocities. Measurements of the particle velocities gave puzzling results. The path of a small particle, on the length scales available from observations in a microscope, is an extremely erratic curve. In modern language we would say that it is a fractal. Such curves are differentiable almost nowhere. Consequently, the particles whose trajectories they represent have no velocity, as usually defined. It was not until the work of *Einstein* and *Smoluchowski* that it was understood that the velocity is not a useful thing to measure in this context.

Albert Einstein (1879–1955), born in Ulm, Germany, is certainly the best known physicist of the twentieth century, perhaps even of all time. In the year of 1905, he published four papers that at once raised him to the rank of a physicist of the highest caliber: the photon hypothesis to explain the photo effect, for which he received the Nobel prize in physics in 1922 for the year 1921, his first paper on BROWNian motion, and his two first papers on relativity theory. At that time Einstein was employed at the *'The Eidgenössische Amt für Geistiges Eigentum'* in Bern, Switzerland. He did not receive the doctoral degree until the following year, 1906. Interestingly, his thesis was on none of the above problems, but rather concerned with the determination of the dimensions of molecules. His first paper on BROWNian motion was entitled *'Die von der molekularkinetischen Theorie der Wärme geforderte Bewegung von in ruhenden Flüssigkeiten suspendierten Teilchen'* [13]. A second paper was entitled *'Zur Theorie der Brownschen Bewegung'* [14]. Einstein published two additional short papers on this topic [15, 16], but these were of relatively minor interest. Einstein was the first to understand, contrary to many scientists of his time, that the basic quantity is not the velocity but the mean-square displacement of particles. He related the mean-square displacement to the diffusion coefficient.

The Polish physicist and mountaineer *Marian Smoluchowski* (1872–1917) was born in Vienna, Austria. During his lifetime, Poland was not an independent country; it was partitioned between Russia, Prussia, and Austria. Marian Smoluchowski entered the University of Vienna and studied physics under *Joseph Stephan* and *Franz Exner*. He was impressed by the work of Ludwig Boltzmann. In his later life he was called *'der geistige Nachfolger Boltzmanns'* (the intellectual successor of Boltzmann). He got his PhD and his 'venia legendi' from the University of Vienna and was appointed full professor at Lvov University (now Ukraine) in 1903. He accepted a chair in physics at the Jagellonian University at Cracow in 1913, when he was a wellknown physicist of worldwide recognition. Smoluchowski also served as president of the Polish Tatra Society and received the *'Silberne Edelweiss'* from the German and Austrian Alpine Society, an award given to distinguished alpinists.

Smoluchowski's interest for molecular statistics led him already around 1900 to consider BROWNian motion. He did publish his results not before 1906 [17, 18], under the impetus of Einstein's first paper. Smoluchowski later studied BROWNian motion for particles under the influence of an external force [19, 20]. Einstein's and Smoluchowski's scientific paths crossed again, when both considered the theory of the scattering of light near the critical state of a fluid, the critical opalescence. Smoluchowski died as a result of a dysentery epidemic, aggravated by wartime conditions in 1917. Einstein wrote a sympathetic obituary for him with special reference to Smoluchowski's interest in fluctuations [21].

Atomic reality – Perrin's experiments: The idea that matter was made up of atoms was already postulated by Demokrit of Abdeira, an ancient Greek philosopher, who lived about four hundred years before Christ. However, an experimental proof had to wait for more than two millennia. The concept of atoms and molecules took strong hold of the scientific community since the time of English scientist *John Dalton* (1766–1844). It was also shown that the ideas of the Italian scientist *Amadeo Avogadro* (1776–1856) could be used to construct a table of atomic weights, a central idea of chemistry and physics. Most scientists were willing to accept atoms as real, since the facts of chemistry and the kinetic theory of gases provided strong indirect evidence. Yet there were famous sceptics. Perhaps the most prominent ones were the German physical chemist and Nobel laureate *Wilhelm Ostwald* (1853–1932) and the Austrian physicist *Ernst Mach* (1938–1916). They agreed that atomic theory was a useful way of summarising experience. However, the lack of direct experimental verification led them to maintain their scepticism against atomic theory with great vigour.

The Einstein-Smoluchowski theory of Brownian motion provided ammunition for the atomists. This theory explains the incessant motion of small particles by fluctuations, which seems to violate the second law of thermodynamics. The question remained, what fluctuates? Clearly, fluctuations can be explained on the basis of atoms and/or molecules that collide with a Brownian particle and push it around. The key question was then, what is the experimental evidence that the Einstein-Smoluchowski theory is quantitatively correct? The answer had to wait for experiments of the French scientist *Jean Baptiste Perrin* (1870–1942), a convinced atomist. The experiments were difficult. In order to study the dependence of the mean-square displacement on the particle radius, it was necessary to prepare monodisperse suspensions. The experiments of Perrin were successful and showed agreement with the Einstein-Smoluchowski theory [22, 23]. He and his students continued refining the work and in 1909 Perrin published a long paper on his own and his students' research [24]. He became an energetic advocate for the reality of atoms and received the 1926 Nobel prize in physics *'...for his work on the discontinuous structure of matter ... '*.

Crystalline solids and atomic defects: Solid-state physics was born when *Max von Laue* (1879–1960) detected diffraction of X-rays on crystals. His experiments demonstrated that solid matter usually occurs in three-dimensional periodic arrangements of atoms. His discovery, published in 1912 together with *Friedrich* and *Knipping*, was awarded with the 1914 Nobel prize in physics.

However, the ideal crystal of Max von Laue is a 'dead' crystal. Solid-state diffusion and many other properties require deviations from ideality. The Russian physicist *Jakov Il'ich Frenkel* (1894–1952) was the first to introduce the concept of disorder in the field of solid-state physics. He suggested that thermal agitation causes transitions of atoms from their regular lattice sites into interstitial positions leaving behind lattice vacancies [25]. This kind of disorder is now called Frenkel disorder and consists of pairs of vacant lattice sites (vacancies) and lattice atoms on interstitial sites of the host crystal (self-interstitials). Only a few years later, *Wagner and Schottky* [26] generalised the concept of disorder and treated disorder in binary compounds considering the occurrence of vacancies, self-interstititals and antisite defects on both sublattices. Nowadays, it is common wisdom that atomic defects are necessary to mediate diffusion in crystals. The German physicist *Walter Schottky* (1886–1976) taught at the universities of Rostock and Würzburg, Germany, and worked in the research laboratories of Siemens. He had a strong influence on the development of telecommunication. Among Schottky's many achievements a major one was the development of a theory for the rectifying behaviour of metal-semiconductor contact, which revolutionised semiconductor technology. Since 1973 the German Physical Society decorates outstanding achievements of young German scientists in solid-state physics with the 'Walter-Schottky award'.

Kirkendall effect: A further cornerstone of solid-state diffusion comes from the work of *Ernest Kirkendall* (1914–2005). In the 1940s, it was still a widespread belief that atomic diffusion in metals takes place via direct exchange or ring mechanisms. This would suggest that in binary alloys the two components should have the same coefficient of self-diffusion. Kirkendall and coworkers observed the inequality of copper and zinc diffusion during interdiffusion between brass and copper, since the interface between the two different phases moves [27–29]. The direction of the mass flow was such as might be expected if zinc diffuses out of the brass more rapidly than copper diffuses in. Such phenomena have been observed in the meantime in many other binary alloys. The movement of inert markers placed at the initial interface of a diffusion couple is now called the *Kirkendall effect*. Kirkendall's discovery, which took the scientific world about ten years to be appreciated, is nowadays taken as evidence for a vacancy mechanism of diffusion in metals and alloys. Kirkendall left research in 1947 and served as secretary of the American Institute of Mining, Metallurgical and Petroleum Engineers. He then became a manager at the United Engineering Trustees and concluded his career as a vice president of the American Iron and Steel Institute.

Thermodynamics of irreversible processes: The Norwegian Nobel laureate in chemistry of 1968 *Lars Onsager* (1903–1976) had widespread interests, which include colloids, dielectrics, order-disorder transitions, hydrodynamics, thermodynamics, and statistical mechanics. His work had a great impact on the 'Thermodynamics of Irreversible Processes'. He received the Nobel prize for the reciprocity theorem, which is named after him. This theorem states that the matrix of phenomenological coefficients, which relate fluxes and generalised forces of transport theory, is symmetric. The non-diagonal terms of the Onsager matrix also include cross-phenomena, such as the influence of a gradient in concentration of one species upon the flow of another one or the effect of a temperature gradient upon the flow of various atomic species, both of which can be significant for diffusion processes.

Solid-state diffusion after World War II: The first period of solid-state diffusion under the guidance of Roberts-Austen, von Hevesy, Frenkel, and Schottky was followed by a period which started in the mid 1930s, when 'artificial' radioactive isotopes, produced in accelerators, became available. Soon after World War II nuclear reactors became additional sources of radioisotopes. This period saw first measurements of self-diffusion on elements other than lead. Examples are self-diffusion of gold [30, 31], copper [32], silver [33], zinc [34], and α-iron [35]. In all these experiments the temperature dependence of diffusion was adequately described by the Arrhenius law, which by about 1950 had become an accepted 'law of nature'.

It is hardly possible to review the following decades, since the field has grown explosively. This period is characterised by the extensive use of radioactive isotopes produced in nuclear reactors and accelerators, the study of the dependence of diffusion on the tracer mass (isotope effect), and of diffusion under hydrostatic pressure. Great improvements in the precision of diffusion measurements and in the accessible temperature ranges were achieved by using refined profiling techniques such as electron microprobe analysis, sputter sectioning, secondary ion mass spectroscopy, Rutherford back-scattering, and nuclear reaction analysis. Methods not directly based on Fick's law to study atomic motion such as the anelastic or magnetic after-effect, internal friction, and impedance spectroscopy for ion-conducting materials were developed and widely applied. Completely new approaches making use of nuclear methods such as nuclear magnetic relaxation (NMR) [36], Mössbauer spectroscopy (MBS), and quasielastic neutron scattering (QENS) have been successfully applied to diffusion problems.

Whereas diffusion on solid surfaces nowadays can be recorded by means of scanning tunnelling microscopy, the motion of atoms inside a solid is still difficult to observe in a direct manner. Nevertheless, diffusion occurs and it is the consequence of a large number of atomic or molecular jumps. The mathematics of the random-walk problem allows one to go back and forth between the diffusion coefficient and the jump distances and jump rates of the diffusing atoms. Once the diffusion coefficient was interpreted in this way, it was only a question of time before attempts were made to understand the measured values in terms of atomistic diffusion mechanisms.

The past decades have seen a tremendous increase in the application of computer modeling and simulation methods to diffusion processes in materials. Along with continuum modeling aimed at describing complex diffusion problems by differential equations, atomic-level modeling such as ab-initio calculations, molecular dynamics studies, and Monte Carlo simulations, play an increasingly important rôle as means of gaining fundamental insights into diffusion processes.

Grain-boundary diffusion: By 1950, the fact that grain-boundary diffusion exists had been well documented by autoradiographic images [37], from which the ratio of grain-boundary to lattice-diffusion coefficients in metals was estimated to be a few orders of magnitude [38]. *Fisher* published his now classical paper presenting the first theoretical model of grain-boundary diffusion in 1951 [39]. That pioneering paper, together with concurrent experimental work by *Hoffman and Turnbull* (1915–2007) [40], initiated the whole area of quantitative studies of grain-boundary diffusion in solids. Nowadays, grain-boundary diffusion is well recognised to be a transport phenomenon of great fundamental interest and of technical importance in normal polycrystals and in particular in nanomaterials.

Distinguished scientists of solid-state diffusion: In what follows some people are mentioned, who have made or still make significant contributions to the field of solid-state diffusion. The author is well aware that such an attempt is necessarily incomplete and perhaps biased by personal flavour.

Wilhelm Jost (1903–1988) was a professor of physical chemistry at the University of Göttingen, Germany. He had a very profound knowledge of diffusion not only for solids but also for liquids and gases. His textbook 'Diffusion in Solids, Liquids and Gases', which appeared for the first time in 1952 [41], is still today a useful source of information. Although the author of the present book never had the chance to meet Wilhelm Jost, it is obvious that Jost was one of the few people who overlooked the whole field of diffusion, irrespective whether diffusion in condensed matter or in gases is concerned.

John Bardeen (1908–1991) and *C. Herring*, both from the Bell Telephone Laboratories, Murray Hill, New Jersey, USA, recognised in 1951 that diffusion of atoms in a crystal by a vacancy mechanism is correlated [42]. After this pioneering work it was soon appreciated that correlation effects play an important rôle for any solid-state diffusion process, when point defects act as diffusion vehicles. Nowadays, a number of methods are available for the calculation of correlation factors. Correlation factors of self-diffusion in elements with cubic lattices are usually numbers characteristic for a given diffusion mechanism. Correlation factors of foreign atom diffusion are temperature dependent and thus contribute to the activation enthalpy of foreign atom diffusion. It may be interesting to mention that John Bardeen is one of the very few scientists, who received the Nobel prize twice. Schockley, Bardeen, and Brattain were awarded for their studies of semiconducors and for the development of the transition in 1956. Bardeen, Cooper, and Schriefer received the 1972 Nobel price for the so-called BCS theory of superconductivity.

Yakov E. Geguzin (1918–1987) was born in the town of Donetsk, now Ukraine. He graduated from Gor'kii State University at Kharkov, Ukraine. After years of industrial and scientific work in solid-state physics he became professor at the Kharkov University. He founded the Department of Crystal Physics, which he headed till his death. The main scientific areas of Geguzin were diffusion and mass transfer in crystals. He carried our pioneering studies of surface diffusion, diffusion and mass transfer in the bulk and on the surface of metals and ionic crystals, interdiffusion and accompanying effects in binary metal and ionic systems. He was a bright person, a master not only to realise experiments but also to tell of them. His enthusiasm combined with his talent for physics attracted many students. His passion is reflected in numerous scientific and popular books, which include topics such as defects in metals, physics of sintering, diffusion processes on crystal surfaces, and an essay on diffusion in crystals [43].

Norman Peterson (1934–1985) was an experimentalist of the highest calibre and a very active and lively person. His radiotracer diffusion studies performed together with *Steven Rothman*, *John Mundy*, *Himanshu Jain* and other members of the materials science group of the Argonne National Laboratory, Illinois, USA, set new standards for high precision measurements of tracer diffusivities in solids. Gaussian penetration profiles of lattice diffusion over more than three orders of magnitude in tracer concentration were often reported. This high precision allowed the detection of small deviations from Arrhenius behaviour of self-diffusion, e.g., in fcc metals, which could be attributed to the simultaneous action of monovacancy and divacancy mechanisms. The high precision was also a prerequisite for successful isotope effect experiments of tracer diffusion, which contributed a lot to the interpretation of diffusion mechanisms. Furthermore, the high precision permitted reliable studies of grain-boundary diffusion in poly- and bi-crystals with tracer techniques. The author of this book collaborated with Norman Peterson, when Peterson spent a sabbatical in Stuttgart, Germany, as a Humboldt fellow. The author and his groups either at the University of Stuttgart, Germany, until 1984 or from then at the University of Münster, Germany, struggled hard to fulfill 'Peterson standards' in own tracer diffusion experiments.

John Manning (1933–2005) had strong interests in the 'Diffusion Kinetics of Atoms in Crystals', as evidenced by the title of his book [44]. He received his PhD from the University of Illinois, Urbana, USA. Then, he joined the metals physics group at the National Bureau of Standards (NBS/NIST) in Washington. Later, he was the chief of the group until his retirement. He also led the Diffusion in Metals Data Center together with *Dan Butrymowics* and *Michael Read*. The obituary published by NIST has the following very rightful statement: *'His papers have explained the significance of the correlation factor and brought about an appreciation of its importance in a variety of diffusion phenomena'*. The author of this book met John Manning on several conferences, Manning was a great listener and a strong advocate, fair, honest, friendly, courteous, kind and above all a gentleman.

Paul Shewmon is professor emeritus in the Department of Materials Science and Engineering at the Ohio State Univeristy, USA. He studied at the University of Illinois and at the Carnegie Mellon University, where he received his PhD. Prior to becoming a professor at the Ohio State University he served among other positions as director of the Materials Science Division of the Argonne National Laboratory, Illinois, and as director of the Division of Materials Research for the National Science Foundation of the United States. Shewmon is an outstanding materials scientists of the United States. He has also written a beautiful textbook on 'Diffusion in Solids', which is still today usefull to introduce students into the field. It appeared first in 1963 and in slightly revised form in 1989 [45].

The diffusion community owes many enlightening contributions to the British theoretician *Alan B. Lidiard* from AEA Technology Harwell and the Department of Theoretical Chemistry, University of Oxford, GB. He co-authored the textbook 'Atomic Transport in Solids' together with *A.R. Allnatt* from the Department of Chemistry, University of Western Ontario, Canada [46]. Their book provides the fundamental statistical theory of atomic transport in crystals, that is the means by which processes occurring at the atomic level are related to macroscopic transport coefficients and other observable quantities. Alan Lidiard is also the father of the so-called 'five-frequency model' [47]. This model provides a theoretical framework for solute and solvent diffusion in dilute alloys and permits to calculate correlation factors for solute and solvent diffusion. It has been also successfully applied to foreign atom diffusion in ionic crystals.

Jean Philibert, a retired professor of the University Paris-sud, France, is an active member and highly respected senior scientist of the international diffusion community. Graduate students in solid-state physics, physical metallurgy, physical and inorganic chemistry, and geophysical materials as well as physicists, metallurgists in science and industrial laboratories benefit from his comprehensive textbook 'Atom Movements – Diffusion and Mass Transport in Solids', which was translated from the French-language book of 1985 by *Steven J. Rothman*, then senior scientist at the Argonne National Laboratory, Illinois, USA [48]. *David Lazarus*, then a professor at the University of Illinois, Urbana, USA, wrote in the preface to Philiberts book: *'This is a work of love by a scientist who understands the field thoroughly and deeply, from its fundamental atomistic aspects to the most practical of its 'real-world' applications.'* The author of the present book often consulted Philibert's book and enjoyed Jean Philibert's well-rounded contributions to scientific discussions during conferences.

Graeme Murch, head of the theoretical diffusion group at the University of Newcastle, Australia, serves the international diffusion community in many respects. He is an expert in computer modeling of diffusion processes and has a deep knowledge of irreversible thermodynamics and diffusion. He authored and co-authored chapters in several specialised books on diffusion, standalone chapters on diffusion in solids, and a chapter about interdiffusion in a data collection [69]. He also edited books on certain aspects of diffusion. Graeme Murch is since many years the editor-in-chief of the international journal 'Defect and Diffusion Forum'. This journal is an important platform of the solid-state diffusion community. The proceedings of many international diffusion conferences have been published in this journal.

Other people, who serve or served the diffusion community with great success, can be mentioned only shortly. Many of them were also involved in the laborious and time-consuming organisation of international conferences in the field of diffusion:

The Russian scientists *Semjon Klotsman*, the retired chief of the diffusion group in Jekaterinburg, Russia, and *Boris Bokstein*, head of the thermodynamics and physical chemistry group at the Moscow Institute of Steels and Alloys, Moscow, Russia, organised stimulating international conferences on special topics of solid-state diffusion.

Deszö Beke, head of the solid-state physics department at the University of Debrecen, Hungary, and his group contribute significantly to the field and organised several conferences. The author of this book has a very good remembrance to DIMETA-82 [49], which took place at lake Balaton, Hungary, in 1982. This conference was one of the very first occasions where diffusion experts from western and eastern countries could participate and exchange experience in a fruitful manner, although the 'iron curtain' still did exist. DIMETA-82 was the starting ignition for a series of international conferences on diffusion in materials. These were: DIMETA-88 once more organised by Beke and his group at lake Balaton, Hungary [50]; DIMAT-92 organised by *Masahiro Koiwa and Hideo Nakajima* in Kyoto, Japan [51]; DIMAT-96 organised by the author of this book and his group in Nordkirchen near Münster, Germany [52]; DIMAT-2000 organised by *Yves Limoge and J.L. Bocquet* in Paris, France [53]; DIMAT-2004 organised by *Marek Danielewski* and colleagues in Cracow, the old capital of Poland [54].

Devendra Gupta, retired senior scientist from the IBM research laboratories in Yorktown Heights, New York, USA, was one of the pioneers of grain-boundary and dislocation diffusion studies in thin films. He organised symposia on 'Diffusion in Ordered Alloys' and on 'Diffusion in Amorphous Materials' and co-edited the proceedings [55, 56]. Gupta also edited a very useful book on 'Diffusion Processes in Advanced Technological Materials', which appeared in 2005 [57].

Yuri Mishin, professor at the Computational Materials Science group of Georg Mason University, Fairfax, Virginia, USA, is an expert in grain-boundary diffusion and in computer modeling of diffusion processes. He co-authored a book on 'Fundamentals of Grain and Interphase Boundary Diffusion' [58] and organised various symposia, e.g., one on 'Diffusion Mechanisms in Crystalline Materials' [59].

Frans van Loo, retired professor of physical chemistry at the Technical University of Eindhoven, The Netherlands, is one of the few experts in multiphase diffusion and of diffusion in ternary systems. He is also a distinguished expert in Kirkendall effect studies. Van Loo and his group have made significant contributions to the question of microstructural stability of the Kirkendall plane. It was demonstrated experimentally that binary systems with stable, unstable, and even with several Kirkendall planes exist.

Mysore Dayananda is professor of the School of Engineering of Purdue University, West Lafayette, Indiana, USA. His research interests mainly concern interdiffusion, multiphase diffusion and diffusion in ternary alloys. Dayananda has also organised several specialised diffusion symposia and co-edited the proceedings [60, 61].

The 150th anniversary of the laws of Fick and the 100th anniversary of Einstein's theory of Brownian motion was celebrated on two conferences. One conference was organised by *Jörg Kärger*, University of Leipzig, Germany, and *Paul Heitjans*, University of Hannover, Germany, at Leipzig in 2005. It was was devoted to the 'Fundamentals of Diffusion' [62]. Heitjans and Kärger also edited a superb text on diffusion, in which experts cover various topics concerning methods, materials and models [63]. The anniversaries were also celebrated during a conference in Moscow, Russia, organised by *Boris Bokstein and Boris Straumal* with the topics 'Diffusion in Solids – Past, Present and Future' [64].

Andreas Öchsner, professor at the University of Aveiro, Portugal, organised a first international conference on 'Diffusion in Solids and Liquids (DSL2005)' in 2005 [65]. The interesting idea of this conference was, to bring diffusion experts from solid-state and liquid-state diffusion together again. Obviously, this idea was successful since many participants also attended DSL2006 only one year later [66].

Diffusion research at the University of Münster, Germany: Finally, one might mention, that the field of solid-state diffusion has a long tradition at the University of Münster, Germany – the author's university. *Wolfgang Seith* (1900–1955), who had been a coworker of Georg von Hevesy at the University of Freiburg, Germany, was full professor of physical chemistry at the

University of Münster from 1937 until his early death in 1955. He established diffusion research in Münster under aggravated war-time and post-war conditions. He also authored an early textbook on 'Diffusion in Metallen', which appeared in 1939 [66]. A revised edition of this book was published in 1955 and co-authored by Seith's associate Heumann [67]. *Theodor Heumann* (1914–2002) was full professor and director of the 'Institut für Metallforschung' at the University of Münster from 1958 until his retirement in 1982. Among other topics, he continued research in diffusion, introduced radiotracer techniques and electron microprobe analysis together with his associate *Christian Herzig*. As professor emeritus Heumann wrote a new book on 'Diffusion in Metallen', which appeared in 1992 [68]. Its German edition was translated to Japanese language by *S.-I. Fujikawa*. The Japanese edition appeared in 2006.

The author of the present book, *Helmut Mehrer*, was the head of a diffusion group at the University of Stuttgart, Germany, since 1974. He was then appointed full professor and successor on Heumann's chair at the University of Münster in 1984 and retired in 2005. Diffusion was reinforced as one of the major research topics of the institute. In addition to metals, further classes of materials have been investigated and additional techniques applied. These topics have been pursued by the author and his colleagues *Christian Herzig*, *Nicolaas Stolwijk*, *Hartmut Bracht*, and *Serguei Divinski*. The name of the institute was changed into 'Institut für Materialphysik' in accordance with the wider spectrum of materials in focus. Metals, intermetallic compounds, metallic glasses, quasicrystals, elemental and compound semiconductors, and ion-conducting glasses and polymers have been investigated. Lattice diffusion has been mainly studied by tracer techniques using mechanical and/or sputter-sectioning techniques and in cooperation with other groups by SIMS profiling. Interdiffusion and multi-phase diffusion was studied by electron microprobe analysis. The pressure and mass dependence of diffusion has been investigated with radiotracer techniques on metals, metallic and oxide glasses. Grain-boundary diffusion and segregation into grain boundaries has been picked up as a further topic. Ionic conduction studied by impedance spectroscopy combined with element-specific tracer measurements, provided additional insight into mass and charge transport in ion-conducting oxide glasses and polymer electrolytes. Numerical modeling of diffusion processes has been applied to obtain a better understanding of experimental data. A data collection on diffusion in metals and alloys was edited in 1990 [69], DIMAT-96 was organised in 1996 and the conference proceedings were edited [52].

Further reading on history of diffusion: An essay on the early history of solid-state diffusion has been given by *L. W. Barr* in a paper on *'The origin of quantitative diffusion measurements in solids. A centenary view'* [71]. *Jean Philibert* has written a paper on *'One and a Half Century of Diffusion: Fick, Einstein, before and beyond'* [72]. Remarks about the more recent history can be found in an article of *Steven Rothman* [70], *Masahiro Koiwa* [73], and

Alfred Seeger [74]. Readers interested in the history of diffusion mechanisms of solid-state diffusion may benefit from *C. Tuijn's* article on *'History of models for solid-state diffusion'* [75]. Steven Rothman ends his personal view of diffusion research with the conclusion that *'... Diffusion is alive and well'*.

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1.2 Bibliography of Solid-State Diffusion

In this section, we list diffusion-related bibliography from the past four or five decades. Textbooks on diffusion in solids and some books that are devoted to the mathematics of diffusion are supplemented by monographs and/or books on specific topics or materials, and by stand-alone chapters on diffusion. Conference proceedings of international conferences on diffusion in solids and comprehensive collections of diffusion data complete the bibliography. The literature is ordered in each section according to the year of publication.

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