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Concrete material science: Past, present, and future innovations☆

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ABSTRACT

Concrete is flying off, but it is simultaneously facing tremendous challenges in terms of environmental impact, financial needs, societal acceptance and image. Based on an historical approach of the science of concrete and reinforced concrete in particular, this paper calls for the exploration of radical changes in three key aspects of concrete use: reinforcement, binder content, and implementation methods. More precisely, it is suggested that, in parallel to the introduction of robotic fabrication methods, digital technologies may be key for the introduction several innovations like (i) rebar-free reinforcement using non-convex granular media; (ii) compression-optimized concrete structures, using topology optimization, architectural geometry, and 3D-printing or origami-patterned formworks; (iii) truly digital concrete through the coupling of massive data collection and deep learning.

1. Concrete: Material, system and icon

Concrete, the mix of aggregates with water and cement, is flying off. Its best commercially traced and documented component, Portland cement or its variants, has been experiencing an unprecedented development since the turn of the millennium, matched only during a few years after WW2 (Fig. 1) [1–3]. Almost twenty years after this rebound, no obvious sign of slowing down is detectable, as this paper is written. Unloved by the majority and yet ubiquitous, concrete is one of the pillars of our developed societies, on equal foot with silicon, oil and gas, each in its own field: infrastructures, high rise, and large residential buildings for concrete; information and communication technologies for silicon; and, so far, transportation for oil and gas. More concrete is produced than any other synthetic material on earth. Twice as much concrete and mortar is used in construction – roughly 35 billion tons [4] – as the total of all other industrial building materials including wood [5], steel [6,7], plastic [8] and aluminium [9]. Roads, bridges, tunnels, dams, power plants, ports, airports, dikes and seawalls, waste- and fresh water plants and networks, all these infrastructures rely on the extensive use of concrete, just like the foundations of our buildings, if not the entire buildings themselves.

There is a wide consensus that the exceptional recent growth of cement and concrete consumption on the global scale is due to a handful of actors only among the emerging countries, China in particular [1,2]. But there are also good reasons to consider that the reason for this lasting growth resides in the current converging needs in developed and developing countries. Beside a huge affordable housing challenge, the world is presently also facing a fantastic infrastructure challenge [10,11]. Infrastructure is the foundation which makes social and economic life possible. It connects people, communities, and businesses. Developed countries face the challenge of maintaining and upgrading their extensive (Box 1) but ageing transport, power, water, and telecommunication networks, whereas developing countries dedicate a large fraction of their national income to satisfy basic human development needs – access to water, sanitation, electricity, and affordable housing – and still fall short of their goal. It is estimated that between now and 2030 an investment larger than the value of today's worldwide infrastructure (over 50 trillion dollars) will be required simply to keep up with projected global GDP growth [11]. The energy transition and the ongoing climate change are probably not going to mitigate the needs. Renewable energy facilities like wind farms require a substantial amount of concrete for their implementation and the rise of the oceans level will likely trigger the construction of thousands of km of protective dams. Actually, there seems to be no other material that could replace concrete in the foreseeable future to meet our societies' legitimate needs for infrastructure, housing, shelter and protection, by the unique property of cement and water transforming a pile of aggregates into rock in a few hours at room temperature.

The social picture is less engaging. Alternatively lauded or execrated, concrete is also the most controversial among all building materials. In spite-, and perhaps because-, of its emblematic role in the development of the modern world, it is crystallizing our expectations,

☆ This paper is dedicated to the memory of Gilles Chanvillard and Ellis Gartner.
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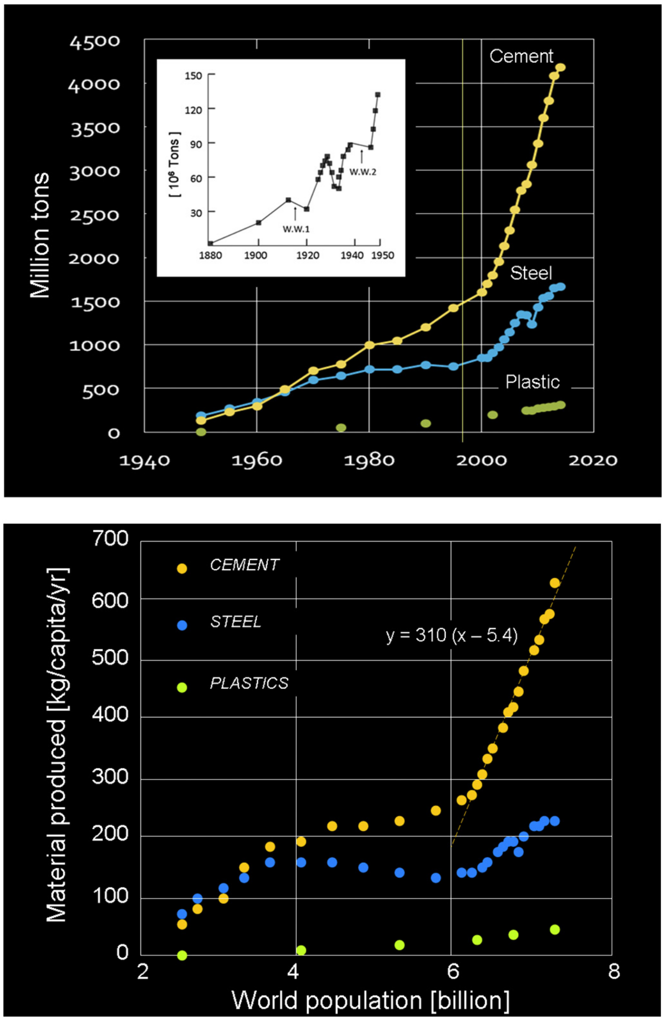

Fig. 1. Top: Comparative evolution of the post-WWII global cement, steel, and plastic productions (data from [1,2] and [6–8]). The inset shows the earlier cement production [3]. Note the strong impact of the 1929 economic crisis on cement production and the hardly noticeable effect of the 2008 crisis, faded away by the growth of emerging economies. Bottom: Same data plotted as material use per capita vs world population.
(Courtesy F.-J. Ulm).

our disappointments and sometimes our hates. Constantly oriented toward modernity and heavily loaded with moral values like honesty, simplicity, functionalism, optimism or communalism by star architects and city planners of the modern movement, concrete has been facing the violence of revolt when people came to compare the promises with the brutality of many urban renewal schemes in the late 20th century and the monotony of contemporary suburban development [20,21]. Widely perceived as dull and repetitive – "in short, a sort of frightening metonymy of the industrial age" [22] – concrete has been so far failing to meet its social promises. In our increasingly ecosensitive early 21st century, concrete is now also blamed for its contribution to carbon emissions and climate change, and to a variety of environmental problems like loss of farm land and increased vulnerability to natural hazards (floods in particular, due to increased imperviousness of soils), destruction of landscapes, loss of biodiversity, destruction of social link, loss of traditional constructive cultures, or depletion of natural resources, sand in particular [23,24]. Taken together, it is an extraordinarily severe indictment that concrete is facing. However, a bit of scrutiny is enough to realize that most of these criticisms are the objections to our dominating socio-economical model itself, concrete just happening to be a particularly ubiquitous and vivid symbol of this paradigm [25].

In a more technical perspective, concrete has also to accept its intrinsically multifaceted or even ambiguous nature. Compared with other building materials, concrete in general and reinforced concrete in particular is indeed heavily loaded with dichotomies [25]. Stretched between liquid and solid, granular and colloidal, gel and crystalline, smooth and rough, compact and porous, metal and mineral, compression and tension, brittle and ductile, material and process, material and structure, experimentation and computation, engineers and architects, technicality and art, worthless and precious, historical and unhistorical, concrete is permanently moving or transgressing the frame of taxonomy [25]. Frank Lloyd Wright went as far as to call it a "mongrel" material, being neither one thing nor another [26].

Actually, whether reinforced with rebars, tendons, fibers, or a combination of those, or even not reinforced at all, concrete is first of all a construction *system*, in which the material itself is intimately coupled to an implementation and a construction method. It was already so in the early days of the mid-nineteenth century, not much after the discovery of modern Portland cement, when the mixture of aggregates, cement and water was implemented in a barely wet state and rammed between movable form to make walls or on a falsework to make arches. Concrete became even more system-like when reinforcement was introduced. The hundreds of patents filed between ~1870 and ~1905 on the subject and the many companies to which they gave birth were all promoting concrete as a particular construction *system*, with a distinctive combination of matrix, reinforcement, structural type, and construction method, sometimes with the help of early computation methods [27–29]. The invention of prestressed concrete in 1928 [30], by removing the dichotomy between tension and compression (perfectly prestressed concrete is supposed to work exclusively in compression), was a radical change in the use of concrete. It might be considered as a step toward simplicity but, in practice, it is the opposite due to its increased computational content and the deep modification in design it led to. More recent developments like self-placing concrete (also termed self-compacting, self-consolidating, or self-leveling concrete) [31] or ultra-high-performance-concrete (UHPC) [32], follow the same trend. Both were initially intended to improve performances and to simplify the construction system (no vibration, less or no passive reinforcement), but the price to pay is a much sharper mix design and a loss of robustness. The same is true for 3D-printable concrete, which has the potential to lead to a totally new construction system. However, while the initial intend was to simplify the traditional construction process and to improve its low productivity [33], the final result is the massive introduction of digital technologies and new stringent requirements in terms of thixotropy and self-adhesion [34].

The *science* of concrete is actually a relatively recent science, contrary to that of cement. Many of the middle nineteenth century inventors were neither scientists nor engineers and several among the main innovators and company leaders of the end of the century were still self-made builders. The marriage between two materials as different as iron and mortar seemed counterintuitive and even counternature to many engineers. For some time, reinforced concrete was considered "uncomputable" [35]. In addition, two questions were shedding doubt on the durability of reinforced concrete. One was the adhesion of hardened cement to iron. Many were convinced that the iron-cement interface would fail soon or later. This led to some hard-to-implement reinforcement systems like the one patented by Paul Cottancin in 1889. Instead of heavy bars, Cottancin was using meshes made of one single iron wire with lots of convolutions supposed to compensate for the bad adhesion [35,36] (note that with the advent of robotic techniques, this may become an attractive technology, see Section 4). A patent was even filed in 1869 on the incorporation of glue in concrete in order to secure adhesion [35].

The other question that was impeding the development of concrete is the matching of the thermal expansion coefficients. Surprisingly, corrosion of the steel reinforcement was apparently not a major

Box 1

An incredible heritage

For non-specialists, it is hard to realize how incredibly large is the infrastructure heritage developed countries have built in a few centuries, beginning in the industrial revolution or even before for many of them. A medium size country like France, for instance, with a mainland area of 550,000 km^2 (roughly, a pentagon with 600 km long sides) has built more than 1 million km of two lane roads and close to 20,000 km of four- or six-lane freeways or national highways, almost exclusively with asphalt pavement [12]. More than 260,000 bridges have been built to avoid crossings [13]. The operating railway network has a total length approaching 30,000 km. Half of it is electrified and 2600 km are devoted to high speed lines operating at more than 300 km/hr [14]. More than 1000 km of road and rail tunnels have been dug [15]. The waste- and rainwater network, made essentially of large diameter concrete pipes, has an estimated length of 250,000 km [16] whereas the fresh water network is about four times longer [17]. Fifty-eight nuclear reactors (approximately one per million inhabitants) deliver about 75% of the total electric power [18].

When it comes to a large country like the United States – approximately ten million km^2, roughly a rectangle of 2500 km × 4000 km – the figures are just incredible [19]. The country is crossed by 6.5 million km of paved public roadways – enough to make eight roundtrips to the moon or 1600 coast-to-coast trips – and 250 thousand km of rail tracks. The interstate system alone has a total length approaching 80,000 km, enough to cross the country from Florida to Washington State more than 17 times. More than 600,000 road bridges and 76,000 railroad bridges are inserted in these entangled networks. However, the average age of the road bridges is currently 44 year and one in nine are rated as structurally deficient. Five hundred airports are serviced with commercial air carriers, and an additional 2800 are open to public use. Large areas of the country are protected by 84,000 dams, but 2000 are considered as deficient high-hazard dams. The wastewater system is already handled by 1.4 million km sewer mains and 15,000 treatment facilities, but fixing and expanding further the pipes is urgent to avoid overflows. With 99 commercial nuclear reactors producing about 20% of the national electric power, the United Sates are the world's largest supplier of electric energy (33%). Needless to say, these infrastructures – and others, not mentioned in this brief survey – rely heavily on the use of concrete.

concern, in spite of the young age of corrosion science. Again, it was not at all obvious that two materials as different as a cementitious matrix and iron could respond similarly to a temperature change. This was an important question because concrete was proposed as a good material for fireproof buildings. Interestingly, the empirical evidence supporting the good adhesion of hydrated cement on iron and the equivalence of their thermal expansion coefficients was rather quickly accumulated, but the fundamental basis for it has not been established yet. We don't understand the interactions responsible for the adhesion of cement on iron and the first paper devoted to a solid state- and statistical physics approach of the thermal properties of cement hydrates has been published only three years ago [37].

Actually, even when it is considered without its reinforcement, concrete is a remarkably rich deposit of interesting and contemporary research questions, all contained in its ambiguities and the actual or desired attributes they contain: granular or continuous?, liquid or solid?, crystalline or glassy?, smooth or rough?, "porous", brittle or ductile?, material or process?, etc.. Thanks to this, concrete and cement science could be a privileged subject in many research disciplines. One could think of the physics and mechanics of granular or porous media [38,39], rheology of dense and multiscale suspensions, heterogeneous nucleation and growth, crystallization in confined media, or else percolation, jamming and other rigidity transitions, just to name a few. Unfortunately, due to its undisputable but probably overestimated complexity and even coupling of complexities (chemical multi-composition coupled with vastly different length and time scales), concrete has been off-putting to many researchers, especially in times when complexity and multiscale problems were not yet fashionable topics and tools were not available to cope with them. In parallel, the science of concrete and the science of cement in particular may have had the tendency to lock themselves into their own world, proposing often material-specific explanations rather than looking for a broader frame. For instance, it is known since Le Chatelier [40,41] that the hydration of cement is a dissolution-precipitation process, but we had to wait till the end of the 20th century and the beginning of the 21st to see the fundamental theories of dissolution [42,43] and those of nucleation and growth at interfaces [44,45] applied to the early kinetics of cement hydration [46–51] (in defense of the many researchers who studied the dissolution of Portland cement, it was realized only recently that dissolution is just the reciprocal of growth and, like growth, a thermally activated kinetic process [52]).

This paper is not intended to be an inventory of the open questions that feed the passion of researchers in the cement and concrete communities. Not only would this be a very long inventory, it is the author's current opinion that it would also overemphasize the questions related to the chemistry of Portland-type cements including their heavily blended variants and their possible substitutes. While being undoubtedly key components of concrete and in particular those that are at the center of concrete's carbon footprint debate, binders and their hydration products do not necessarily represent the focal point where transformational progress of concrete as a construction *system* is possible. The approach adopted in this paper is to start from an historical analysis of a few basic aspects of concrete use − reinforcement, formulation, and implementation − in order to identify where potentially radical progress is possible.

2. Reinforced concrete: The perfect marriage?

"*Should human beings suddenly disappear from the face of the earth, the last century of our existence will be clearly discernable on hundred million years in the future by a unique, rust-colored layer of sediment found all over the planet…consisting of crushed and recrystallized concrete, tinged reddish brown by the oxidation of its now-vanished steel reinforcement bars*." This is how Robert Courland, in his Introduction to *Concrete Planet - The Strange and Fascinating Story of the World's Most Common Man-Made Material*, describes the geological stratum that might denote the reign of *Homo sapiens* [29]. It makes sense. The total mass of concrete manufactured since the middle of the nineteenth century should very soon exceed one trillion metric tons or 10^{15} kg (estimate based on the integral of the world's cement production curve, considering that the mass of concrete is 6 to 7 times larger than that of cement (Fig. 1)) and about one fourth of it is made of concrete reinforced with steel bars [4]. In a few more years this will be enough to cover the total surface of land masses (~150 × 10^6 km^2) with a layer a few mm thick, a thickness comparable to that of the Ir- and Ni-rich "K-T" boundary that marks the transition between the Cretaceous and Tertiary periods and the extinction of many mesozoic species, including dinosaurs [53].

The extensive use of reinforced concrete comes from the simple observation that the Achilles' heal of plain (unreinforced) concrete is its poor tensile strength. Concrete withstands very well large loads in compression thanks to its very dense network of granular contacts, but because it is still porous, brittle, and crossed by a myriad of water-covered interfaces, it doesn't withstand very well tensile efforts. It breaks suddenly when its elastic limit is reached. Raw (unbaked) earth is facing the same weakness, even more dramatically. This did not prevent builders from using plain concrete or raw earth for construction, by choosing the appropriate architecture. The world cultural heritage list contains hundreds of houses, churches, mosques, fortresses, or entire cities built with sun-dried clay bricks or rammed earth [54]. Using plain concrete made with pozzolanic cement, the Romans constructed one of the most elegant domes in the world – the dome of the Pantheon – still proudly standing in perfect shape more than two thousand years after its construction.

In the early 19th century, soon after the publication in 1818 of Louis Vicat's *Recherches expérimentales sur les chaux de construction, les bétons et les mortiers ordinaires* [55], François-Martin Lebrun applied the traditional technique of *pisé* (ramming earth layers between movable wall forms) to barely wet mixtures of aggregates and Portland-type cement, without reinforcement [27]. A few decades later in the mid-1870ies, another Frenchman, François Coignet, filed a patent and published a treatise on *Bétons agglomérés* ("compacted concrete") *appliqués à l'art de construire*, in which he was promoting basically the same technique as Lebrun and the monolithic character of the result [56]. Coignet's achievements were often not very durable but he succeeded in several remarkable cases, among which the Cleft Ridge span bridge, Brooklyn, in 1874, credited with being the first concrete arch in the United States [27]. Interestingly, Coignet was the first to use slag-enriched cements.

The idea to use iron or steel reinforcement to overcome the tensile weakness of concrete emerged almost concomitantly with the rise of the cement industry and in parallel with the use of plain concrete. It seems that the real discoverer of reinforcement of cement paste or mortars with metal is Joseph Louis Lambot, a farmer established in the south of France [27,28,35]. As soon as 1845, Lambot was making crates for his private use with an iron mesh covered with several layers of mortar, up to a total thickness of 3 to 4 cm. In 1848, he made a rowboat using the same technique. He used it for several years before filing a patent in 1855 for "a combination of iron and cement aimed at replacing timber". Lambot made a second rowboat and showed his invention at the Universal exhibition in Paris the same year, but it did not gain the success expected [57,58].

Another inventor, Joseph Monier, obtained more recognition. Monier started as a modest gardener, passionate by landscaping. Wooden crates and reservoirs were deteriorating rather rapidly and iron was still expensive and rusting. So, he made experiments with basically the same type of truss as Lambot – "armored cement" – for flower crates, pipes and water reservoirs. His first patent was filed in 1867, but many more (eighteen) followed from 1861 to 1891, for larger water reservoirs, staircases, fences, beams, floors, sewage pipes, and even "hygienic and economic, portable homes", as described in his French Patent no. 175,513 filed in 1886 [57]. In his patents for aqueducts, beams, footbridges, and railway sleepers, filed in 1877 and 1878, Monier was describing "a frame of round or square iron rods of any size and thickness, according to the strength that I want to give to them" [57], which is much closer to modern reinforcement than his initial trusses of iron wire. However, Monier himself had only moderate success and, probably, did not fully understand the mechanism of reinforcement. For instance, he did not understand that the reinforcement rods in beams or slabs had to be in the lower regions under tension [27].

In parallel with and following Monier's patents, the two last decades of the nineteenth century and the first years of the twentieth saw an abundance of developments and patents in Europe and in the United States which shows that the invention of reinforced concrete for construction cannot be granted to a single inventor [27–29,35]. In Germany and Austria, Conrad Freytag, Rudolph Schuster, and Gustav-Adolf Wayss bought the rights for Monier's reinforced beam. This led finally in 1885 to a very successful company, Wayss & Freytag, with numerous subsidiaries. Wayss himself wrote a book on "Das System Monier" in order to popularize – and to improve – the "MonierBau" [59].

The idea to use metal pieces to reinforce buildings was actually not totally new. Metal tie rods were already used in gothic and classical architecture in order to prevent stone walls from moving away. However, reinforcing mortar or concrete with metal is obeying a somewhat different logic. The very essence of reinforced concrete is to "link the load-bearing to the load-carried parts" [35]. It took several decades to discover how to achieve this with rebars of relatively small diameter in different configurations (beams, slabs, columns). In England, William Boutland Wilkinson was granted a patent in 1854 for "improvements in the construction of fire-proof dwellings, ware-houses, and other buildings" [29]. He emphasized the need to put iron girders or other metal pieces in the regions under tension and demonstrated his technology by building a cottage. However, just like Lambot's one, his patent didn't attract much attention. About twenty years later, in the United States, William E. Ward, a mechanical engineer, went one step further by realizing the importance of the strong adhesion of cement on iron in order to allow for the smooth load transmission from the concrete to the metal. He did experiments showing that the metal girders should be positioned in the lower part of the beam "in order for its tension force to resist to the load under the neutral axis when the composite beam is heavily loaded, while the beton above this line is resisting to the compression efforts" [60]. Ward built a large house – now known as "Ward's castle" – for himself in Port Chester, NY, using only Portland cement concrete reinforced with **I**- or **T**-shaped girders and rods of iron, but he didn't realize that he could replace them by something simpler and more efficient [29]. At about the same time, another American, Thaddeus Hyatt, made an important contribution by showing that the thermal expansion coefficient of iron and plain concrete are very close, proving that reinforced concrete beams would not collapse in case of fire [61]. In France, François Hennebique, a self-educated builder, was filed a patent in 1992, with an addition one year later on what became known as the "Hennebique system", with hangers linking iron rods. Hennebique was an ambitious businessman. In spite of losing a lawsuit for anteriority against two competitors, his company developed remarkably. In less than twenty years, it grew to a global size with 63 offices worldwide, including twelve in the USA. He even launched a monthly technical journal – Le Béton Armé – entirely devoted to his construction system [57].

Although the first true reinforced concrete civil engineering work was the bridge built in Wiggen, Switzerland, by Hennebique's company in 1892, the most iconic work of those early days is probably the sixteen stories Ingalls Building, the first reinforced concrete high rise building (16-story, 64 m), in Cincinnati [29,62]. Until then, high rise buildings were built in brick masonry. The Ingalls Building, completed in 1904, was built by the Ferro-Concrete Construction Company which had licensed the construction method of yet another self-educated builder and probably the last one at this level of innovation, Ernest L. Ransome. Ransome was an English immigrant established in San Francisco. Among several other inventions, he patented the incorporation of expansion joints in concrete slabs in order to prevent their cracking due to early age shrinkage. Above all, he may be considered as the inventor of the modern reinforcement iron bar, the "rebar" [29]. Experimenting with two-inch-thick square rods, he discovered that by twisting them he could obtain not only a better grip of the concrete but also a higher tensile strength. His U.S. patent no. 305,226 on a construction system using these "cold twisted" bars was granted in 1884.

The early decades of the twentieth century saw the spreading use of reinforced concrete. With limited computational help and often with a lot of intuition, engineers like Robert Maillart in Switzerland, Max Berg in Germany and Poland, Pier Luigi Nervi in Italy or Eugène Freyssinet in

France succeeded in building bold structures with slender simplicity [27–29,35,36]. Despite using himself the technique with great success, Freyssinet was considering that this alliance between materials mechanically as different as metal and concrete, in which both materials are forced to strain in the same way, was somehow counter-natural [63]. Freyssinet graduated from Ecole Polytechnique in Paris in 1899 and from Ecole Nationale des Ponts et Chaussées right after. Like others before him, he realized that in a system like reinforced concrete in which the tensile loads are carried by the metal, the concrete would inevitably crack, due to the much larger elasticity of the metal. His transformational idea was to design a system such that all the efforts – compression, tension, and shear – could be carried by the concrete alone, without cracking [63]. After many years of experimentation, he patented in 1928 the principle of pre-stressed concrete in which, thanks to steel tendons (cables or rods), a high permanent compressive load is applied to the concrete prior to any tensile effort [30]. The level of compressive loading is calculated to be at any time larger than the largest tensile effort which might be asked to the concrete. Thus, the concrete is never working in tension. When used with high strength (in compression) concrete and with steel tendons with a very high elastic limit (in tension), pre-stressing has made possible the construction of structures of remarkable slenderness. Freyssinet made several other discoveries [63]. In particular, we owe him the discovery of the irreversible delayed deformations of concrete under load – creep – which may ruin pre-stressing if they are too large. He was also precursor in the introduction of vibration and curing.

Reinforcement with rebars and pre-stressing improve the performances of structural elements but they leave the cementitious matrix unchanged. The matrix itself is still brittle, with a poor impact resistance. The introduction of discontinuous fibers – short with respect to the size of the structural element but long with respect to the size of the largest grains – is a simple way to improve both the tensile strength (moderately) and the fracture energy (very significantly) of concrete. By bridging advancing cracks, fibers increase many times the fracture strain of the matrix. Earth builders have been reinforcing mud bricks with bio-sourced fibers (straw, sisal, jute, …) almost since ever. In concrete, a first patent on the use of asbestos fibers was filed in 1902. Steel fibers were introduced around 1923 whereas glass fibers were introduced around 1950 when the harmful character of asbestos was recognized and alkali-resistant glass fibers were developed. Polymer fibers followed immediately.

With a moderate load of polymer, brass or steel fibers (typically a few % by weight, which is moderate in terms of weight or volume fraction but considerable in terms of mechanical percolation) well-dispersed in a well-graded and very dense cement-based granular matrix (see Section 3), concrete is hitting a world of properties in terms of strength, toughness, and ductility which brings it close to metallic materials [64,65]. The enhancement of compressive and tensile strength in these materials – fiber-reinforced ultra-high-performance-concrete or UHPC – is such that remarkably slender bridges or thin shells can now be built without passive reinforcement (rebars) (Fig. 2).

Yet other forms of reinforcement may find soon a significant development thanks to the advent of robotic construction techniques. The thin iron wire meshes used by Joseph Lambot to make his “ferro-cement” boat or those used by Joseph Monier to make his crates, pipes and reservoirs were not only intended to give bending strength to the layers of fine mortar which were pressed on it. They were also, in a very simple way, giving their shape – often curvilinear – to the final objects, something which is very attractive in contemporary architecture but which, using casting formworks and classical reinforcement, can be difficult, time-consuming and costly. Rediscovered and described by Pier Luigi Nervi in 1943, “ferro-cemento” evolved in a slightly different form. In order to increase the contact area between the reinforcement and the matrix, and to obtain a more homogeneous mechanical behavior, Nervi designed a system of multiple steel meshes fixed on bars of a larger diameter [66]. The mortar was poured within the stacked meshes, making the use of casting forms useless. This technique allowed him to design free complex shapes with thin concrete shells and to build several motor boats and an experimental warehouse as well [66].

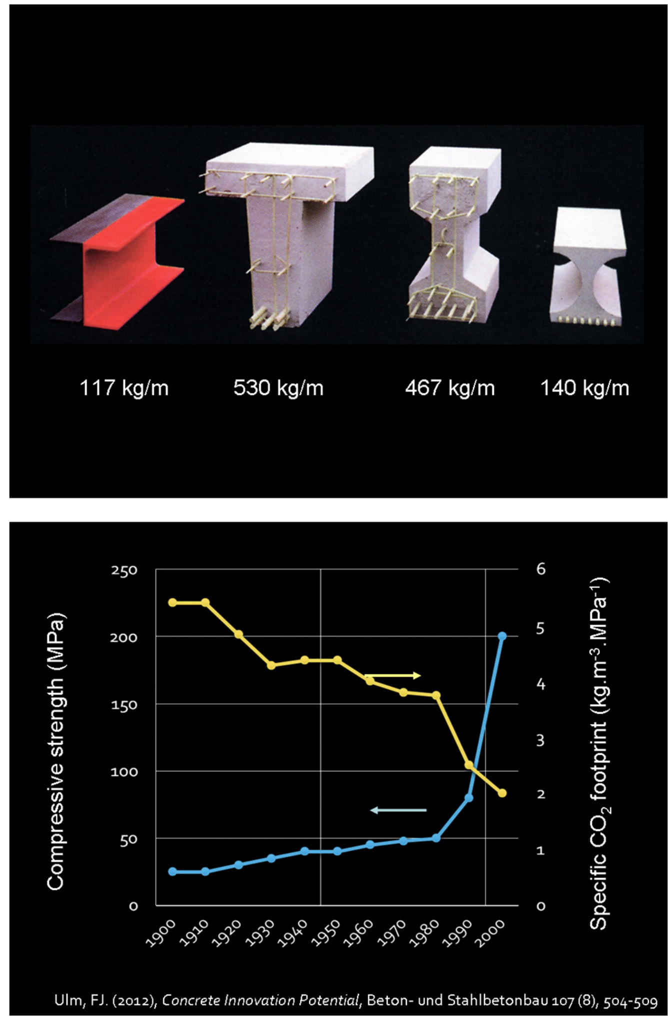

Fig. 2. Top: The four beams on this figure have the same load bearing capacity. The first is made of mild steel. The second is made of classical reinforced concrete with iron rebars, as it was used in the nineteen thirties. The third is made of prestressed high-performance concrete, with both rebars and prestress cables. The last one is in fiber-reinforced Ultra-High-Performance Concrete (UHPC), without any rebar, but with prestressing cables. It is only slightly heavier per unit length than the steel beam (+20%), whereas the more classical concretes are much heavier (+450% and +400%, respectively) (courtesy: Ph. Gégout, Bouygues). Bottom: Evolution of the compressive strength and average specific carbon dioxide footprint of concrete during the 20th and early 21st century.
(F.-J. Ulm, *Concrete Innovation Potential*, Beton- und Stahlbetonbau 107 (2012) 504–509, with permission).

In spite of their distinct advantages – they merge reinforcement and formwork into one single material item – mesh-like supports or moulds would probably remain anecdotal achievements had robotic fabrication not appeared on the construction scene. In 2013, Norman Hack and Willi Lauer demonstrated the feasibility of automated construction of free form meshes (they coined the word “mesh-mould”) using an industrial robot equipped with a polymer extruder [67]. Soon after, an automated robotic wire bending and welding tool for steel meshes was developed [68], opening the door to real reinforcement capabilities [69]. Fabrication of the mesh-mould is by far not the only difficult step in this technology. The development of computational tools for the design of topologically differentiated mesh morphologies is as important as fabrication itself. On the chemical and rheological side, the relationship between mesh aperture and concrete formulation and rheology (particle size distribution, viscosity, yield stress, thixotropy, surface tension, etc.) is critical for an appropriate concrete filling strategy [68,69]. The mesh-mould metal (M3) technology is described in more details elsewhere in this issue, in the broader context of robotic in situ manufacturing techniques [70].

At this point it is worth to stop and think. In a review of the Proceedings of the International Congresses on cement chemistry written after the 2007 conference in Montreal, Canada, J. Francis Young concluded that "*designing structures for 50-year or 100-year service life is already a reality*, *although our approach is probably not optimal*" [71]. It may be understood that in Young's mind, a 100-year service life was a good result, but that this was not solving all problems. Indeed, stepping back, one may wonder whether a power of ten is not missing in this conclusion. Are we ready to spend fifty trillion dollars (see Section 1) every century to renew our infrastructure? Obviously, the answer to this question is no. We cannot afford it. Accepting a 100-year service life is like building a disposable infrastructure world with a material – reinforced concrete – that would have a vanishing financial and environmental cost, something that present reinforced concrete doesn't have. On the other hand, building *all* our structures with a life span of one or two thousand years or more like the Romans did, doesn't make sense either due to the obsolescence of technologies. Just think of what nuclear power plants may look like – if they still exist – in a few centuries from now. In the same vein, the roads of the future will probably no longer be just the inert support for vehicles that we use today. They will most probably incorporate active functionalities allowing them to interact with vehicles and to adapt to climatic conditions. To try to keep the roads of today forever may be a wrong choice. On the contrary, designing an office or commercial building with a life span of several centuries could make sense. Europe is covered with wonderful ancient buildings, sometimes from the renaissance or classical periods that were reconverted for contemporary usage by skilled architects.

The question of durability and life span of concrete structures has to be raised here because corrosion of the reinforcement (rebars and pre- or post-tensioning cables) is the main reason for the decay of concrete. Thin slabs of robotically fabricated concrete with mesh-like steel reinforcement should be no different, or perhaps even more sensitive to corrosion. Ferrous materials are normally protected by a thin layer of iron oxide in the highly alkaline interstitial solution of concrete made with Portland cement or alkali-activated binders [72,73]. However, two processes are able to destabilize this "passive" layer. One is the carbonation and the subsequent pH drop < 9 due to ingress of atmospheric CO_2. The other is chloride ions penetration, mainly in marine environment, and the subsequent pH drop < 5 on the steel surface. This is why the absence of cracks and a very small diffusion coefficient D of gaseous and dissolved species are the key to durability. Quantitatively, in the absence of (micro)cracks, in order to have a penetration depth of CO_2 or Cl^- smaller than, say, 1 cm in a century, D should be of the order of a few $10^{-17}\,m^2\,s^{-1}$, to be compared to the experimental value of $\sim 10^{-8}\,m^2\,s^{-1}$ or higher for CO_2 diffusion and $\sim 10^{-12}\,m^2\,s^{-1}$ for chlorine ion diffusion in ordinary concrete [74,75]. The reliable prediction of the onset of corrosion in a reinforced structure is certainly more complex than this rough calculation of a root mean square displacement using a one-dimensional diffusion model in a semi-infinite medium, but the result gives an approximate estimate of what we must be looking for in terms of diffusivity if we want to go on using steel as reinforcement (in passing, it shows also that in a 100 year perspective the CO_2 uptake of concrete may be a significant fraction of its carbon footprint [76]). Values as low as $10^{-17}\,m^2\,s^{-1}$ are achievable with very compact UHPC-type matrices [77], but is the use of this type of concrete justified in all circumstances? On the other hand, corrosion may also be slowed down or arrested by a variety of means like the switch from steel to stainless steel, the use of corrosion inhibitors, the use of polymer coatings of rebars and tendons, or active control by impressed current cathodic protection [72,73,78].

Actually, if we allow ourselves to think out of the box, there is only one good solution: divest from reinforcement and perhaps also from prestressing as we practice it now, and design either a new type of reinforcement with non-corrodible materials, or a new type of concrete which does not require reinforcement thanks to a higher tensile strength and ductility. This will be addressed further later in this paper.

3. Concrete formulation: The art of packing, flowing, and gluing grains

In 1900, David Hilbert presented to the International Mathematical Congress in Paris a list of problems which he hoped would guide mathematical research in the beginning century. Problem # 18 was formulated as follows (cited by Aste & Weaire [79]): *I point out the following question* (…) *important to number theory and perhaps sometimes useful to physics and chemistry*: *How one can arrange most densely in space an infinite number of equal solids of given form*, e.g. *spheres with given radii or regular tetrahedral with given edges* (*or in prescribed positions*), *that is*, *how can one so fit them together that the ratio of the filled to the unfilled space may be as great as possible*? Considering the date at which Hilbert established his list, it is unlikely that he had the formulation of concrete in mind as a possible application of his challenge. Yet, the problem of finding the optimal granular formulation for concrete is not far from his problem # 18. Being essentially a load-bearing brittle material (for now), plain concrete is at its best in the state of highest compactness and lowest porosity. There is one important difference with Hilbert's problem though. In concrete, the "solids" to be arranged are not equal, and they have little chance to be ever arranged in an ordered fashion. They span a whole range of shapes and sizes, and their arrangement is the result of a "random" mixing process. This makes a significant difference.

The first to experiment seriously about this question was John Desmond Bernal. Bernal (1901–1971) was a prominent Irish-born scientist, with broad scientific interests going from crystallography to the origin of life and to social studies [80]. With his group at Birkbeck College, University of London, he made decisive contributions to the structure of biomolecules including cholesterol, vitamins, sex hormones, haemoglobin (with Max Perutz), ribonuclease (with Andrew Booth), and viruses (with Rosalind Franklin). With J.W. Jeffery and H.F.W. Taylor, he made also pioneering contributions to the crystallography of Portland cement and its hydration products [81]. Bernal was an exceptionally brilliant and charismatic person, attracted by frontier problems and unconventional approaches [80]. He was fascinated by the liquid state, this bridge between perfect order (the crystal) and perfect disorder (the gas) [82]. One of his great insights was to recognize that the lack of long-range order was no obstacle to local order. His main interest was on water but he considered also "simple" liquid, in which the molecules would be spheres clumping together in an unorganized packing [82]. By shaking and kneading thousands of ball bearings in bags, then compressing the assembly by winding it round with thick rubber bands, he managed to build dense random packings of hard spheres, which became known as Bernal packings. As the random clusters were disassembled, the positions of the balls were carefully measured and used to calculate the radial distribution functions. He found that, although particular local arrangements form recursively, they are variable in shape and random in distribution [83]. The solid volume fraction of Bernal packing was found to be only ≅64%, well below the ≅74% of Kepler's fcc and hcp dense regular packings.

The Bernal packing, with a solid volume fraction (or packing fraction, compactness, or density) of 0.64, can be obtained in an infinitely large number of ways, but many experiments and computer simulations do reproduce the same value (approximately, say within a percent or so). A consensus was rapidly reached that the value of 0.64 corresponds to the maximum value attainable by a packing of equal spheres in a random and dense arrangement [84,85]. Bernal packing is therefore often called *dense random packing* or, yet more frequently, *random close packing* (rcp), which may itself be replaced by the more rigorous *maximally random jamming* idea [86].

Looser packings can also be obtained if the densification step is skipped or made lighter. Not much after Bernal's investigations with ball bearings in bags, David Scott in Toronto decided to do similar experiments with spherical vessels of different sizes. In addition, he compared the results obtained in vessels that were gently shaken with those obtained without shaking. He observed that both the vessel size and the shaking did matter [85,87]. The smaller the vessel size, the looser the packing can be while keeping its mechanical stability. When the vessel was gently shaken to optimize the packing, the density was found to depend on vessel size as $\rho = 0.6366 - 0.33N^{-1/3}$, with N being the number of balls. When the vessel was not shaken, he obtained $\rho = 0.60 - 0.37N^{-1/3}$. The limit packing obtained with a large number of spheres in the later case is called the *random loose packing* (rlp) Its density has probably a less universal value than that of the random *close* packing, for it depends on external factors such as gravity and the friction between balls and with the vessel walls. The lowest densities that have been experimentally obtained for mechanically stable random packings of equal spheres are close to 0.555 [88].

Let us come back to the formulation of concrete. One of the properties of the dense random packing of equal spheres is that its solid volume fraction is independent of the sphere diameter (as long as the local arrangement of particles is not perturbed by friction or non-contact – attractive or repulsive – interactions, like in fine powders for instance), just like that of the hcp and fcc packings. Bernal's experiment performed with large or with small ball bearings would yield similar results, around $\rho \cong 0.64$, as long as short-range attractive forces do not perturb the game. There is no need of complex calculations to understand that this limit can easily be overcome by packing two or more populations of spheres together. Provided their diameters are sufficiently far apart, the small spheres will fit in the interstices of larger ones, leaving even smaller interstices to be filled by the third generation of spheres and so on (Fig. 3). With many carefully chosen populations, the mixture may have a density approaching unity in the infinite limit.

This type of recursive packing, known as Apollonian Packing, was already imagined around 200 BCE by Apollonius of Perga, a mathematician of the Alexandrine school. Apollonius studied the general problem of finding the circle that is tangent to three given objects – point, line, or circle – in a plane, but he could not solve the three-circle problem, also called the “kissing-circle” problem [79]. The exact solution, in the case of three mutually tangent circles of unequal radii, was obtained by René Descartes in 1643.

The Apollonian packing, with circles in a plane or spheres in three-dimensional space, is a classical example of a fractal, a word coined by Benoît Mandelbrot in 1975 to describe structures that are made of many similar elements with sizes that cover an infinitely large range of length scales [92,93]. Fractals are characterized by an effective dimension called the fractal dimension, d_f. It describes the structural cascade and the way a measure – length, area, mass, porosity, etc. – is scaling with the relative size of the measuring elements. In Apollonian packings it is the space left over between the circles or the spheres that is a fractal and that follows a scaling law:

$$\phi = 1 - \rho \approx \left(\frac{r_{\min}}{r_{\max}}\right)^{d-d_f} \tag{1}$$

In this relation, $r_{\min}$ and $r_{\max}$ are the radius of the largest and the smallest circle (or sphere), respectively, and d is the Euclidean dimension of space. ϕ is the porosity, i.e. the fraction of total space occupied by voids, and $\rho = 1 - \phi$ is the compactness or density, i.e. the fraction of total space occupied by the disks or by the spheres. Actually, this expression is valid for any fractal packing in the limit of a very broad power law size distribution ($r_{\min} < < r_{\max}$).

The fractal dimension of Apollonian packings is notoriously difficult to determine analytically, but its value has been closely approached by numerical simulations. For circles in a plane, it is close to 1.30 [94] and for spheres in 3D space it is close to 2.47 [95]. Both estimates were confirmed by adapting an approximate theory for the random close packing of polydisperse spheres [95]. Thus, according to Eq. (1), the residual porosity of a Apollonian packing of spherical grains should

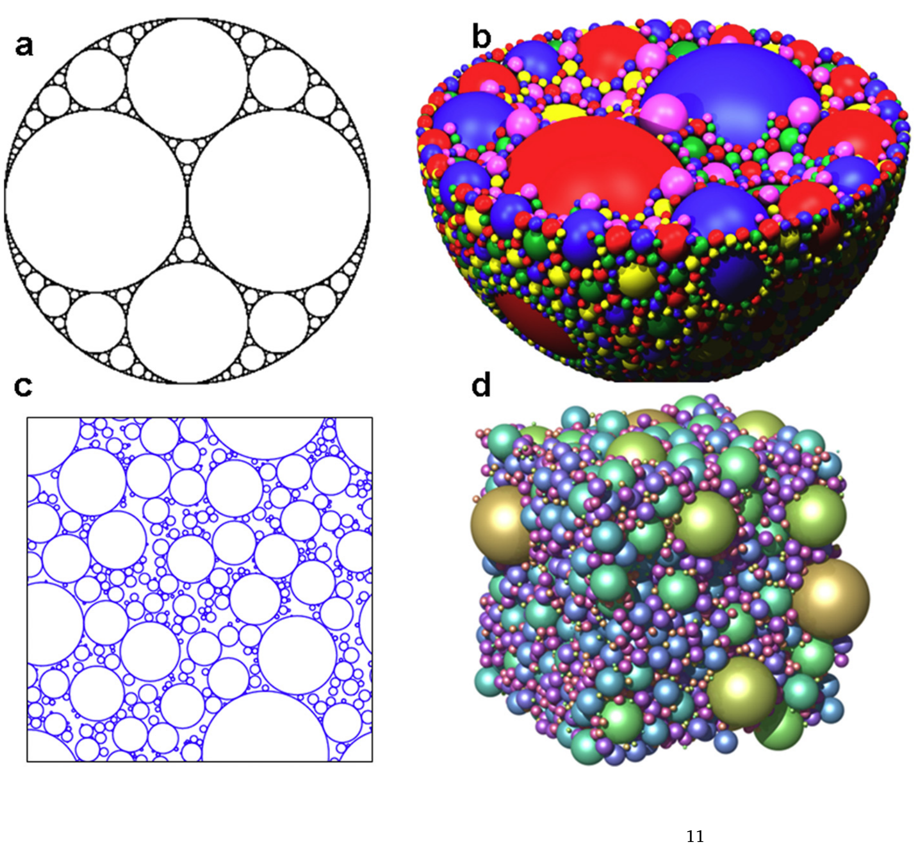

Fig. 3. (a): A “deterministic” or regular Apollonian packing of disks in a circular box; (b): same with spheres in a hemi spherical box (from [89]; (c): A random Apollonian packing of disks (from [90]). The centers of the disks are chosen randomly and the disks grow simultaneously with a linear growth rate until they collide with another disk; (d) random Apollonian packing of spheres [91].

scale as r_{min}/r_{max} to a power 3–2.47 ≅ 0.5.

Well before mathematicians and physicists addressed the question, civil engineers knew that there is a decreasing relationship between the minimum porosity of fresh concrete and its aggregate size range [96–100]. Remarkably, on the basis of simple symmetry and dimensional arguments, Caquot, a French engineer of the "Ponts et Chaussées" Corps, established a power law relationship that writes [101] (we owe also to Caquot the structure of the Corcovado Christ in Rio de Janeiro):

$$(e + v)_{min} = \alpha\left(\frac{r_{min}}{r_{max}}\right)^{1/5} \tag{2}$$

In this equation, α is a dimensionless parameter, and e and v are the volume of water and air, per unit volume (1 m^3) of fresh concrete, respectively. Their sum is the fresh concrete porosity, measured before any chemical reaction between cement and water. The suffix "min" means that the relationship applies to the minimum porosity (or maximum compactness) that can be reached by optimizing the amount of each granular population between r_{min} and r_{max}. This search for the most compact packing corresponds to common practice, although it is often not formulated like that. Any amateur builder knows that, in order to obtain a "good" concrete – a concrete that is easily workable right after mixing and strong after setting – he has to mix the "right" proportion of gravel, sand, cement and water. Professionals go one step further by introducing on purpose additional populations of smaller grains called "fillers" in the mix and decreasing thereby the ratio between r_{min} and r_{max} (Fig. 4) This is the basis, not only for higher mechanical performances, but also for improved durability.

Besides the fact that it was known well before fractals were "invented", Eq. (2) should also be awarded the merit to provide an estimate for the fractal dimension of the (pseudo)Apollonian packings of particles in optimized concretes. As illustrated in Fig. 5, where the minimum porosity of a number of concretes with very different compositions has been plotted vs their r_{min}/r_{max} ratio, the exponent 1/5 in Eq. (2) is remarkably robust. Comparing Eq. (1) with Eq. (2), this suggests that the fractal dimension looked for is close to (3–1/5) = 2.8, which is significantly larger than the fractal dimension of the ideal Apollonian packing of spheres in three-dimensional space (~2.47).

Actually, the perfect Apollonian packing of spheres is a very peculiar arrangement of particles, which requires the careful positioning of each grain. It is characterized by a discrete distribution of sizes and a rapidly decreasing cumulative mass- or volume distribution (mass or volume of particles passing a sieve with opening r). The distribution is clearly dominated by the larger grains (Fig. 3). In parallel, the residual porosity is also rapidly decreasing – with an exponent 0.53 – as finer

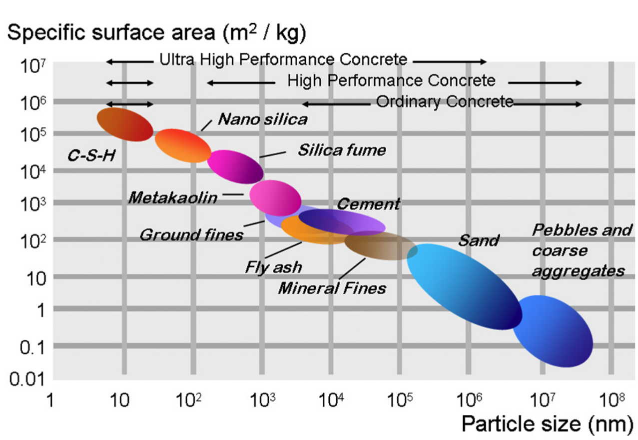

Fig. 4. A surface area vs particle size plot for the different granular populations − aggregates, fillers, cement, and hydrates − used in modern concretes. C-S-H is the main product of reaction of Portland cement with water.
Adapted from [102].

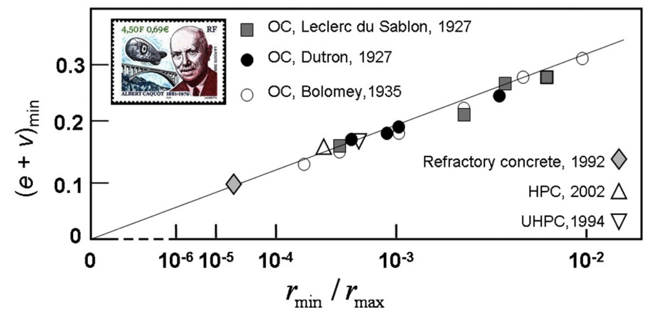

Fig. 5. Minimum porosity of 17 different concretes, plotted vs the ratio between the smallest and the largest particles in their formulation, r_{min}/r_{max}. The abscissa scale is proportional to (r_{min}/r_{max}) to the power 1/5. The radii used in this correlation are the harmonic averages for each population, $r = 3/\sigma$, where σ is the specific surface area per unit volume, measured by the Blaine method, a permeability-based classical method for surface area measurements in the cement and concrete industry. The stamp in the top left corner is a picture of Albert Caquot, who was able to predict the correct form of Eq. (2) on the simple basis of symmetry considerations [100].
Graph drawn with data compiled by J. Baron in [103].

populations are added and as r_{min}/r_{max} gets smaller. In real concretes formulated for minimal porosity in real implementation conditions, the exponent is much smaller (0.2) and the influence of adding populations of smaller grains doesn't diminish that much the residual void space. In addition, if there is no excess of water (not more water than what is needed to fill the residual porosity), the packing is incompressible. As far as mixing and flow are concerned, incompressibility is not a desirable property, because a system of hard convex particles cannot shear without dilating [38].

The difference between Apollonian packings and real optimal concrete mixes may be understood in terms of workability. It is unlikely that an Apollonian packing would spontaneously form simply by mixing and/or vibrating the right proportion of particles. The arrangement is so particular that it may only be obtained by placing the particles one by one by hand. On the other hand, a somewhat more spaced packing with an excess of particles of order i - 1 with respect to what would be needed in an Apollonian packing (Fig. 6) is much more likely to form spontaneously in a mixing process by a sort of lubricated coating mechanism. It should also be more workable and the residual porosity should decrease less rapidly as more populations are added, in agreement with the difference between fractal exponents. An additional difference between real concrete and the models discussed here is that the particle size distribution in real materials is not discrete. Each population has some distribution of sizes, so that the total distribution is much more continuous. Several "optimal" continuous particle size distributions have been proposed one of the most popular being the Andreasen and Andersen distribution [104].

Not all concretes are optimized for obtaining the minimum possible porosity with a given set of aggregates, sand, cement, sand and fillers, and not all concretes contain a variety of granular populations broad enough to adopt a fractal model. Yet, it is important to predict the properties of the mix, starting from those of each granular population, and taking into account the geometrical interactions between populations in the mix. This has led to several models [105–112]. Among those which take the interactions into account, the simplest one is probably the so-called *Linear Packing Density Model* for granular mixes, in which the perturbation introduced by a new class of particles in the mix is supposed to scale linearly with the volume introduced [110]. Two main perturbations are considered (Fig. 8): the "wall" effect on the one hand, and the "spacing" or "loosening" effect on the other hand. The wall effect accounts for the non-trivial fact that the local compactness of a granular medium is decreasing in the vicinity of a wall. The wall may be that of the container (like in David Scott's experiments described above [85]), but it may also be the surface of a larger grain.

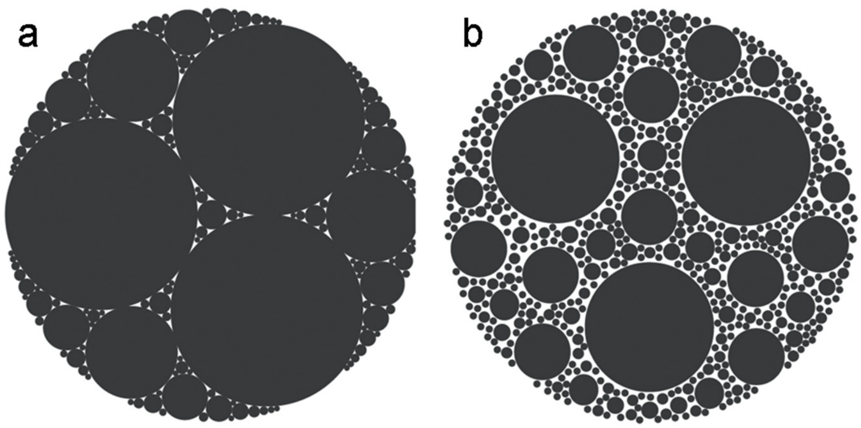

Fig. 6. Schematic illustration of the difference between an Apollonian packing (a) and a "spaced" packing (b) in which there is an increase in the clearance between particles of a given size by means of some excess of smaller particles.

The effect is noticeable over a distance from the wall approximately equal to ten times the grain diameter. The much more obvious spacing effect accounts for the fact that the forced introduction of a grain too large to naturally fit in the central cavity of a locally compact packing will increase the center-to-center distance. In addition to these two effects, the compactness of a given mix may be also limited by friction, aggregation of particles or, in more general terms, by the energy used to compact the mix. This led to the so-called *Compressible Packing Model*, with improved accuracy with respect to the linear model [111,112] and which recovers Caquot's relationship in the limit of infinite compaction energy.

Increasing the number of distinct populations of grains and broadening the overall particle size distribution toward the smaller end leads to the counterintuitive result that a more compact concrete may also be more fluid. This "liquid stone paradox" can be qualitatively understood by looking at how much water is needed to move the grains away from each other and to transform the dry packing into slurry. In a dry random close packing of equal spheres, we know that ~ (1–64%) = ~36% of the total volume is void space. Thus, one has to add at least that same quantity of water before the grains get somewhat spaced and become able to flow, that is, to pass over each other. This is a pretty large quantity of liquid. On the contrary, in an optimally formulated polydisperse packing, the void space is much smaller and even a tiny amount of liquid may be enough to space the grains out and to transform a dry heap into a slurry. Attending the preparation of a UHPC mix is in this respect an astonishing experience. As the operator is pouring a ridiculously small volume of liquid (water with some superplasticizer) in the carefully graded powder mix, the audience looks, sceptical and dubious that this could ever change anything. The mixing paddle keeps turning into the bowl but the powder remains desperately dry. Yet, after a few tens of seconds, the mixture suddenly turns into remarkably homogeneous and fluid slurry. The waiting time during mixing was just the time necessary to expel the air and to replace it by water.

This behavior may be approached in a more quantitative but still simplified way by starting from the other end, the pure liquid, and looking how the viscosity of that liquid is evolving as solid particles are added. It is a universal observation that the dispersion of particles in a fluid makes the mixture more viscous than the neat suspending fluid [113–115]. It can even – if the particles interact with non-contact attractive or repulsive forces – transform the liquid into a soft solid which flows only when a threshold stress (the "yield stress") is exceeded. The main control parameter is, once more, the volume fraction of particles. In general terms, a larger viscosity means that additional sources of energy dissipation have been introduced with the particles. Two sources of energy dissipation have to be considered. The first one is hydrodynamic lubrication, which is the hydrodynamic force required to squeeze the liquid out of the confined space between two approaching particles or, symmetrically, to suck the liquid up in the gap between two separating particles. Lubrication forces tend reversibly toward infinity as the gap closes to zero so that, in theory, particles never touch each other in a suspension. In reality, all particles exhibit some surface roughness which, provided the pressure is high enough, allows for direct contact on asperities. This introduces a second source of dissipation through frictional contact, provided the particle concentration and the shear stress are large enough. This contribution of non-hydrodynamic frictional forces to the bulk viscosity of dense suspensions – and cement paste as well as concrete belong to this category – has long been neglected in spite of experimental evidence [116–119], but it is now recognized as a possible major contribution to the shear stress, even in suspensions of Brownian (colloidal) particles [120]. Frictional contacts may also contribute to the yield stress of mortars and concrete. For a more detailed discussion of the physical parameters which govern the rheological behavior of cement-based materials, including yield stress, shear rate dependence and thixotropy, the reader is referred to [121] and to the contribution by N. Roussel in this issue [122].

It is not straightforward to quantitatively determine the contribution of hydrodynamic dissipation and that due to contact forces in the macroscopic viscosity of suspensions but, all together, the viscosity of a dense suspension of particles can usually be accounted for by the following equation, proposed empirically by Krieger and Dougherty [123]:

$$\eta = \eta_0 \left[1 - \left(\frac{\phi}{\phi_m}\right)\right]^{-[\eta]\phi_m} \tag{3}$$

In this expression, η and η_0 are the viscosity of the suspension and of the neat suspending fluid, respectively; ϕ is the actual solid volume fraction in the slurry and ϕ_m is its maximum possible value in the actual flow conditions. [η] is a numerical parameter which was chosen equal to 2.5 by Krieger and Dougherty in order to recover the very dilute suspension limit obtained analytically by Einstein in 1906 [124]: $\eta = \eta_0(1 + 2.5\phi)$.

As ϕ approaches $\phi_{\max}$, the system approaches a state of jamming and the viscosity diverges toward infinity. Beyond ϕ_m the suspension is completely jammed and homogeneous flow is impossible. With equal spherical particles in quasi-static conditions (very low shear rates), we know that ϕ_m is $\cong 0.64$, but ϕ_m may be somewhat larger at high shear rate when the particles tend to align in the flow lines [105]. On the contrary, when friction is important, ϕ_m may be smaller, down to the random loose packing limit (rlp $\cong$ 0.55) when the friction coefficient becomes very large [120]. Whatever the flow and friction conditions, ϕ_m is larger with a broad distribution of particle sizes than in the monodisperse case. Hence, for a given value of ϕ, the ratio ϕ/ϕ_m is lower in the later case and the viscosity is also lower. Thus, by broadening and optimizing its hierarchical grain size distribution, one can simultaneously improve both the strength and the fluidity of a concrete. This leads to the quasi-magic practical result that adequately formulated very dense concretes may be simultaneously particularly easy to pump, particularly resistant to solid-liquid segregation (large grains tend to sediment while fine particles tend to form a supernate), and particularly strong after setting.

The properties just mentioned – density, fluidity (or "workability"), and resistance to fluid-solid segregation – are shared, to different degrees, by ultra-high-performance concrete (UHPC) and self-compacting concrete (SCC). SCC is a highly flowable, non-segregating, and generally high-performance and durable concrete that can spread into place, fill the formwork and encapsulate the reinforcement without any mechanical help (no vibration) [31,125]. It represents a significant improvement for concrete use, both in pre-cast plants and on the work site. In addition to fluidity and absence of segregation, SCC must also have a good passing ability, which is imposing a reinforcement-specific upper limit to the size of the largest aggregates [126]. It must also have a vanishing yield stress in order to avoid complete arrest as the flow rate decreases [126]. While being somewhat less fluid than SCC due to its lower water content and its even higher density, UHPC conserves these attributes. Needless to say, the particle size distribution is critical in the formulation of SCCs [127,128] and UHPCs [110,111,129].

None of these development would have been possible without what R. Flatt called the "spices of concrete", the chemical admixtures which, when added in the right proportions, can radically change essential properties of the mix [130]. Chemical admixtures may take care of various tasks [131]: to inhibit corrosion of rebars (to some extent), to reduce the shrinkage due to capillary stresses, to retard or to accelerate setting, to entrain air in order to protect concrete from frost damage, to increase the viscosity of the interstitial solution in order to prevent segregation or, most important, to prevent the formation of aggregates (or flocs) of cement and other fine particles.

The formation of flocs is due to short-range attractive forces (short-range with respect to the cement particles size), mainly van der Waals or ionic correlation forces. It has important consequences on the rheology of suspensions, and cementitious slurries are no exception. Flocs have generally a complex, often fractal structure in which a significant volume of suspending fluid is screened from the hydrodynamic forces that surround the floc (a common wording is that some fluid is "trapped" in the flocs) [132,133]. Due to this, a floc is behaving as if it had an effective flow-free volume larger than the real volume fraction of its constituting particles. Thus, flocculation is increasing the viscosity of a suspension (at least as long as the flocs do not sediment). Flocculation may also lead to gelation of the whole mixture if the floc network percolates. This gives the medium the properties of a soft solid, with a yield stress which can be quantitatively related to the degree of flocculation [134].

The admixtures in charge of preventing the formation of flocs are dispersants, called "water reducers" or "superplasticizers" in the cementitious nomenclature. Most of them are negatively charged polyelectrolytes that adsorb on the positively charged surfaces of the dissolving cement particles and probably also on other fine particles like silica fume for instance [135,136]. Thanks to mobile molecular segments protruding into the solution, or to clouds of mobile counterions retained captive by the surface charge, or both, they generate a repulsive force of osmotic nature when two particles approach each other. Provided their action is not impeded by other attractive interactions, the net result is that flocculation is prevented and the mix turns into a less viscous suspension of individual grains. Without this, the earlier discussion on the void space of granular packings in mix design would be meaningless.

In spite of tremendous progress in the design and synthesis of robust dispersants adapted to the surface chemistry of Portland-type cement, the present generation of superplasticizers is probably still perfectible. Current superplasticizers are designed to avoid floc formation, which is indeed a priority objective in the overwhelming majority of situations. However, they are not designed to lubricate contacts and to avoid friction which, as pointed out above, may be a major contributor to viscosity, in particular at large solid fraction and at high shear stress. This is all the more important since superplasticizers, by transforming soft and deformable particles (flocs) into hard and rigid particles (the individual grains), favour the intervention of direct contact forces [118,137]. In forming processes like extrusion or 3D-printing, this cannot be neglected (for a full discussion of the role of admixtures in the context of digital concrete, the reader is referred to the contribution of D. Marchon in this issue [138]).

At this point, just like in Section 2, it is worth to stop and think. The point here is about the best possible use of cement. The current correlation between concrete performance – strength and durability – and cement use is simple: higher performances require higher cement contents, lower water to cement weight ratios (W/C), and also an extended population of grains toward the smaller sizes. Typically, the cement content of an ordinary concrete (OC) reaching a compressive strength of 25 MPa after 28 days will be around 350 kg/m^3, with a W/C between 0.4 and 0.6 depending on the targeted consistency, most often without superplasticizer; That of a 60 MPa high performance concrete (HPC) will be over 450 kg/m^3 with W/C around 0.35 and addition of silica fume, and that of a 200 MPa UHPC will approach 700 kg/m^3, with a W/C under 0.20, addition of silica fume and other fines, and the compulsory use of superplasticizers. Simultaneously, the quality of the cement – that is, the compressive strength of the cement itself – will also increase as we go from OC to HPC and UHPC.

The simplest way to understand the logic behind this evolution is to go back to the empirical relationship discovered more than a century ago by R. Féret [139]. Féret was a tireless experimentalist and thanks to a huge number of experiments he established a simple relationship between compressive strength and formulation which writes:

$$f_c(t) = k_f f_{mc}(t)[c/(c + w + v)]^2 \tag{4}$$

In this equation $f_c(t)$ and $f_{mc}(t)$ are the compressive strength of the concrete and the normalized mortar prepared with the chosen cement at time t, respectively; $f_{mc}(t)$ is an indicator of the "quality" of cement. The quadratic term contains three variables, c, w and v which are the volume of cement, water and air per unit volume of concrete, respectively. The parameter k_f (the "f" subscript stands for "Féret") is supposed to account for everything else, in particular the granular formulation of the aggregates. Alternatively, instead of time, one can use the advancement of the cement hydration reaction α as time-dependent parameter:

$$f_c(\alpha) = k_f f\,[\alpha c/(\alpha c + w + v)]^2 \tag{5}$$

In this modified equation f is the strength of the normalized mortar when the cement is fully hydrated.

A few decades after Féret and independently, J. Bolomey [100,140] obtained another relationship which writes:

$$f_c(t) = k_b f_{mc}(t)[c/(w + v) - 0.5] \tag{6}$$

Bolomey's equation (where the subscript "b" stands for "Bolomey") gives results very close to that of Féret for ordinary concretes.

A few more decades and T.C. Powers showed that the strength of concrete at a given stage of cement hardening was a power function of a variable Y such that [141,142]:

$$f_c = C.\chi^3 \quad \text{with} \quad \chi = \frac{V_{hydrates}}{V_{hydrates} + V_{capillary\ voids}} \tag{7}$$

χ, termed the "gel space ratio", has a straightforward physical meaning. It is a measure of the effectiveness with which the hydrates generated by the reaction of cement with water are able to fill the void space. Powers showed also that the volume of Portland cement hydrates is ~2.15 times larger than the volume of the anhydrous cement they come from [141,142]. So, the volume of hydrates generated at a given time t is $2.15\alpha(t)c$. Assuming that a part of the hydrates is just replacing the free space generated by the reaction of cement, the volume of hydrates able to seal off the capillary void space is $1.15\alpha(t)$c. The volume of capillary voids left over at any time is just the difference between the initial void volume $e + v$ and the volume of hydrates sealing off the voids. The Y parameter from Powers writes now:

$$Y = \frac{2.15\alpha c}{2.15\alpha c + w + v - 1.15\alpha c} = \frac{2.15\alpha c}{\alpha c + w + v} \tag{8}$$

This is formally equivalent to the rhs term between brackets in Féret's relationship (5), provided the 2.15 factor is included in the k_f parameter. To our best knowledge, this formal equivalence of Féret's and Powers relations (with the exception of the exponent) was first pointed out by P.-C. Aïtcin, J. Baron and J.-P. Bournazel [143].

The point here is not to discuss details of these relationships (or the many others that have been proposed) such as the numerical value of the exponent for instance. The focus of our discussion is the fact that they all show that there are two routes to improve the compressive strength of concrete. One route is to optimize the hydration-dependent term that is, to generate as much hydration product as possible to fill the void space. The other is to optimize the multiscale granular packing that is, to minimize the void volume to be filled by the hydration products. As far as the first option is concerned, Féret's formulation leads to the straightforward conclusion that increasing the cement content and decreasing the water and air content is the simplest way to increase strength. Powers formulation goes deeper into the hydration process in the sense that it suggests that what really matters is the space-filling character of the hydrates. In practice, it is clear that increasing the cement content of concrete is (unfortunately) the route chosen by most builders. Controlling the space-filling character of the hydrates is subtler task.

Among the different hydrates that Portland-type cements are generating, calcium-silicate-hydrate (C-S-H) [144,145] and its Al-enriched version (C-A-S-H) [146,147] remain undoubtedly those to consider in priority, due to their leading role in the hardening of cement paste and concrete [148] and also in their creep [149,150]. There is now ample evidence that C-S-H in real cement pastes is an assembly of "nano-particles" [149,151–156], these nano-particles being themselves stacks of highly disordered layers, with an increasingly three-dimensional bonding scheme as the C/S ratio increases [157,158] (for a recent review of the various models proposed for calcium silicate hydrates, the reader is referred to [159]). Their precipitation mechanism is usually considered to proceed primarily via a heterogeneous nucleation mechanism, either on the surface of the anhydrous cement grains or on the surface of supplementary materials like slag or silica fume [49,160–164]. Therefore, a possible way to modify the space-filling ability of C-S-H or C-A-S-H − and simultaneously the overall hydration kinetics − is to favour homogeneous nucleation in parallel with heterogeneous nucleation. This can be achieved by the introduction of nucleation seeds in the interstitial solution, as it was already suggested some decades ago, essentially to accelerate setting and hardening [165], and investigated later for more fundamental purposes [166,167]. The main problem for all practical purposes is to prepare a stable (no or limited aggregation, no sedimentation, no adsorption) colloidal suspension of C-(A)-S-H seeds. This can be achieved by protecting the seeds with adsorbed polymer molecules, but the operation is tricky. Too much protection and the seeds are no longer able to incorporate monomers and to grow. Too little, and the seeds may aggregate and loose accessible surface area. A right combination of protecting molecule and surface coverage of the C-S-H seeds was found in 2010 at BASF, using comb-type copolymers akin to superplasticizers [168–170]. In parallel with massive precipitation of C-S-H in the capillary pores leading to a much more homogeneous distribution of the hydrates (Fig. 8), a large increase in the early rise of compressive strength of concrete was obtained, especially at low temperature [168,171]. However, seeding failed to improve the long term strength. This shows that, apart from a kinetic benefit (which is very important in current construction practice and even more in several digital fabrication technologies with concrete [172,173]), homogeneous seeding did not succeed in increasing the average density of the hydrate paste. After all, this is what Powers relation is telling us: as far as compressive strength is concerned, it doesn't really matter where and under which form the hydrates precipitate, as long as the molar volume ratio between hydrates and anhydrous products is constant.

A lesson to be learned from this is that, as far as the long term compressive strength of concrete is concerned, there is probably little practical progress (but a huge cognitive progress and interesting opportunities as far as early age properties are concerned) to expect from attempts to unveil and to modify the nucleation and "pseudo-growth" process of hydrates and that of C-(A)-S-H in particular at the meso-scale, that is, the process leading to its structure at length scales going from a few nm to the size of the capillary pores [174]. In our present state of knowledge, this meso-scale structure is considered to be the result of a multi-step process involving (i) nucleation and limited growth of the nuclei to nano-sized particles [49,50,162], (ii) self-catalyzed reproduction by secondary nucleation [175,176], (iii) aggregation, driven by a non-monotonous interaction potential between nanoparticles [177–180], as evidenced by experimental AFM measurements [181] and, finally, (iv) possible crystallization of dense multi-particle domains [175,176,182,183]. The seeding experiments tell us that manipulating this process with the aim of changing the meso-scale structure can only have a second order influence on the final properties. The first order parameter remains the relative volume of hydrates.

The straightforward consequence of this is that the simplest way to increase the compressive strength of concrete via the hydration reactions (the other route, via the granular skeleton will be examined next) is to increase the amount of hydrates generated per unit volume of clinker, that is, to increase the "2.15" factor in Eq. (8). In theory, this can be achieved by adding so-called "Supplementary Cementitious Materials" (SCM) [4184]. The major families of SCM are silica fume (SF), fly ashes (FA), blast furnace slag (BFS), metakaolin (MK) or more generally calcined clays (CC), and crushed limestone (CLS). In a CaO-Al_2O_3-SiO_2 composition diagram [183], slag is not very far from clinker and lies on the high basicity corner of the diagram. It is itself a hydraulic material, like clinker, but it reacts more slowly. It forms a variety of hydrates similar to those formed by neat Portland cement. On the other hand, silica fume, fly ash, metakaolin and calcined clays are closer to the acidic side of the phase diagram. They have no hydraulic character, but they share with volcanic ashes – in particular, those of the city of Pozzuoli near Naples – the ability to react with lime and to produce C-(A)-S-H-type hydrates. This was the way Romans were preparing their binder and this property is called pozzolanicity. Most volcanic ashes are glassy silica-aluminas and upon reaction with lime they produce C-A-S-H hydrates. Fly ash and metakaolin (and similar types of calcined clays), have also a high alumina content and they produce the same type of C-A-S-H hydrate. On the other hand, fume silica, which is almost pure amorphous SiO_2, produces essentially Al-free C-S-H. Finally, crushed limestone is neither hydraulic nor pozzolanic, but its surface is able to nucleate C-S-H hydrates [185]. Thus, whether by their hydraulic (BFS), pozzolanic (SF, FA, MK, CC), or nucleating (CLS) properties, the addition of SCMs to clinker or the substitution of clinker by SCMs leads basically to the same result: as much or even more hydrates than in unblended cement.

However, going from this qualitative statement to a quantitative prediction of strength is far from obvious. The gel space ratio concept has to be revisited or re-calibrated for each SCM, due to the unequal contributions of the various hydrates to strength. Actually, a much more sophisticated approach emerged. Thermodynamic modeling is the first step [186]. It enables us to predict the nature of the hydrates as well as the composition of the aqueous phase after long hydration times. Remarkably, comparison with experiments shows that with the exception of C-(A)-S-H which remains metastable, the phase assemblage readily reaches equilibrium [186]. The next step is the coupling with a kinetic model which, on the basis of the thermodynamic model, enables us to predict quite accurately the evolution of the phase assemblage with time during hydration of Portland and blended cements [187,188]. The last step is the calculation of mechanical properties − elasticity and compressive strength − using multi-scale micromechanical modeling [148,189,190]. This represents a major evolution with respect to empirical equations. It is the right framework to incorporate the detailed meso-scale models [175–180] but much progress is still needed before it could be considered as a routine method.

Returning to Féret's or Powers equations, we have to examine the second possible route leading to stronger concrete, buried in the k_f (or k) parameter. As extensively discussed in this Section, hardened concrete is primarily a multi-scale granular packing or, more exactly, a cohesive-frictional granular packing. The structure of this packing controls the way the applied stress propagates and possibly concentrates, leading to failure. At first order, its average density (or the dual quantity, its average porosity) is the primary factor controlling strength. The value of k_f is a measure of the effectiveness of a given packing to sustain stresses without failure, for a given degree of filling of the capillary pores of the cement paste by the hydrates.

It is not obvious to untangle the improvement coming from a larger content in cement and/or from addition of reactive fine materials, from that of a better (i.e. higher density) granular formulation, for both factors are generally modified concomitantly. An interesting attempt to achieve this separation is to calculate the so-called *binder intensity index*, *bi*, as proposed by Damineli et al. [191,192]. This index measures the total amount of binder necessary to deliver one unit of a given performance indicator, e.g. 1 MPa of compressive strength. It reads:

$$bi = \frac{b}{p} \tag{9}$$

where b is the total consumption of binder (kg·m^{-3}) and p is the performance requirement, e.g. compressive strength after curing. Fig. 7 illustrates the distribution of bi_{cs} as a function of the compressive strength for close to one thousand different concretes of international origin [83]. A first interesting observation relates to the scatter of the bi_{cs} values, which is very broad on the low performance side of the data points cloud but becomes considerably narrower toward the high-performance end. The "mechanical yield" of cement in some low strength ordinary concretes is quite good (low bi_{cs} values), but in some others it is very bad (high bi_{cs} values). This illustrates the fact that poor strength performances can be obtained in many different ways (i.e. with many different formulations), while high performance concrete obeys more accurate formulation rules.

A second observation is that the average binder intensity index tends clearly to decrease with the increase in compressive strength, down to a plateau value of about 5 kg cement·m^{-3}·MPa^{-1}. This shows that cement is best used in high performance mixes, using superplasticizers and addition of fines in order to have a quasi-continuous distribution of particle size (Fig. 4). However, in this type of concrete, the continuity of the particle size distribution is further improved by using a water/cement ratio well below the minimum required for complete hydration (0.42 [141,142]), which leaves a significant fraction of cement particles unhydrated. Thus, in this type of mix, cement is not only a source of hydrates. It behaves also as filler and it occupies a slot in the particle size distribution which otherwise would be empty. This is an efficient strategy in terms of strength, but it is not the best choice in terms of environmental impact. In addition, on a more fundamental level, it makes it not straightforward to use the binder intensity index as a direct quantitative indicator of the "quality" of the granular packing, at least in the high strength part of the graph where some correction should apply.

To close this section, let us come back to Féret's relationship or its quasi-equivalents. As far as it is correctly describing the formulation-dependence of compressive strength, there is one obvious prediction that cannot be missed: provided one keeps the same cement and the same granular formulation, strength will not change as long as the c/(c + w + v) ratio is kept constant. In other words, to take extreme examples, the same concrete strength can be targeted either with a lot of cement and a lot of water and air, or alternatively with a little cement and a little water and air. This opens the way to a class of concrete with low cement content, as it has already been suggested, though on somewhat different basis [193]. It remains to establish how far can this logic can be pursuit. Féret's equation applies the best to ordinary and medium strength concretes, say up to 60 MPa. So, at least in this range is it worth to investigate further the mechanical properties of optimized granular matrices with very low binder content (and appropriate admixture). It may require the development of new admixtures and novel or ancient (back to Roman-type [194]) ways to produce cohesion.

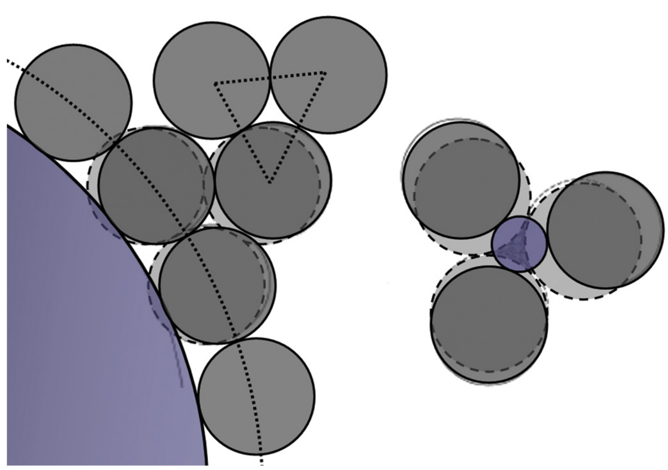

Fig. 7. Two perturbations occurring in a dense assembly of particles with at least two populations of particles. Left: the "wall effect". Close to a wall, like the surface of a formwork or the surface of a large particle for instance, the density of the packing of smaller particles is decreasing due to the forced switching from 3D (tetrahedral-like, with a local packing fraction of ~0.74) to pseudo-2D (hexagonal-like with a local packing fraction of ~0.66) arrangement. Right: the "spacing" effect. A particle larger than the central void forced to fit into a random close-packed arrangement is pushing the larger particles away.

4. Toward radical changes

In 2012, Robert Flatt, Nicolas Roussel and Christopher Cheeseman published a paper entitled "*Concrete*: *An eco-material that needs to be improved*" [193]. This title is an excellent, though somewhat optimistic, condensate of the situation that concrete is facing today. On a unit mass or volume basis, concrete is indeed a reasonably good eco-material in spite of the use of Portland cement or its blended versions [4], thanks to its large content in aggregates. Its important environmental footprint comes from the huge and still increasing volumes in which it is used. If we succeed in switching to fossil fuel-free transportation and if we find enough resources to transform our built dwellings heritage into zero-energy constructions, concrete may well become the major contributor to greenhouse gas emission in the future [4]. Pursuing our search for less CO_2– and energy-intensive binders [184,195–197] and our efforts for recycling aggregates including sand (or, at least, not using sand from fragile natural settings like river beds, beaches, or continental shelves), will definitely improve the carbon footprint of concrete and its use of natural resources, and it could improve the image and the societal acceptance of concrete. However, it will not solve the huge financial challenge that the renewal or the construction of infrastructure is raising on a global scale, in part due to the notoriously low productivity of the construction industry [198–201].

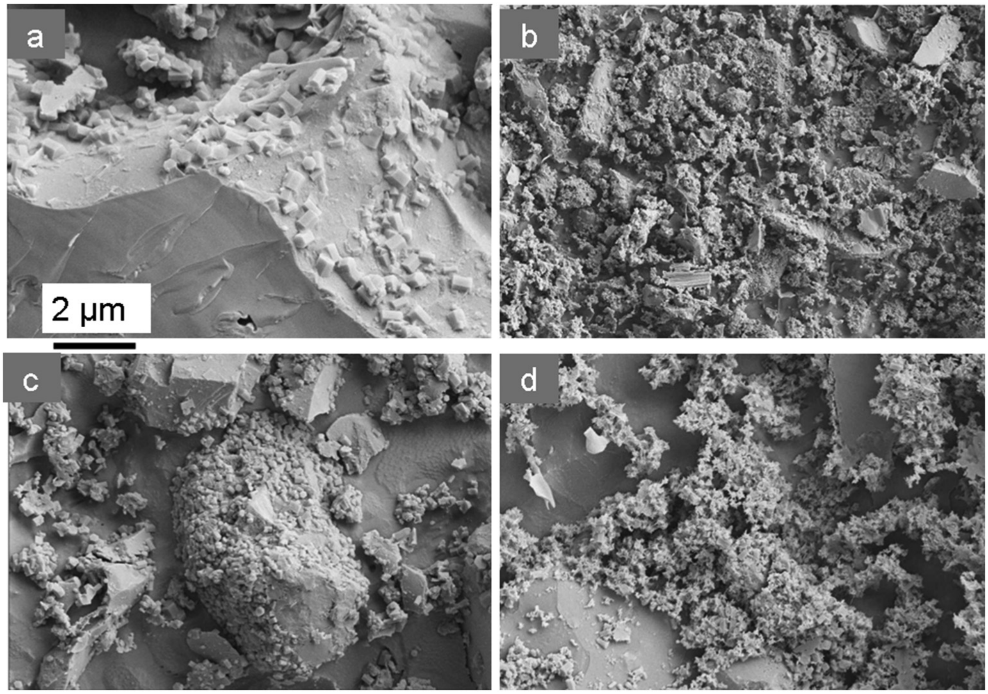

Fig. 8. SEM images of cement pastes hydrated without (a and c) or with (b and d) polymer-stabilized C-S-H seeds. Micrographs (a) and (b) were taken after the same hydration time (1 h), whereas (c) and (d) were taken at the hydration degree α reached after 6H20' for paste (c) and after 3 h20′ for paste (d). The first noticeable effect of seed addition is a faster hydrate precipitation (there is more C-S-H in (b) than in (a)). Another effect is a more homogeneous distribution of C-S-H which is present in the whole capillary void space in (b) and (d), and only on the surface of the cement particles in (a) and (c).
Courtesy Luc Nicoleau [168].

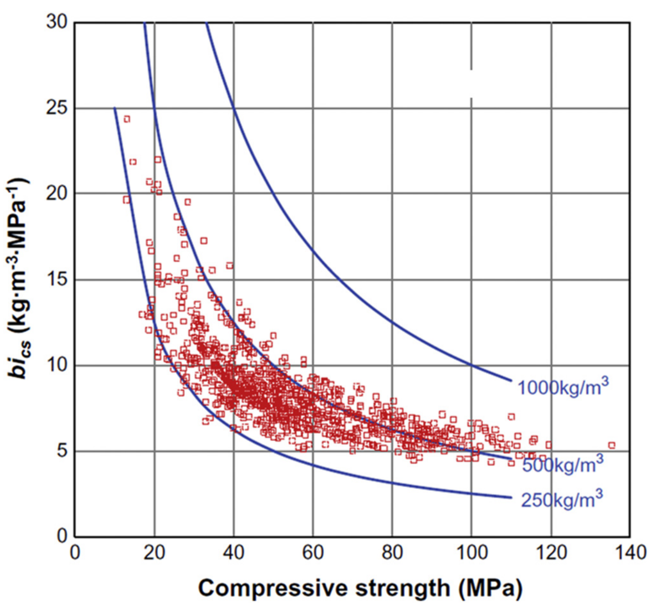

Fig. 9. Binder intensity index as a function of 28 days strength for close to one thousand concretes of international origin.
(From Damineli et al. [192]).

This paper, together with the other contributions in this issue, calls for the exploration of radical science- and technology-based changes in our construction practice with concrete. Most important is the need to abandon a purely material-centred approach of concrete and return to the integrated system-centred approach of the pioneers, taking in particular advantage of the opportunities offered by digital and unconventional construction technologies.

Two priority objectives have been identified in this paper. The first, with the purpose of improved durability, is the need to reinvent reinforcement and in particular to abandon or, at the least, minimize passive reinforcement with steel rebars. Going in this direction could also significantly improve productivity. The second objective, with the purpose of improved strength, workability, durability, and carbon footprint is the optimization of the granular formulation and, concomitantly, the decrease of the binder content.

4.1. Toward rebar-free concrete

Conventional reinforcement – especially passive reinforcement – has many advantages. It is inexpensive and robust technology. It is easy to build on site, with a minimum of training. It provides tensile strength, ductility, and crack growth resistance. Whatever the future cement and concrete technologies, the need for reinforcement will probably not disappear soon. This being said, the first question to address in the context of this special issue of CCR is whether the currently experimented digital or robotic fabrication technologies with concrete (Smart Dynamic Casting, Mesh-Mould, 3D-printing, binder jetting, etc.) are compatible with reinforcement, either passive or active. This question is addressed by Asprone et al. elsewhere in this issue [202]. The answer is that several techniques are indeed suitable for incorporating reinforcement either in its traditional form (Smart Dynamic Casting) or in near-traditional form (light robotically fabricated meshes in Mesh-Mould, or direct in-print entrainment of a continuous reinforcement cable in 3D-printing [202], reminiscent of continuous wire reinforcement for jammed dry granular architecture [203]). The printability of fiber-reinforced concrete has also been demonstrated [202]. Adding external reinforcement after the digital fabrication step is another option [202].

The approach adopted in this paper is slightly different. The question addressed is whether digital technologies may help in decreasing the need for (passive) reinforcement or in discovering new steel-free reinforcement methods. A preliminary comment is that robotic fabrication is not the only way for digital technologies to introduce innovations in construction with concrete. Innovations may also come from digital design methods. For instance, digital methods may indeed help in designing compression only structures or structures with minimal levels of tension, with much reduced reinforcement needs. This point is addressed in Asprone's et al. and in Block's contributions to this issue [202,204], but we will also add some comments later in this section.

We will start our search for new reinforcement methods by looking at what *autonomous assembly* may bring forth. The concept of

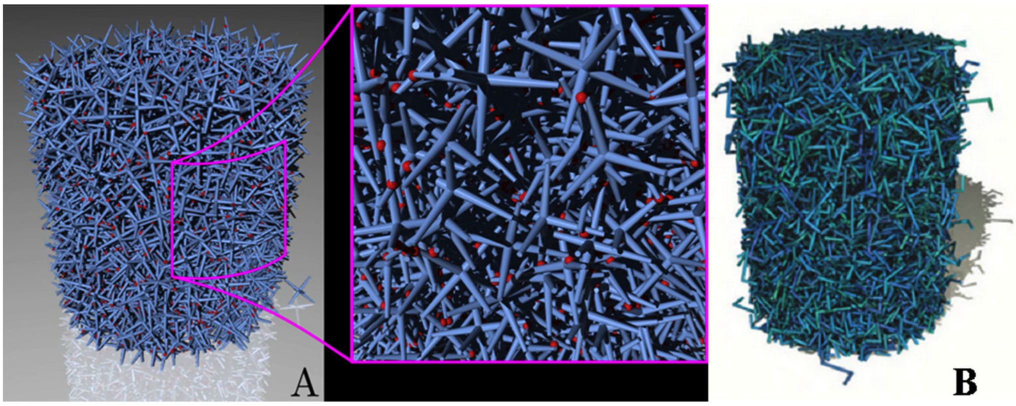

Fig. 10. Entangled assemblages of non-convex particles which could provide self-assembled reinforcement for concrete: (a) hexapods (six arms star-shaped) [218], the red dots indicate contact spots; (b) Z-shaped particles (credit Kieran Murphy, Leah Roth, Heinrich Jaeger, Project Z-Form). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 11. Free-form architecture with UHPFRC concrete: (A) Curved panels of the Louis Vuiton Foudation building in Paris, architect Frank Gehry; (B) The "fishnet" façade of the Mucem museum in Marseille, architect Rudy Ricciotti.

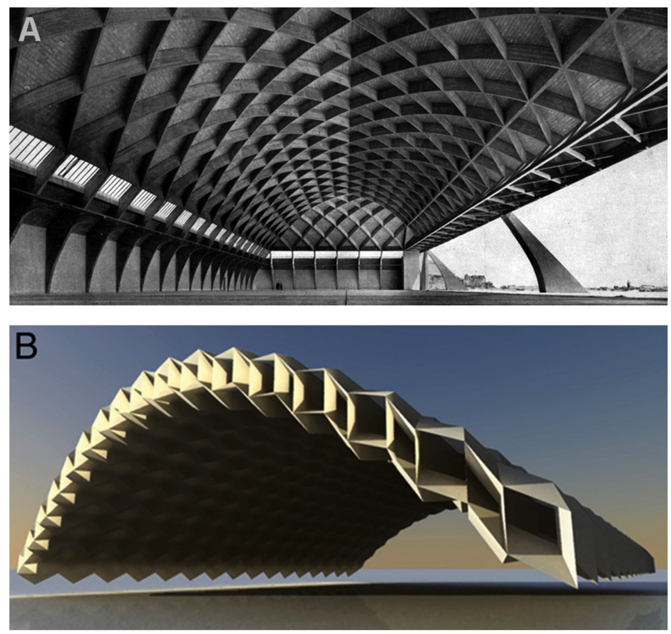

Fig. 12. (A) Aircraft hangar, Orvieto, architect-engineer Pier Luigi Nervi, 1935; (B) A computer-generated tubular origami-patterned architectural canopy illustrating its possible use as formwork. Adapted from refs. [234] and [236].

autonomous assembly may be viewed as a generalization of the concept of self-organization or self-assembly which has been widely popularized as a key concept in our understanding of life and many other natural phenomena [205]. It focuses on autonomy, the ability of materials, components (machines, humans, robots) or even processes to come together independently and have agency [206]. It is currently intensively explored in the context of fabrication, architecture, or structural engineering under various names like "designer matter" [207], "aleatory architecture" [208], or "aggregate architecture" [209]. A common key underlying idea is that, with properly designed building blocks, functionality may emerge from the interaction of a large number of building blocks during the assembly process which, in the simplest case, may be totally random. Disorder, mechanical instabilities and geometric nonlinearities are no longer considered as problems but as new opportunities [207].

Jammed assemblies of non-convex particles represent a particularly simple example of this type of system. For instance, contrary to the stadium of the 2008 Olympics ubiquitously referred to as the 'Bird's Nest', which owes its shape and stability to massive pieces of steel bound together [208,210], natural bird nests show solid-like behavior with elasticity and cohesion by the unique virtue of entanglement of independent twigs [211,212]. As another example, fire ants form remarkably strong viscoelastic aggregates (rafts, bridges, bivouacs) of 100,000 and more individuals, just by interlocking limbs and mandibles [208,213,214]. This contrasts with the behavior of ordinary non-cohesive and convex particles (dry sand for instance) which exhibit solid-like behavior only when they reach the jamming limit by confinement [215]. In frictionless, monodisperse spheres the rigid, jammed state is reached at a packing density corresponding to the rcp limit, $\phi_J = \phi_{rcp} \cong 0.64$. Even at such a high density, frictionless spheres can be sheared past one another without dilation of the packing. With friction,

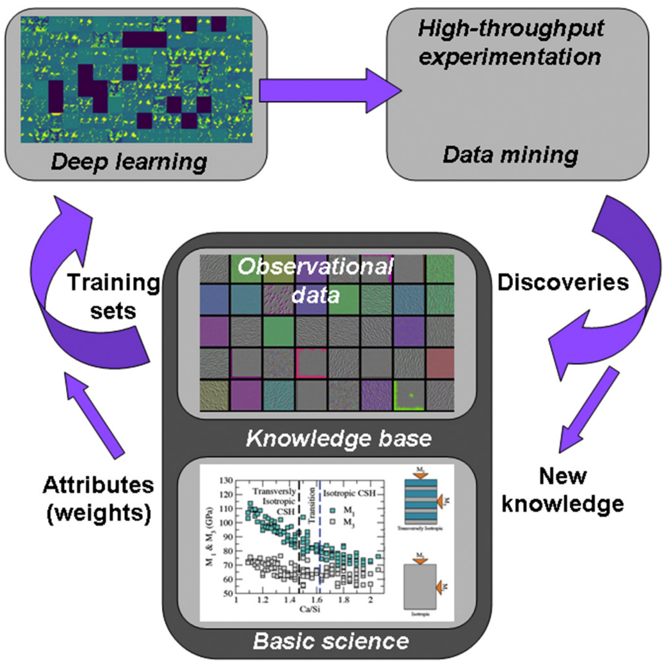

Fig. 13. Cartoon depicting a paradigm for transformative evolution of research on concrete through massive data collection and deep learning fed with basic research in physics and chemistry.
Adapted from ref. [257].

mechanically stable packings of spheres can exist in a range of densities extending to ~0.55 (remember Bernal's and Scott's experiments [84,85,87]), but shearing requires some dilation. In bird nests and ant aggregates, the same state of jamming is reached by "geometric cohesion" or "self-confinement" at much lower densities [212].

On the experimental side, U-shaped [211], *Z*-shaped [216], and star-shaped [217,218] particles as well as flexible chains of beads [219] have been shown to exhibit strong self-confinement (Fig. 10). They make it possible to create freestanding walls and columns, or even overhangs and domes, by simply pouring the particles into a mould and removing the mould as soon as it is full [216]. Alternatively, robotic arms may be used to grasp handfuls of particles and to build the structural element without the need of mould [217]. By changing the number of segments, the segment length and the angles between segments, the degree of entanglement may be fine-tuned. Just like the vein-like network of force chains that propagate compressive stresses in confined packings of convex particles, a minority of particles tugging their neighbors inwards propagate tensile stresses in assemblies of non-convex particles [216]. The same morphological parameters (number, length, angle of segments) control the degree of reversibility of the structure, that is, the ability to disentangle the aggregate [220,221]. Reversibility, together with the very open structure of the aggregates, is a very attractive feature for lightweight, reconfigurable architecture.

Here we suggest that random aggregates of non-convex particles provide also an interesting alternative to traditional passive reinforcement. For instance, synthetic particles made of UHPFRC by robotic fabrication could be randomly poured in the formwork and self-entangle. A self-placing concrete matrix could then be poured in the formwork. Alternatively, the formwork could be removed before pouring the concrete matrix and the entangled packing could be filled with an appropriate concrete mix, as in the mesh-mould technique briefly described in Section 2 [67–69] and more extensively elsewhere in this issue [70]. More work would definitely be needed to find the particle shape corresponding to the best trade-off between flowability and entanglement, but this should not be an issue. Optimizers using evolutionary algorithms have already been successfully applied to similar problems with compound particles comprised of bonded spheres, like finding which particle shape forms the densest random packing when poured under gravity [222], or finding the shape that leads to maximum stiffness or softness under compressive loading [223].

Autonomous assembly of non-convex aggregates is just one possible innovation among the many that could probably be imported in concrete technology from the physics of granular media. Another possible example is the use of vacuum-induced jamming to build re-configurable free-form formworks [224,225]. 3D-printing is currently the most straightforward way to get rid of formworks and to explore the world of free forms, but this doesn't prevent from looking for alternative and effective ways to build complex shapes. Vacuumatics – a term coined by J. Gilbert and coworkers (quoted in ref. [225]) – is a method inspired from the traditional sand casting technique used in the metal manufacturing industry. The granular material is first enclosed in a flexible membrane. In this state, it can be shaped in any form. Under moderate vacuum (moderate confinement), it acquires plastic properties, which still allows for shaping while providing enough yield stress to keep the newly given shape, like with modeling clay. Under stronger vacuum (strong confinement), the material becomes fully rigid (like a bag of ground coffee packed under vacuum) and is now able to accept the load of the poured concrete. After use, the system is fully reconfigurable if needed.

The design of free-form complex shapes is not just an aesthetic issue. It is also potentially a way to reach minimal tensile stresses and to minimize the need for reinforcement. Nature is full of complex, often hierarchical structures which succeed in optimizing simultaneously mechanical, optical, hydrodynamic, etc. properties and economy of material use. They are a fountain of inspiration for architects and engineers [226–228]. Typical examples are the cellular structure of trabecular bone, the hierarchical structure of nacre, or the ribbed structure of some large leaves which has been an obvious source of inspiration for Pier Luigi Nervi for his amazingly thin floors and roofs which owe their stiffness to their ribbed structure. What nature achieved through natural evolution, engineers may try to accomplish through *topology optimization*. Topology optimization is a mathematical method that optimizes material layout within a given design space, for a given set of loads, boundary conditions and constraints [229,230]. The design can attain any shape within the design space. Among the constraints, an upper limit to tensile efforts may be included. For instance, the maximum stiffness of a floor structure may be sought for, with a given amount of concrete and the minimal amount of reinforcement. Topology optimization is usually implemented in a finite element context assuming linear elastic material behavior, but more sophisticated models may be used.

Due to the free forms that naturally emerge from this, the result is often difficult to build or to manufacture. Vacuumatic formworks may help if 3D-printing is not available, but recent developments suggest that transformational progress may soon come from the convergence of two relatively recent theoretical research areas. One is the area now called *Architectural Geometry* [231,232]. The other is the science of paper folding or, more precisely, the science of deployable origami-patterned structures [233,234]. The two ingredients of architectural geometry are discrete differential geometry [235] and numerical optimization. It aims at transforming ideal free-form surfaces into buildable structures. This often means panelization, i.e. finding a collection of smaller flat or curved polyhedral elements that can be assembled to meshes with planar or curved faces and, desirably, with torsion-free nodes in the support structure. This last criterion makes planar quadrilateral meshes preferable to triangle meshes, with less degrees of freedom though. Another advantage of quad meshes is that they allow multiplayer construction. As far as the panel and support structure materials are concerned, steel-and-glass or the wood-and-wood are the most frequently encountered associations, but UHPFRC is also perfectly adapted to free-form architecture (Fig. 11).

On the other hand, *origami*, the art of folding paper, has recently emerged as a method for creating deployable and reconfigurable structures [236]. "Origamics" [233] is at the center of an intense exploration activity for (possible) applications as diverse as solar arrays [237], deployable curtain walls for light control [234], self-foldable robots [238], or self-deployable stent grafts [239], just to name a few. It is a common experience that folding makes sheets more rigid. However, folded sheets are not yet structures [240]. They tend to keep some flexibility and because folding is reversible they need to be locked into a fixed configuration in order to behave as a real structure [236,240]. Therefore, conferring load-bearing characteristics to a deployable origami-patterned structure is a tricky business, but it has been achieved at least once by coupling origami tubes in a zipper-like fashion [236].

Origami patterning is an interesting method for conferring dynamic properties to building facades (optimizing shading for instance), but it is also an interesting approach for manufacturing formworks able to cast complex shapes, with potentially much less labor than with traditional formworks. What is suggested here is that topology optimization, architectural geometry, and origami patterning may well become, on equal foot with printing, privileged methods for building optimally-designed structures in concrete, using reconfigurable formworks (Fig. 12).

Finally, an important question remains. Is it realistic to consider that we may soon be able to improve the bending and tensile strength, the elastic limit and modulus, and the total fracture energy of hardened cement paste to a point such that reinforcement would no longer be needed, even in ordinary concrete? Improving the mechanical properties of cement or cement hydrates by incorporating polymers has been a permanent quest of chemists for more than forty years. Often nature-inspired, this strategy has led to remarkable results. With calcium-aluminate cement and PVA, it led to the development of the so-called macro-defect-free (MDF) composite cement with a flexural strength up to 70 MPa [241]. Recently, highly oriented mesocrystalline deposits of C-S-H could be prepared by careful destabilization of an initially fully dispersed suspension of C-S-H nanoparticles ($\sim 60 \times 30 \times 5\,nm^3$) [242]. Total dispersion was obtained using copolymer dispersants selected from a phage display assay [243]. During deposition, from ~5 to ~20% (wt%) of polymer are incorporated in the material. Tests performed on micro-cantilevers milled from these deposits revealed an amazing bending strength approaching 200 MPa, which is close to that of nacre and outperforms the flexural strength of ordinary concrete by a factor of 40 to 100 [243]. Unfortunately, fracture remains brittle and the strain at fracture is. In spite of the scientific beauty of this nanochemical feat, enormous progress is clearly still needed before both strength, ductility, could be reached with polymer contents compatible with material availability (10 wt% polymer with respect to cement would lead to a doubling of the global polymer use), not considering cost issues.

4.2. Less cement, better concrete

Beyond its light provocative character, this heading points to a very serious point: cement (more exactly, clinker) and other hydraulic or pozzolanic materials are currently far from being used at their best level, even in (ultra)high-performance concrete. In spite of the relatively large "mechanical yield" of cement in this type of concrete (number of MPa of compressive strength/mass of cement per unit volume, of the order of $20\,MPa/kg{\cdot}m^{-3}$ in (U)HPC, Fig. 9), less than ~40% of the tri- and di-calcium silicates have been hydrated [183]. When the pozzolanic silica fume is included in the calculation, the figure goes down to less than 20%. As discussed in Section 3, this leads to the counter-intuitive conclusion that in the best performing cement-based materials, cement and reactive SCM are primarily used as inert fillers, and only secondarily for their hydrate-forming capacity. The relatively inexpensive character of cement is partly responsible for this, but in rational terms, it is a highly questionable choice. Optimization of the particle packing over the whole particle size distribution with, concomitantly, optimal use of the adhesive and space-filling properties of hydrates is – as far as we can do it – a preferable choice.

In spite of conceptual and methodological progress (as skimmed through in Section 3), concrete formulation with a given set of target properties (workability, structural build-up, hardening kinetics, ultimate strength, creep, etc.) remains a difficult task, due in part to the increasing complexity of concrete compositions and that of cement itself. Research in basic science (thermodynamics, kinetics, micromecanics), in molecular and coarse grain modeling, and in semi-empirical experimental-theoretical proportioning methods has undoubtedly to be continued but, considering the difficulty of the task, one may wonder whether a parallel route using the recent advances in machine learning, together with high-throughput experimentation, should not be explored. Within the limited landscape of the 4-component ordinary concrete (gravel, sand, Portland cement, water), the compressive strength is usually predicted using classical statistical (or probabilistic modeling) methods like linear or non-linear regression, but these methods prove to be unsuitable to cope with the complexity of higher performance concrete. To compensate for these drawbacks machine learning (ML) algorithms have been introduced, not only for proportioning and strength prediction (for a short selection, see [244–253]), but also for a wide variety of civil engineering problems [254] (an extended literature bibliography of this field is beyond the scope of this paper).

Machine learning (ML) is basically different from the early versions of artificial intelligence (AI) [255,256]. In these early versions (symbolic AI, expert systems), the programmer introduces a large set of rules (a program). The system is fed with data to be processed according to these rules and the computer generates answers. This is a suitable method to solve logical problems. In ML, a system is trained rather than programmed. It is given a large set of examples (data and answers) relevant to a given task (recognize cats in a large number of images for instance) and it finds the statistical structure (the rules) underlying these examples. Eventually, it allows the system to come up with rules for automating the task. At the core of the learning process is the transformation of the data (inputs) into successively more meaningful representations called layers. The data transformation from one layer to the next one occurs through simple operations chosen among a predefined set. Each operation is characterized by a weight. Learning means finding a set of values for the weights of all layers in the algorithm.

The simplest ML methods or *Shallow Learning* (early neural networks; kernel methods like the *Support Vector Machine*; Decision trees like the *Random Forest* algorithm; etc.) are using a very limited number of layers and weights and the layers are trained in succession. So far, all ML techniques applied to concrete proportioning and strength prediction belong to this category, as far as we are aware. On the other hand, all the spectacular advances made in speech and image recognition or in autonomous driving in the last few years are all related to *deep learning* [255,256]. Deep learning is characterized by the large number of layers in the input-to-target mapping process, the large number of weights, up to tens of millions, and the fact that all layers are trained jointly. This gives deep learning systems their remarkable learning capability, but very large data sets of data are necessary for efficient training.

This is where the coupling with high-throughput experimentation and with data harvesting at the global scale comes in. The combinatorial space of the cement-SCM-filler-aggregate-admixture-water-curing-ageing system is virtually infinite and cannot be explored completely in a single laboratory or consortium. But millions of concrete mixes are prepared daily all over the world and could – provided a well-defined protocol is established – deliver millions of useful data sets. By complementing this with high-throughput experimentation in a network of selected laboratories, an unprecedented big data-type amount of information could be collected, covering a parameter space which could go well beyond simple proportioning ratios and include, on the input side (i) cement composition and crystal chemistry data; (ii) cement, fines, filler and SCM nature, particle size distribution, morphology through 3D imaging, and surface electrochemical data; (iii) admixture and water data; (iv) physical parameters (T, mixing method and energy); (v) degree of hydration and composition of the hydrate assemblage; (vi) curing and ageing conditions, etc. On the property (output) side, as many useful mechanical and microstructural data as possible could be recorded. All the input and output boxes of such an extensive combinatorial space will obviously never be filled, but this doesn't matter. By feeding them to deep learning algorithms guided by a strong physical and chemical basis, a transformative new optimization paradigm may emerge which would allow us to use each constituent of concrete, including cement, at its best, for each particular application (Fig. 13). We might call this *digital concrete*.

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References

[1] U.S. Geological Survey (USGS), Cement statistics and information, https://minerals.usgs.gov/minerals/pubs/commodity/cement/.
[2] CEMBUREAU, Activity report, https://cembureau.eu/media/1635/activity-report-2016.pdf, (2016).
[3] G.M. Idorn, Concrete Progress From Antiquity to Third Millennium, Thomas Telford, London, 1997.
[4] K.L. Scrivener, V.M. John, E.M. Gartner, Eco-Efficient Cements: Potential, Economically Viable Solutions for a Low-CO_2, Cement-Based Materials Industry, United Nations Environment Program, 2016 (available on), www.unep.org.
[5] M.H. Ramage, H. Burridge, M. Busse-Wicher, G. Fereday, T. Reynolds, D.U. Shah, G. Wu, L. Yu, P. Fleming, D. Densley-Tingley, J. Allwood, P. Dupree, P.F. Linden, O. Scherman, The wood from the trees. The use of timber in construction, Renew. Sust. Energ. Rev. 68 (2017) 333–359.
[6] World Steel Association (WSA), Steel statistical yearbooks 1978 to 2016, www.worldsteel.org/steel-by-topic/statistics/steel-statistical-yearbook-.html.
[7] U.S. Geological Survey (USGS), Iron and steel statistics and information, https://minerals.usgs.gov/minerals/pubs/commodity/iron_&_steel/mcs-2018-feste.pdf.
[8] R. Geyer, J.R. Jambeck, K.L. Law, Production, use, and fate of all plastics ever made, Sci. Adv. 3 (2017) e1700782(Supplementary material), http://advances.sciencemag.org/cgi/content/full/3/7/e1700782/DC1.
[9] U.S. Geological Survey (USGS), Aluminium statistics and information, https://minerals.usgs.gov/minerals/pubs/commodity/aluminum/mcs-2018-alumi.pdf.
[10] J. Woetzel, S. Ram, J. Mischke, N. Garemo, S. Sankhe, A blueprint for addressing the global affordable housing challenge, McKinsey Global Institute Report, 2014 https://www.mckinsey.com/global-themes/urbanization/tackling-the-worlds-affordable-housing-challenge.
[11] R. Dobbs, H. Pohl, D.-Y. Lin, J. Mischke, N. Garemo, J. Hexter, S. Matzinger, R. Palter, R. Nanavatty, Infrastructure productivity: how to save 1 trillion USD a year, McKinsey Global Institute Report, 2013 https://www.mckinsey.com/industries/capital-projects-and-infrastructure/our-insights/infrastructure-productivity.
[12] http://www.bison-fute.gouv.fr/reseau-routier-public.html.
[13] http://www.planete-tp.com/ponts-en-france-r58.html.
[14] https://www.sncf-reseau.fr/fr/le-reseau.
[15] http://www.cetu.developpement-durable.gouv.fr/etat-recapitulatif-du-nombre-et-des-longueurs-de-a480.html.
[16] http://www.statistiques.developpement-durable.gouv.fr/lessentiel/ar/306/1168/assainissement-collecte-eaux-usees-pluviales.html.
[17] http://www.eaufrance.fr/comprendre/l-eau-potable-et-l-assainissement/distribution-d-eau-potable#ancre_chiffre.
[18] https://www.asn.fr/Informer/Dossiers-pedagogiques/La-surete-des-centrales-nucleaires/Le-parc-francais-des-centrales-nucleaires.
[19] ASCE's, Infrastructure report card, https://infrastructurereportcard.org, (2017).
[20] A. Picon, Architecture and technology: two centuries of creative tension, in: J.-L. Cohen, G. Martin MoellerJr. (Eds.), Liquid Stone. New Architecture in Concrete, Birkhäuser, Basel, 2006, pp. 8–19.
[21] J.-L. Cohen, G.M. Moeller, Introduction, in: J.-L. Cohen, G. Martin MoellerJr. (Eds.), Liquid Stone. New Architecture in Concrete, Birkhäuser, Basel, 2006, pp. 6–7.
[22] J.-L. Cohen, Modern architecture and the saga of concrete, in: J.-L. Cohen, G. Martin MoellerJr. (Eds.), Liquid Stone. New Architecture in Concrete, Birkhäuser, Basel, 2006, pp. 20–33.
[23] A. Torres, J. Brandt, K. Lear, J. Liu, A looming tragedy of the sand commons, Sci. Mag. 357 (6355) (2017) 970–971.
[24] UNEP Global Environmental Alert Service (GEAS), Sand, rarer than one thinks (downloadable on),, 2014. www.unep.org/geas, .
[25] A. Forty, The material without a history, in: J.-L. Cohen, G. Martin MoellerJr. (Eds.), Liquid Stone. New Architecture in Concrete, Birkhäuser, Basel, 2006, pp. 34–45.
[26] Frank Lloyd Wright, In the cause of architecture VII: the meaning of materials – concrete (1928), reprinted in, in: B.B. Pfeifer (Ed.), F.L. Wright, Collected Writings, Vol. 1 Rizzoli, New York, 1992, pp. 297–301.
[27] Peter Collins, Concrete. The Vision of a New Architecture, 2nd ed., McGill-Queen's University Press, Montreal & Kingston, 2004.
[28] C. Simonnet, Le Béton, Histoire d'un Matériau, Editions Parenthèses, Marseille, (2005).
[29] R. Courland, Concrete Planet: The Strange and Fascinating Story of the World's Most Common Man-Made Material, Prometheus Books, Amherst, N.Y, 2011.
[30] E. Freyssinet, L'amélioration des constructions en béton armé par l'introduction de deformations élastiques systématiques, Le Génie Civil, 15 sept 1928, pp. 254–257 no. 2405.
[31] H. Okamura, K. Ozawa, Mix-design for self-compacting concrete, Concrete Library of JSCE, 25 1995, pp. 107–120.
[32] P. Richard, M. Cheyrezy, Composition of reactive powder concrete, Cem. Concr. Res. 25 (1995) 1501–1511.
[33] B. Khoshnevis, Innovative rapid prototyping process making large sized, smooth surface complex shapes in a variety of materials, Mater. Technol. 13 (1998) 52–63.
[34] T. Wangler, E. Lloret, L. Reiter, N. Hack, F. Gramazio, M. Kohler, M. Bernhard, B. Dillenburger, J. Buchli, N. Roussel, R. Flatt, Digital concrete: opportunities and challenges, RILEM Technol. Lett. 1 (2016) 67–75.
[35] J. Gubler, Histoire du béton: naissance et développement, de 1818 à nos Jours, CIMbéton, Paris, downloadable on www.infociments.fr/telecharger/CT-B90A.pdf (Retrieved on 2018-02-26).
[36] M. Wells, Engineers: A History of Engineering and Structural Design, Rouledge, New York, 2010.
[37] M.J. Aldolhosseini Qomi, F.-J. Ulm, R.J.-M. Pellenq, Physical origins of thermal properties of cement paste, Phys. Rev. Appl. 3 (2015) 064010.
[38] B. Andreotti, Y. Forterre, O. Pouliquen, Granular Media. Between Fluid and Solid, Cambridge University Press, Cambridge, UK, 2013.
[39] O. Coussy, Mechanics and Physics of Porous Solids, Wiley, Chichester, UK, 2010.
[40] H. Le Chatelier, Recherches expérimentales sur la constitution des ciments et la théorie de leur prise, C. R. Acad. Sci. Paris 94 (1882) 867–869.
[41] H. Le Chatelier, Recherches Expérimentales sur la Constitution des Mortiers Hydrauliques, Dunod, Paris, 1904.
[42] A.C. Lassaga, A. Lüttge, Variation of crystal dissolution rate based on a dissolution stepwave model, Science 291 (2001) 2400–2404.
[43] P.M. Dove, N. Han, J.J. De Yoreo, Mechanisms of classical crystal growth theory explain quartz and silicate dissolution behaviour, PNAS 102 (2005) 15357–15362.
[44] W.A. Johnson, R.F. Mehl, Reaction kinetics in processes of nucleation and growth, Trans. Am. Inst. Min. (Metall.) Eng. 135 (1939) 416–442.
[45] J.W. Cahn, The kinetics of grain boundary nucleated reactions, Acta Metall. 4 (1956) 449–459.
[46] P. Juilland, E. Gallucci, R. Flatt, K.L. Scrivener, Dissolution theory applied to the induction period in alite hydration, Cem. Concr. Res. 40 (2010) 831–844.
[47] P. Juilland, E. Gallucci, Morpho-topological investigation of the mechanisms and kinetic regimes of alite dissolution, Cem. Concr. Res. 76 (2015) 180–191.
[48] L. Nicoleau, A. Nonat, D. Perrey, The di- and tricalcium silicate dissolutions, Cem. Concr. Res. 47 (2013) 14–30.
[49] S. Garrault-Gauffinet, A. Nonat, Experimental investigation of calcium silicate hydrate (C-S-H) nucleation, J. Cryst. Growth 200 (1999) 565–574.
[50] S. Garrault, A. Nonat, Hydrated layer formation on tricalcium and dicalcium silicate surfaces: experimental study and numerical simulations, Langmuir 17 (2001) 8131–8138.
[51] J.J. Thomas, A new approach to modeling the nucleation and growth kinetics of tricalcium silicate hydration, J. Am. Ceram. Soc. 90 (2007) 3282–3288.
[52] P.M. Dove, N. Han, Kinetics of mineral dissolution and growth as reciprocal microscopic surface processes across chemical driving force, in: M. Skowronski, J.J. De Yoreo, C.A. Wang (Eds.), Perspectives on Inorganic, Organic, and Biological Crystal Growth: From Fundamentals to Applications, 13th International Summer School on Crystal Growth, Amer. Inst. Phys. Conf. Proc. 916, 2007, pp. 215–234.
[53] E.A.M. Koutsoukos, The K-T boundary, chap. 7, in: E.A.M. Koutsoukos (Ed.), Applied Stratigraphy, Chap. 7, Springer, Berlin, 2005, pp. 147–161.
[54] H. Van Damme, H. Houben, Earth concrete: stablization revisited, Cem. Concr. Res. (2018), http://dx.doi.org/10.1016/j.cemconres.2017.02.035 (in press).
[55] L.J. Vicat, Recherches expérimentales sur les chaux de construction, les bétons et les mortiers ordinaires, Goujon, Paris, 1818.
[56] F. Coignet, Des bétons agglomérés appliqués à l'art de construire, Librairie scientifique, industrielle et agricole E. Lacroix, (1861) (378p).
[57] J.-L. Bosc, J.-M. Chauveau, J. Clément, J. Degenne, B. Marrey, M. Paulin, Joseph Monier et la naissance du ciment armé, Editions du Linteau, Paris, (2001).
[58] P. Dorveaux, Lambot-Miraval, Monier, et le ciment armé, La Maison Rustique,

Paris, extr. from La Revue Agricole, issue 17 and 18, may 1919.
[59] G.A. Wayss, Das system monier, in: G.A. Wayss (Ed.), In Seiner Anwendung auf das Gesamte Bauwesen, Seydel & Cie, Berlin, 1887.
[60] W.E. Ward, Beton in combination with iron as a building material, Trans. Am. Soc. Mech. Eng. 4 (1883) 388–404.
[61] T. Hyatt, An Account of some Experiments With Portland-Cement-Concrete Combined with Iron, as a Building Material, With Reference to Economy of Metal in Construction, and for Security Against Fire in the Making of Roofs, Floors and Walking Surfaces, Chickwick Press, London, 1877.
[62] C.W. Condit, The first reinforced concrete system: the Ingalls building in Cincinnati and its place in structural history, Technol. Cult. 9 (1968) 1–33.
[63] E. Freyssinet, Un amour sans limite, presentation by H. Lemoine and P. Xercavins, Notes by B. Marrey., Editions du Linteau, Paris, (1993).
[64] M. Bohnsdalen Eide, J.-M. Hisdal, Ultra High Performance Fibre Reinforced Concrete (UHPFRC) – State of the Art, SINTEF Building and Infrastructure, Oslo, 2012.
[65] J. Resplendino, F. Toulemonde (Eds.), Designing and Building With UHPFRC, Wiley-ISTE, 2011.
[66] C. Greco, The "Ferro-Cemento" of Pier Luigi Nervi. The new material and the first experimental building, Spatial Structures: Heritage, Present and Future, SGD Editorial, Milano, 1995, pp. 309–316.
[67] N. Hack, W.V. Lauer, Mesh-Mould: robotically fabricated spatial meshes as reinforced concrete formwork, Archit. Des. 84 (3) (2014) 44–53.
[68] N. Hack, W.V. Lauer, F. Gramazio, M. Kohler, Mesh Mould: robotically fabricated metal mashes as concrete formwork and reinforcement, in: W. Brameshuber (Ed.), FERRO-11, Proceedings of the 11th Int. Symp. on Ferrocement and 3rd Int. Conf. on Textile Reinforced Concrete, RILEM Pub. SARL, Aachen, Germany, 2015, pp. 347–359.
[69] N. Hack, T. Wangler, J. Mata-Falcon, K. Dörfler, N. Kumar, A.N. Walzer, K. Graser, L. Reiter, H. Richner, J. Buchli, W. Kaufmann, R.J. Flatt, F. Gramazio, M. Kohler, Mesh Mould: an on site, robotically fabricated, functional formwork, in: Hammer Tor Arne (Ed.), Proceedings of the Eleventh High Performance Concrete (11th HPC) and Second Concrete Innovation Conference (2nd CIC), Norwegian Concrete Association/Tecna, Tromso, Norway, 2017(paper no. 19).
[70] J. Buchli, M. Giftthaler, N. Kumar, M. Lussi, T. Sandy, K. Dörfler, N. Hack, Digital *in situ* fabrication, Cem. Concr. Res. (2018) (this special issue).
[71] J.F. Young, Looking ahead from the past: the heritage of cement chemistry, Cem. Concr. Res. 38 (2008) 111–114.
[72] J.P. Broomfield, Corrosion of Steel in Concrete: Understanding, Investigation, and Repair, 2nd ed., CRC Press, 2006.
[73] L. Bertolini, B. Elsener, P. Pedeferri, R. Polder, Corrosion of Steel in Concrete: Prevention, Diagnosis, Repair, Wiley, Weinheim (Germany), 2004.
[74] S. Zofia, Z. Adam, Theoretical model and experimental tests on chloride diffusion and migration processes in concrete, Proc. Eng. 57 (2013) 1121–1130.
[75] S.H. Jung, M.K. Lee, B.H. Oh, Measurement device and characteristics of diffusion coefficient of carbon dioxide in concrete, Amer. Concr. Inst. Mater. J. (2011) 589–595 (Nov-Dec 2011, Title no. 108-M62).
[76] C. Pade, M. Guimaraes, The CO_2 uptake of concrete in a 100-year perspective, Cem. Concr. Res. 37 (2007) 1348–1356.
[77] C.P. Vernet, Ultra-durable concretes: structure at the micro- and nanoscale, Mater. Res. Soc. Bull. 29 (2004) 324–327.
[78] G. Gaber, Corrosion Protection of Reinforcing Steel in Concrete, Scholar's Press, 2017.
[79] T. Aste, D. Weaire, The Pursuit of Perfect Packing, I.O.P. Pub, Bristol and Philadelphia, 2000.
[80] A. Brown, J.D. Bernal, The Sage of Science? Oxford University Press, Oxford, 2005.
[81] J.D. Bernal, J.W. Jeffery, H.F.W. Taylor, Crystallographic research on the hydration of Portland cement. A first report on investigations in progress, Mag. Concr. Res. 4 (1952) (1952) 49–54.
[82] J.L. Finney, Bernal's road to random packing and the structure of liquids, Philos. Mag. 93 (2013) 3940–3969.
[83] J.D. Bernal, J. Mason, Co-ordination of randomly packed spheres, Nature 188 (1960) 910–911.
[84] J.D. Bernal, J.L. Finney, Random packing of spheres in non-rigid containers, Nature 214 (1967) 265–266.
[85] G.D. Scott, D.M. Kilgour, The density of random close packing of spheres, Brit. J. Appl. Phys. Ser. 2 (1969) 863–866.
[86] S. Torquato, F.H. Stillinger, Jammed hard-particle packings: from Kepler to Bernal and beyond, Rev. Mod. Phys. 82 (2010) 2633–2672.
[87] G.D. Scott, Packing of spheres, Nature 188 (1960) 908–909.
[88] G.Y. Onoda, E.G. Linger, Random loose packing of uniform spheres and the dilatancy onset, Phys. Rev. Lett. 64 (1990) 2727–2730.
[89] M. Borkovec, W. De Paris, R. Peikert, The fractal dimension of the apollonian sphere packing, Fractals 2 (1994) 521–526.
[90] G.W. Delaney, S. Hutzler, T. Aste, Relation between grain shape and fractal properties in random apollonian packing with grain rotation, Phys. Rev. Lett. 101 (2008) 120602.
[91] R.S. Farr, London Institute for Mathematical Sciences, www.london-institute.org/people/farr/packing.html.
[92] B.B. Mandelbrot, Les objets fractals, Herrmann, Paris, 1975.
[93] B.B. Mandelbrot, The Fractal Geometry of Nature, Freeman, New York, 1977.
[94] S.S. Manna, H.J. Herrmann, Precise determination of the fractal dimensions of Apollonian packing and space-filling bearings, J. Phys. A Math. Gen. 24 (1991) L481–L490.
[95] R.S. Farr, E. Griffiths, Estimate for the fractal dimension of the Apollonian gasket in *d* dimensions, Phys. Rev. E 81 (2010) (061403-1 to 061403-4).
[96] A.N. Talbot, F.E. Richart, The strength of concrete: its relation to the cement, aggregate and water, Bull. of the Eng. Exp. Station, University of Illinois, Urbana, IL, 1923, p. 137 Bull.
[97] S. Walker, Modulus of elasticity of concrete, Bull. of the Struct. Mater. Res. Lab. Lewis Institute, Chicago, IL, 1923, p. 5 Bull.
[98] M. Leclerc du Sablon, M., Le béton rationnel. Méthodes Pratiques pour la Réalisation de Mortiers et de Bétons Offrant les Qualités Désirées au Prix de Revient Minimum. Annales des Ponts et Chaussées, 97, Tome 1, Fasc, 1 (1927), p. 149.
[99] R. Dutron, Dosage Rationnel des Mortiers et Des Bétons, Revue des Matériaux de Construction et des Travaux Publics, 216 (1927) 304–306; 217 (1927) 329–333; 218 (1927) 374–377, 219 (1927), pp. 406–414.
[100] J. Bolomey, Granulation et prévision de la résistance probable des bétons, Travaux 19 (30) (1935) 228–232.
[101] A.I. Caquot, Le rôle des Matériaux inertes dans le béton, Mémoire de la Société des Ingénieurs Civils de France, July-August issue, Fasc. 4 (1937), pp. 562–582.
[102] K. Sobolev, M. Ferrada Gutiérrez, How nanotechnology can change the concrete world, Am. Ceram. Soc. Bull. 84 (2005) 14–18.
[103] J. Baron, Compacité des mélanges de grande étendue granulaire, in: J. Baron, J.-P. Ollivier (Eds.), Les Bétons: Bases et Données Pour Leur Formulation, Eyrolles, Paris, 2002.
[104] A.H.M. Andreasen, J. Andersen, Ueber die beziehung zwischen kornabstufung und zwischenraum in produkten aus losen köenern (mit einigen experimenten), Kolloid Zeitschrift 50 (1930) 217–228.
[105] X. Chateau, Particle packing and the rheology of concrete, in: N. Roussel (Ed.), Understanding the Rheology of Concrete, Woodhead Publishing, Cambridge, 2012, pp. 117–143.
[106] H.J.H. Browers, Particle-size distribution and packing fraction of geometric random packings, Phys. Rev. E 74 (2006) 031309.
[107] C.C. Furnas, Grading aggregates: I. Mathematical relations for beds of broken solids of maximum density, Ind. Eng. Chem. 23 (1931) 1528–1958.
[108] N. Ouchiyama, T. Tanaka, Porosity estimation from particle size distribution, Ind. Eng. Chem. Fundam. 25 (1986) 125–129.
[109] S. Liu, Z. Ha, Prediction of random packing limit for multimodal particle mixtures, Powder Technol. 126 (2002) 283–296.
[110] F. de Larrard, T. Sedran, Optimization of ultra-high-performance concrete by the use of a packing model, Cem. Concr. Res. 24 (1994) 997–1009.
[111] F. de Larrard, T. Sedran, Mixture-proportioning of high-performance concrete, Cem. Concr. Res. 32 (2002) 1699–1704.
[112] F. de Larrard, Concrete Mixture Proportioning: A Scientific Approach, Modern Concrete Technology Series, No. 9 E & FN SPON, London, 1999.
[113] J. Mewis, N.J. Wagner, Colloidal Suspension Rheology, Cambridge Univ. Press, Cambridge, 2012.
[114] P. Coussot, Rheometry of Pastes, Suspensions, and Granular Materials: Applications in Industry and Environment, Wiley-Interscience, 2005.
[115] N. Roussel (Ed.), Understanding the Rheology of Concrete, Woodhead Publishing, Cambridge, 2012.
[116] V.G. Kolli, E.J. Pollauf, F. Gadala-Maria, Transient normal stress response in a concentrated suspension of spherical particles, J. Rheol. 46 (2002) 321–334.
[117] N.Y.C. Lin, B.M. Guy, M. Hermes, C. Ness, J. Sun, W.C.K. Poon, I. Cohen, Hydrodynamic and contact contributions to continuous shear thickening in colloidal suspensions, Phys. Rev. Lett. 115 (2015) 228304.
[118] D. Lootens, P. Hébraud, E. Lécolier, H. Van Damme, Gelation, shear-thinning and shear-thickening in cement slurries, Oil & Gas Tech. – Rev. IFP 59 (1) (2004) 31–40.
[119] D. Lootens, H. Van Damme, Y. Hémar, P. Hébraud, Dilatant flow of concentrated suspensions of rough particles, Phys. Rev. Lett. 95 (2005) 268302.
[120] M.M. Denn, J.F. Morris, D. Bonn, Shear thickening in concentrated suspensions of smooth spheres in Newtonian suspending fluids, Soft Matter 14 (2018) 170–184.
[121] N. Roussel, A. Lemaître, R.J. Flatt, P. Coussot, Steady state flow of cement suspensions: a micromechanical state of the art, Cem. Concr. Res. 40 (2010) 77–84.
[122] N. Roussel, Rheological requirements for printable concrete, Cem. Concr. Res. (2018) (this special issue).
[123] I.M. Krieger, T.J. Dougherty, A mechanism for non-Newtonian flow in suspensions of rigid spheres, Trans. Soc. Rheol. 3 (1959) 137–152.
[124] A. Einstein, Eine neue bestimmung der moleküldimensionen, Ann. Phys. 19 (2006) 289–306.
[125] H. Okamura, M. Ouchi, Self-compacting concrete, J. Adv. Concr. Technol. 1 (2003) 5–15.
[126] A. Yahia, P.-C. Aïtcin, Self-consolidating concrete, in: P.-C. Aïtcin, R. Flatt (Eds.), Science and Technology of Concrete Admixtures, Woodhead Publishing, Amsterdam, 2016.
[127] H.J.H. Browers, H.J. Radix, Self-compacting concrete: the role of the particle size distribution, First Int. Symp. on Design, Performance and Use of Self-Consolidating Concrete, SCC, 2005.
[128] C. Shi, Z. Wu, K. Lv, L. Wu, A review on mixture design methods for self-compacting concrete, Constr. Build. Mater. 84 (2015) 387–398.
[129] R. Yu, P. Spiesz, H.J.H. Brouwers, Mix design and properties assessment of ultra-high performance fibre reinforced concrete (UHPFRC), Cem. Concr. Res. 56 (2014) 29–39.
[130] R. Flatt, Conclusions and outlook on the future of concrete admixtures, in: P.-C. Aïtcin, R. Flatt (Eds.), Science and Technology of Concrete Admixtures, Woodhead Publishing, Amsterdam, 2016, pp. 527–530.
[131] P.-C. Aïtcin, R. Flatt (Eds.), Science and Technology of Concrete Admixtures, Woodhead Publishing, Amsterdam, 2016.
[132] R.C. Sonntag, W.B. Russel, Structure and breakup of flocs subjected to fluid

stresses, J. Colloid Interface Sci. 115 (1987) 378–389.
[133] P. Mills, P. Snabre, The fractal concept in the rheology of concentrated suspensions, Rheol. Acta 26 (1988) 105–108.
[134] R.J. Flatt, P. Bowen, Yodel: a yield stress model for suspensions, J. Amer. Ceram. Soc. 89 (4) (2006) 1244–1256.
[135] G. Gelardi, S. Mantellato, D. Marchon, M. Palacios, A.B. Eberhardt, R.J. Flatt, Chemistry of chemical admixtures, in: P.-C. Aïtcin, R. Flatt (Eds.), Science and Technology of Concrete Admixtures, Woodhead Publishing, Amsterdam, 2016, pp. 149–218.
[136] G. Gelardi, R.J. Flatt, Working mechanism of water reducers and superplasticizers, in: P.-C. Aïtcin, R. Flatt (Eds.), Science and Technology of Concrete Admixtures, Woodhead Publishing, Amsterdam, 2016, pp. 257–278.
[137] M. Cyr, C. Legrand, M. Mouret, Study of the shear-thickening effect of superplasticizers on the rheological behaviour of cement pastes containing or not mineral additives, Cem. Concr. Res. 30 (2000) 1477–1483.
[138] D. Marchon, S. Kawashima, H. Bessaies-Bey, S. Mantellato, S. Ng, Hydration and rheology control of concrete admixtures for digital fabrication. Cem. Concr. Res., (this special issue).
[139] R. Feret, Sur la compacité des mortiers hydrauliques. Mémoires et documents relatifs à l'art des constructions au service de l'ingénieur, Annales des Ponts et Chaussées, (1892), pp. 5–161 2nd semester.
[140] J. Bolomey, Détermination de la résistance probable d'un béton connaissant son dosage et sa densité au moment du gâchage, Bull. Tech. de la Suisse Romande, 55ème année, No. 17, (1929), pp. 193–197.
[141] T.C. Powers, T.L. Brownyard, Studies of Physical Properties of Hardened Portland Cement Paste, Portland Cem. Assoc. Bull. (no. 22 (1948) 342 p.; Reprinted from J. Amer. Concr. Inst. (Oct-Dec 1946; Jan-Apr 1947); Proceedings, 43, 101–132, 249–336, 469–504, 549–602, 669–712, 845–857, 865–880, 933–969, 971–992).
[142] T.C. Powers, Structure and physical properties of hardened Portland cement paste, J. Amer. Ceram. Soc. 41 (1958) 1–6.
[143] P.-C. Aïtcin, J. Baron, J.-P. Bournazel, Viser une résistance à la compression, in: J. Baron, J.-P. Ollivier (Eds.), Les Bétons. Bases et Données Pour Leur Formulation, Eyrolles, Paris, 1996, pp. 277–305.
[144] H.F.W. Taylor, Cement Chemistry, Academic Press, 1997.
[145] Y. Suda, T. Saeki, T. Saito, Relation between chemical composition and physical properties of C-S-H generated from cementitious materials, J. Adv. Concr. Technol. 13 (2015) 275–290.
[146] J. Haas, A. Nonat, From C-S-H to C-A-S-H: experimental study and thermodynamic modelling, Cem. Concr. Res. 68 (2015) 124–138.
[147] D. Hou, Z. Li, T. Zhao, Reactive force field simulation on polymerization and hydrolytic reactions in calcium aluminate silicate hydrate (C-A-S-H) gel: structure, dynamics and mechanical properties, RSC Adv. 5 (2015) 448–461.
[148] P. Termkhajornkit, Q. Huy Vu, R. Barbarulo, S. Daronnat, G. Chanvillard, Dependence of compressive strength on phase assemblage in cement pastes: beyond gel-space ratio – experimental evidence and micromechanical modelling, Cem. Concr. Res. 56 (2014) 1–11.
[149] M. Vandamme, F.-J. Ulm, Nanogranular origin of concrete creep, Proc. Natl. Acad. Sci. U. S. A. 106 (26) (2009) 10552–10557.
[150] I. Pignatelli, A. Kumar, R. Alizadeh, Y. Le Pape, M. Bauchy, G. Sant, A dissolution-precipitation mechanism is at the origin of concrete creep in moist environments, J. Chem. Phys. 145 (2016) 054701.
[151] A. Nonat, The structure and stoichiometry of C-S-H, Cem. Concr. Res. 34 (2004) 1521–1528 (in particular, Fig. 1).
[152] A.J. Allen, J.J. Thomas, H.M. Jennings, Composition and density of nanoscale calcium-silicate-hydrate in cement, Nat. Mater. 6 (2007) 311–316.
[153] G. Constantinides, F.-J. Ulm, The nanogranular nature of C-S-H, J. Mech. Phys. Solids 55 (2007) 64–90.
[154] F.-J. Ulm, M. Vandamme, C. Bobko, J.A. Ortega, K. Tai, C. Ortiz, Statistical indentation techniques for hydrated nanocomposites: concrete, bone, shale, J. Am. Ceram. Soc. 90 (2007) 2677–2692.
[155] F. Ridi, E. Fratini, P. Baglioni, Cement: a two thousand year old nano-colloid, J. Colloid Interface Sci. 357 (2011) 255–264.
[156] W.-S. Chiang, E. Fratini, P. Baglioni, D. Liu, S.-H. Chen, Microstructure determination of calcium-silicate-hydrate globules by small-angle neutron scattering, J. Phys. Chem. C 116 (2012) 5055–5061.
[157] M.J. Abdolhosseini Qomi, K.J. Krakowiak, M. Bauchy, K.L. Stewart, R. Shahsavari, D. Jagannathan, D.B. Brommer, A. Baronnet, M.J. Buehler, S. Yip, F.-J. Ulm, K.J. Van Vliet, R.J.-M. Pellenq, Combinatorial molecular optimization of cement hydrates, Nat. Commun. 5 (2014) 4960.
[158] A. Kumar, B.J. Walder, A.K. Mohamed, A. Hofstetter, B. Srinivasan, A.J. Rossini, K. Scrivener, L. Emsley, P. Bowen, The atomic-level structure of cementitious calcium silicate hydrate, J. Phys. Cem. C 121 (2017) 17188–17196.
[159] S. Papatzani, K. Paine, J. Calabria-Holley, A comprehensive review of the models on the nanostructure of calcium silicate hydrates, Constr. Build. Mater. 74 (2015) 219–234.
[160] J.J. Thomas, A new approach to modeling the nucleation and growth kinetics of tricalcium silicate hydration, J. Amer. Ceram. Soc. 90 (10) (2007) 3282–3288.
[161] G.W. Scherer, J. Zhang, J.J. Thomas, Nucleation and growth models for the hydration of cement, Cem. Concr. Res. 42 (2012) 982–993.
[162] S. Bishnoi, K.L. Scrivener, Studying nucleation and growth kinetics of alite hydration using µic, Cem. Concr. Res. 39 (2009) 249–260.
[163] J.W. Bullard, A determination of hydration mechanism for tricalcium silicate using a kinetic cellular automaton model, J. Amer. Ceram. Soc. 91 (7) (2008) 2088–2097.
[164] J.W. Bullard, H.M. Jennings, R.A. Livingston, A. Nonat, G.W. Scherer, J.S. Schweitzer, K.L. Scrivener, J.J. Thomas, Mechanisms of cement hydration, Cem. Concr. Res. 41 (2011) 1208–1223.
[165] M. Duriez, R. Lévy, Possibilités nouvelles dans le durcissement rapide des ciments, mortiers et bétons, Ann. De l'Inst. Tech. Des B.T.P. 98 (1956) 137–158.
[166] J.J. Thomas, H.M. Jennings, J.J. Chen, Influence of nucleation seeding on the hydration mechanisms of tricalcium silicate and cement, J. Phys. Chem. C 113 (2009) 4327–4334.
[167] R. Alizadeh, L. Raki, J.M. Makar, J.J. Beaudoin, I. Moudrakovski, Hydration of tricalcium silicate in the presence of synthetic calcium-silicate-hydrate, J. Mater. Chem. 19 (2009) 7937–7946.
[168] L. Nicoleau, New calcium silicate hydrate network, J. Transp. Res. Board (2010) 42–51 No. 2142.
[169] L. Nicoleau, T. Gädt, L. Chitu, G. Maier, O. Paris, Oriented aggregation of calcium silicate hydrate platelets by the use of comb-like copolymers, Soft Matter 9 (2013) 4864–4874.
[170] J. Rieger, M. Kellermeier, L. Nicoleau, Formation of nanoparticles and nanostructures – an industrial perspective on $CaCO_3$, cement, and polymers, Angew. Chem. Int. Ed. 53 (2014) 12380–12396.
[171] M. Kutschera, L. Nicoleau, M. Bräu, Nano-optimized construction materials by nano-seeding and crystallization control, in: K. Gopalakrishnan, B. Birgisson, P. Taylor, N. Attoh-Okine (Eds.), Nanotechnology in Civil Infrastructure, Springer-Verlag, Berlin Heidelberg, 2011, pp. 175–205.
[172] L. Reiter, T. Wangler, N. Roussel, R.J. Flatt, The role of early age structural buildup in digital fabrication with concrete, Cem. Concr. Res. (2018) (this special issue).
[173] T. Wangler, E. Lloret, L. Reiter, N. Hack, F. Gramazio, M. Kohler, M. Bernhard, B. Dillenburger, J. Buchli, N. Roussel, R. Flatt, Digital concrete: opportunities and challenges, RILEM Technol. Lett. 1 (2016) 67–75.
[174] S.D. Palkovic, D.B. Brommer, K. Kupwade-Patil, A. Masic, M.J. Buehler, O. Büyüköztürk, Roadmap across the mesoscale for durable and sustainable cement paste – a bioinspired approach, Constr. Build. Mater. 115 (2016) 13–31.
[175] L. Nicoleau, Accelerated growth of calcium silicate hydrates: experiments and simulations, Cem. Concr. Res. 41 (2011) 1339–1348.
[176] M. Reza Andalibi, A. Kumar, B. Srinivasan, P. Bowen, K. Scrivener, C. Ludwig, A. Testino, On the mesoscale mechanism of synthetic calcium-silicate-hydrate precipitation: a population balance modeling approach, J. Mater. Chem. A 6 (2018) 363–373.
[177] K. Ioannidou, R.J.-M. Pellenq, E. Del Gado, Controlling local packing and growth in calcium-silicate-hydrate gels, Soft Matter 10 (2014) 1121–1133.
[178] K. Ioannidou, K.J. Krakowiak, M. Bauchy, C.G. Hoover, E. Masoero, S. Yip, F.-J. Ulm, P. Levitz, R.J.-M. Pellenq, E. Del Gado, Mesoscale texture of cement hydrates, Proc. Natl. Acad. Sci. U. S. A. 113 (2016) 2029–2034.
[179] K. Ioannidou, M. Kanduc, L. Li, D. Frenkel, J. Dobnikar, E. Del Gado, The crucial effect of early-age gelation on the mechanical properties of cement hydrates, Nat. Commun. 7 (2016) 12106.
[180] I. Shvab, L. Brochard, H. Manzano, E. Masoero, Precipitation mechanisms of mesoporous nanoparticle aggregates: off-lattice, coarse-grained, kinetic simulations, Cryst. Growth Des. 17 (2017) 1316–1327.
[181] C. Plassard, E. Lesniewska, I. Pochard, A. Nonat, Nanoscale experimental investigation of particle interactions at the origin of the cohesion of cement, Langmuir 21 (2005) 7263–7270.
[182] D. Viehland, J.-F. Li, L.-J. Yuan, Z. Xu, Mesostructure of calcium silicate hydrate (CSH) gels in Portland cement paste: short-range ordering, nanocrystallinity, and local compositional order, J. Am. Ceram. Soc. 79 (1996) 1731–1744.
[183] L. Gatty, S. Bonnamy, C. Clinard, A. Feylessoufi, P. Richard, H. Van Damme, A transmission electron microscopy study of interfaces and matrix homogeneity in ultra-high-performance cement-based materials, J. Mater. Sci. 36 (2001) 4013–4026.
[184] B. Lothenbach, K. Scrivener, R.D. Hooton, Supplementary cementitious materials, Cem. Concr. Res. 41 (2011) 1244–1256.
[185] S.J. Jiang, H. Van Damme, Influence of fillers on textural and mechanical properties of C3S pastes, in: P. Colombet, A.-R. Grimmer, H. Zanni, P. Sozzani (Eds.), Nuclear Magnetic Resonance Spectroscopy of Cement-Based Materials, Springer-Verlag, Berlin, 1998, pp. 379–386.
[186] D. Damidot, B. Lothenbach, D. Herfort, F.P. Glasser, Thermodynamics and cement science, Cem. Concr. Res. 41 (2011) 679–695.
[187] B. Lothenbach, F. Winnefeld, Thermodynamic modelling of the hydration of Portland cement, Cem. Concr. Res. 36 (2006) 209–226.
[188] K. De Weerdt, M. Ben Haha, G. Le Saout, K.O. Kjellsen, H. Justnes, B. Lothenbach, Hydration mechanisms of ternary Portland cements containing limestone powder and fly ash, Cem. Concr. Res. 41 (2011) 279–291.
[189] J. Sanahuja, L. Dormieux, G. Chanvillard, Modelling elasticity of a hydrating cement paste, Cem. Concr. Res. 37 (2007) 1427–1439.
[190] B. Pichler, C. Hellmich, Upscaling quasi-brittle strength of cement paste and mortar: a multi-scale engineering mechanics model, Cem. Concr. Res. 41 (2011) 467–476.
[191] B.L. Damineli, F.M. Kemeid, P.S. Aguiar, V.M. John, Measuring the eco-efficiency of cement use, Cem. Concr. Compos. 32 (2010) 555–562.
[192] B.L. Damineli, R.G. Pileggi, V.M. John, Lower binder intensity eco-efficient concretes, in: F. Pacheco-Torgal, S. Jalali, J. Labrincha, V.M. John (Eds.), Eco-Efficient Concrete, Woodhead Publishing in Materials, 2013, pp. 26–44.
[193] R.J. Flatt, N. Roussel, C.R. Cheeseman, Concrete: an eco-material that needs to be improved, J. Eur. Ceram. Soc. 32 (2012) 2787–2798.
[194] L. Bertrand, C. Gervais, A. Masic, L. Robbiola, Paleo-inspired systems: durability, sustainability, and remarkable properties, Angew. Chem. Int. Ed. Eng. (2017), http://dx.doi.org/10.1002/anie.201709303.
[195] E.M. Gartner, D.E. Macphee, A physico-chemical basis for novel cementitious binders, Cem. Concr. Res. 41 (2011) 736–749.

[196] E. Gartner, T. Sui, Alternative cement clinkers, Cem. Concr. Res. (2018), http://dx.doi.org/10.1016/j.cemconres.2017.02.002 (in press).
[197] K. Kupwade-Patil, C. De Wolf, S. Chin, J. Ochsendorf, A.E. Hajiah, A. Al-Mumin, O. Büyüköztürk, Impact of embodied energy on materials/buildings with partial replacement of ordinary Portland cement (OPC) by natural Pozzolanic volcanic ash, J. Clean. Prod. 177 (2018) 547–554.
[198] P. Teicholz, P.M. Goodrum, C.T. Haas, U.S. construction labor productivity trends, 1970–1998, J. Constr. Eng. Manag. 127 (2001).
[199] A. Dubois, L.-E. Gadde, The construction industry as a loosely coupled system: implications for productivity and innovations, Constr. Manag. Econ. 20 (2002) 621–631.
[200] R. Fulford, C. Standing, Construction industry productivity and the potential for collaborative practice, Int. J. Proj. Manag. 32 (2014) 315–326.
[201] V.J. Shinde, M.N. Hedaoo, Productivity improvement in construction industry, Int. J. Eng. Technol. Sci. Res. 4 (2017) 1169–1175.
[202] D. Asprone, C. Menna, F.P. Bos, T.A.M. Salet, J. Mata-Falcon, W. Kaufmann, Rethinking reinforcement for digital fabrication with concrete, Cem. Concr. Res. (2018) (this special issue).
[203] M. Fauconneau, F.K. Wittel, H.J. Herrmann, Continuous wire reinforcement for jammed granular architecture, Granul. Matter 18 (2016) (paper 27).
[204] P. Block, Compression only structures, Cem. Concr. Res., this special issue.
[205] G.M. Whitesides, B. Grzybowski, Self-assembly at all scales, Science 295 (2002) 2418–2421.
[206] S. Tibbits, From automated to autonomous assembly, in: S. Tibbits (Ed.), Autonomous Assembly, Architectural Design, 87(4) 2017, pp. 7–15 Profile No. 248.
[207] P.M. Reis, H.M. Jaeger, M. van Hecke, Designer matter: a perspective, extreme, Theor. Appl. Mech. Lett. 5 (2015) 25–29.
[208] S. Keller, H.M. Jaeger, Aleatory architectures, Granul. Matter 18 (2016) (paper 29).
[209] K. Dierichs, A. Menges, Aggregate structures: material and machine computation of designed granular substances, Archit. Des. 82 (2012) 74–81.
[210] S. Keller, Bidden City (Beijing Olympics), Artforum Int. 46 (2008) 137–145.
[211] S.V. Franklin, Extensional rheology of entangled granular materials, Europhys. Lett. 106 (2014) (paper 58004).
[212] S.V. Franklin, Geometric cohesion in granular materials, Phys. Today 65 (9) (2012) 70.
[213] M. Tennenbaum, Z. Liu, D. Hu, A. Fernandez-Nieves, Mechanics of fire ants aggregations, Nat. Mater. 15 (2016) 54–59.
[214] D.L. Hu, S. Phoneko, E. Altshuler, F. Brochard-Wyart, Entangled active matter: from cells to ants, Eur. Phys. J. Special Topics 225 (2016) 629–649.
[215] H.M. Jaeger, Toward jamming by design, Soft Matter 11 (2015) 12–27.
[216] K.A. Murphy, N. Reiser, D. Choksy, C.E. Singer, H.M. Jaeger, Freestanding load-bearing structures with Z-shaped particles, Granul. Matter 18 (2016) (paper 26).
[217] Y. Zhao, K. Liu, M. Zheng, J. Barés, K. Dierichs, A. Menges, R.P. Behringer, Packings of 3D stars: stability and structure, Granul. Matter 18 (2016) (paper 24).
[218] J. Barès, Y. Zhao, M. Renouf, K. Dierichs, R. Behringer, Structure of hexapod 3D packings: understanding the global stability from the local organization, Eur. Phys. J. Web Conf. 140 (2017) 06021, , http://dx.doi.org/10.1051/epjconf/201714006021.
[219] D. Dumont, M. Houze, P. Rambach, T. Salez, S. Patinet, P. Damman, Emergent strain stiffening in interlocked granular chains, Phys. Rev. Lett. 120 (2018) 088001.
[220] P. Aejmelaeus-Lindström, J. Willmann, S. Tibbits, F. Gramazio, M. Kohler, Jammed architectural structures: towards large-scale reversible construction, Granul. Matter 18 (2016) (paper 28).
[221] K. Dierichs, A. Menges, Toward an aggregate architecture: designed granular systems as programmable matter in architecture, Granul. Matter 18 (2016) (paper 25).
[222] M.Z. Miskin, H.M. Jaeger, Evolving design rules for the inverse granular packing problem, Soft Matter 10 (2014) 3708–3715.
[223] M.Z. Miskin, H.M. Jaeger, Adapting granular materials through artificial evolution, Nat. Mater. 12 (2013) 326–331.
[224] F. Huijben, F. van Herwijnen, Vacuumatics: vacuumatically prestressed (adaptable) structures, in: J.F. Abel, J.R. Cooke (Eds.), Proc. 6th Int. Conf. on Computation of Shell and Spatial Structures IASS-IACM 2008, Multi Science Publishing, Brentwood, UK, 2008.
[225] F. Huijben, Vacuumatic formwork: a novel granular manufacturing technique for producing topology-optimized structures in concrete, Granul. Matter 18 (2016) (paper 23).
[226] M. Pawlin, Biomimicry in Architecture, RIBA Publishing, London, 2011.
[227] J. Vincent, New materials and natural design, in: R. Allen (Ed.), Bulletproof Feathers. How Science Uses Nature's Secrets to Design Cutting-Edge Technology, Univ. of Chicago Press, 2010, pp. 132–171.
[228] S.W. Cranford, M.J. Buehler, Biomateriomics, Springer, 2012.
[229] A.M. Mirzendehdel, K. Suresh, A Hands-On Introduction to Topology Optimization, CreateSpace Independent Publishing Platform, 2017.
[230] J. Zhu, T. Gao, Topology Optimization in Engineering Structure Design, Elsevier, 2016.
[231] H. Pottmann, A. Asperl, M. Hofer, A. Kilian, Architectural Geometry, Bentley Institute Press, 2007.
[232] H. Pottmann, M. Eigensatz, A. Vaxman, J. Wallner, Architectural geometry, Comput. Graph. 47 (2015) 145–164.
[233] I. Stewart, Mathematics: some assembly needed, Nature 448 (2007) 419.
[234] P.M. Reis, F. Lopez Jimenez, J. Marthelot, Transforming architectures inspired by origami, Proc. Natl. Acad. Sci. U. S. A. 112 (2015) 12234–12235.
[235] A.I. Bobenko, Y.B. Suris, Discrete differential geometry: integrable structure, graduate studies in mathematics, J. Am. Math. Soc. vol. 98, (2009).
[236] E.T. Filipov, T. Tachi, G.H. Paulino, Origami tubes assembled into stiff, yet reconfigurable structures and metamaterials, Proc. Natl. Acad. Sci. U. S. A. 112 (2015) 12321–12326.
[237] S.A. Zirbel, R.J. Lang, M.W. Thomson, D.A. Sigel, P.E. Walkemeyer, B.P. Trease, S.P. Magleby, L.L. Howell, Accomodating thickness in origami-based deployable arrays, J. Mech. Des. 135 (2013) 111005.
[238] S. Felton, M. Tolley, E. Demaine, D. Rus, R. Wood, Applied origami: a method for building self-folding machines, Science 345 (2014) 644–646.
[239] K. Kuribayashi, K. Tsuchiya, Z. You, D. Tomus, M. Umemoto, T. Ito, M. Sasaki, Self-deployable origami stent grafts as a biomedical application of Ni-rich TiNi shape memory alloy foil, Mater. Sci. Eng. A 419 (2006) 131–137.
[240] A. Lebée, From folds to structures, a review, Int. J. Space Struct. 30 (2015) 55–74.
[241] J.D. Birchall, A.J. Howard, K. Kendall, Flexural strength and porosity of cements, Nature 289 (1981) 388–390.
[242] A. Picker, L. Nicoleau, Z. Burhard, J. Bill, I. Zlotnikov, C. Labez, A. Nonat, H. Cölfen, Mesocrystalline calcium silicate hydrate: a bioinspired route toward elastic concrete materials, Sci. Adv. 3 (2017) e1701216.
[243] A. Picker, L. Nicoleau, A. Nonat, C. Labbez, H. Cölfen, Identification of binding peptides on calcium silicate hydrate: a novel view on cement additives, Adv. Mater. 26 (2014) 1135–1140.
[244] N. Hong-Guang, J. Zong Wang, Prediction of compressive strength of concrete by neural networks, Cem. Concr. Res. 30 (2000) 1245–1250.
[245] A. Baykasoglu, T. Dereli, S. Tanis, Prediction of cement strength using soft computing techniques, Cem. Concr. Res. 34 (2004) 2083–2090.
[246] S. Akkurt, G. Tayfur, S. Can, Fuzzy logic model for the prediction of cement compressive strength, Cem. Concr. Res. 34 (2004) 1429–1433.
[247] A. Oztas, M. Pala, E. Ozbay, E. Kanca, N. Caglar, M.A. Bhatti, Predicting the compressive strength and slump of high strength concrete using neural network, Constr. Build. Mater. 20 (2006) 769–775.
[248] I.B. Topçu, M. Saridemir, Prediction of compressive strength of concrete containing fly ash using artificial neural networks and fuzzy logic, Comput. Mater. Sci. 41 (2008) 305–311.
[249] C. Deepa, K. Sathiya Kumari, V. Pream Sudha, Prediction of the compressive strength of high performance concrete mix using tree based modeling, Int. J. Comput. Appl. 6 (2010) 18–24.
[250] J.-S. Chou, C.-F. Tsai, A.-D. Pham, Y.-H. Lu, Machine learning in concrete strength simulations: multi-nation data analytics, Constr. Build. Mater. 73 (2014) 771–780.
[251] P. Chopra, R.K. Sharma, M. Kumar, Predicting compressive strength of concrete for varying workability using regression models, Int. J. Eng. Appl. Sci. 6 (2014) 10–22.
[252] D. Prayogo, M.-Y. Cheng, J. Widjaja, H. Ongkowijoyo, H. Prayogo, Prediction of concrete compressive strength from early age test result using an advanced metaheuristic-based machine learning technique, Proc. 34th International Symposium on Automation and Robotics in Construction (ISARC 2017), 2017.
[253] P. Chopra, R.K. Sharma, M. Kumar, T. Chopra, Comparison of machine learning techniques for the prediction of compressive strength of concrete, Adv. Civil Eng. vol. 2018, (2018) 9 (Article ID 5481705).
[254] B. Boukhatem, S. Kenai, A. Tagnit-Hamou, M. Ghrici, Application of new information technology on concrete: an overview, J. Civ. Eng. Manag. 17 (2011) 248–258.
[255] I. Goodfellow, Y. Bengio, A. Courville, Deep Learning, MIT Press, Cambridge, MA, 2016.
[256] F. Chollet, Deep Learning With Python, Manning Publications Co., Shelter Island, NY, 2018.
[257] F. Ren, L. Ward, T. Williams, K.J. Laws, C. Wolverton, J. Hattrick-Simpers, A. Mehta, Accelerated discovery of metallic glasses through iteration of machine learning and high-throughput experiments, Sci. Adv. 4 (2018) eaaq1566.