Sustainability in Construction Materials: From Waste Valorization to Circular Economy

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Abstract Already from the beginning, 50 years ago, the first nucleus of researchers establishing the research group, was convinced that the construction sector was the best option for the valorization of industrial by-products as "secondary raw materials". In fact, this sector is probably the largest consumer of resources and the largest waste generator, consequently it has huge environmental impact. On the other hand,

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construction materials affect the performance of buildings with respect to safety, health, environmental performance and energy efficiency. Manufacturing of construction products using alternative raw materials; recycling to manage construction and demolition waste; durability and environmental compatibility of materials: all these were the different and challenging fields of research that the group has faced in a continuous effort of innovation and cooperation at national and international level. The focus of the group was already perfectly in line with what is now called "Circular Economy", which at present is considered a revolution in the way of human economic development. We are sure the group thus contributed to this revolution even before the term was in current use. We feel ready for the next 50.

1 The First Nucleus of Researchers

The first Department that was formed at the Engineering Faculty of the University of Ancona, today the Polytechnic University of the Marche, towards the end of the 70s of the last century was the Department of Materials and Earth Sciences.

The constitution was made possible by the aggregation of three institutes: Chemistry, Physics and Geology. The first Director was Prof. Savino Melone who organized the Department in three sections that were based on the founding institutes, coordinating the planning of organizational activities and the development through the establishment of a departmental council.

1.1 Short History: People and Structures

They were pioneering years because, being the newly founded Faculty of Engineering and the location at a converted industrial warehouse, the laboratories were lacking in equipment and they had difficulty to start also because of the precariousness of the teaching staff that for the most part was not resident, but coming from other universities.

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The research activities on which to invest began to take shape towards the second half of the 70s, when the young structured researchers had identified their areas of interest and when they were assigned the first chairs as a full professor who, coming from other universities, to transfer their research guide lines at the Department.

For the Chemistry section of the Department, are the cases of Prof. Paolo Bruni, already present at the Department since the early 70's and of Prof. Mario Collepardi who, coming from the University of Rome, remained in the Department for 25 years, giving a fundamental mark to the research on cementitious materials.

Over the years, of course, there have been significant changes, the first of all the transfer to the new headquarters of the Engineering Faculty, in 1984, which saw a consistent implementation of the research laboratories, but also the reorganization of the disciplines related to the Department. The geological section expanded its disciplines and research lines in the engineering sector of geotechnics, as well as in the Physics section the research sectors in the fields of optics and microstructure of materials were diversified with the entry of new full professors.

Also the Chemistry section, grouping different disciplinary scientific sectors, has undergone splitting and re-aggregation over time relating to staff afferents (Fig. 1) within the department, without however eliminating collaborations in research topics.

Over time the Department has changed its name several times from Materials Science and Earth, to Physics and Engineering of Materials and Territory, to Science and Engineering of the Materials of the Environment and Urban Planning, and of course there have been several Department Directors among which in addition to Prof. Savino Melone, in order of time, prof. Lucedio Greci, Prof. Giacomo Moriconi, Prof. Erio Pasqualini up to the current Prof. Oriano Francescangeli.

Fig. 1 Group photo (not to scale: the research group is bigger!)

1.2 Short History: Research Challenges and Opportunities

Already from the beginning, 50 years ago, the first nucleus of researchers establishing the research group, was convinced that the construction sector was the best option for the valorization of waste, industrial by-products and other recycled materials as "secondary raw materials".

The Department in its brief history, has assumed a peculiar configuration in which the academic community has consolidated an intense relationship with the regional territory, promoting at the same time, a strong and attractive activity both at national and international level. Its activity started enquiring on synergistic researches in the field of the interaction between the building materials and the environment, bearing in mind the specificities of the different scientific areas interconnected.

From this point of view, the department created a sort of "balancing of the skills and use of resources" through the creation of different sectors to give life to a structure in which different competences could contribute to a teaching of high level and a research of high international profile in the field of materials and environmental engineering.

At the foundation, professors came from many Italian universities and different cultural background, from those with a more fundamental vocation, such as Chemistry, Physics and Earth Sciences, to those more markedly applicative, such as materials engineering, and environment.

In this context the study of material-environment interactions, chemical-physical phenomena of interest in the industrial sector, corrosion and protection of materials, environmental defence and quality of living environments received the larger consideration, and represented the main focus of research of the STM group (Scienza e Tecnologia dei Materiali). The interest of the STM group was placed on sustainable construction materials, recycling industrial by-products or biomass ashes in mortars and concretes keeping in mind particularly the environment through the monitoring of soil, water and air quality with the assessment of chemical risk and, diagnosis and prognosis of living and working environments.

Among the most relevant applications, the use of photocatalytic materials applied in the motorway area with the creation of vertical surfaces (piers and tunnel vault, masonry surfaces) and on the road pavement with monitoring of short-term and longer-term effects on the reduction of NOx emissions and precursors of atmospheric pollution.

2 New Products from Industrial By-Products

A judicious use of natural resources, achieved by the use of by-products and recyclable materials, and a lower environmental impact, achieved through reduced carbon dioxide emission and reduced natural aggregate extraction from quarries, represent two main actions that meet the needs for sustainable construction development. It is fundamental to discuss and define the criteria on the basis of which the use of by-products and recyclable materials in concrete can be optimized.

When using recycled materials, for instance, the fresh concrete behaviour during placing can change. Moreover, when using recycled materials appropriately, some important properties of the hardened concrete such as ductility and durability can be better engineered, as several works of the STM group explains and emphasizes.

2.1 Low CO_2 Cement from Waste

With an annual production of almost 3 Gt Ordinary Portland cement (OPC) is the dominant binder of the construction industry. The cement industry contributes about 7% of the total worldwide CO_2 emissions. The urge to reduce carbon dioxide emissions and the fact that OPC structures which have been build a few decades ago are still facing disintegration problems points out the handicaps of OPC. The early deterioration of reinforced concrete structures based on OPC is a current phenomenon with significant consequences both in terms of the cost for the rehabilitation of these structures, or even in terms of environmental impacts associated with these operations.

2.1.1 Alkali Activated Binders Products

Increased attention to the environmental impact of OPC has prompted researchers to study the optimization of alternative clinkerless construction materials [8, 9, 26].

Alkali Activated Cements (AAC) are a novel class of cement-like materials obtained by the polymerization reaction of a solid aluminosilicate with an aqueous solution of alkali hydroxide, silicate, carbonate or sulphate [32] (Fig. 2).

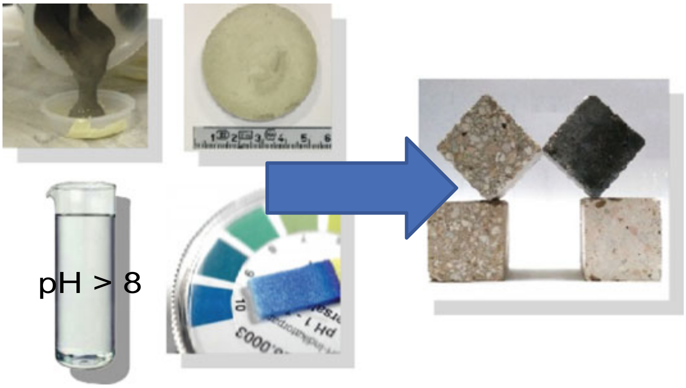

Fig. 2 Alkali activated binders

Aluminosilicate precursors are generally industrial by-products, and thanks to the absence of limestone and a general energy savings in the production of AAC, a reduction of 40–90% of greenhouse gases emissions compared to that of OPC materials with the same characteristics has been estimated [10].

The STM Group contributed further to the state of art of AAC mortars by investigating their behaviour when cured at room conditions and compared with traditional cementitious mortars at the same compressive strength class. In particular, according to UNI EN 1504-3:2006, mortars belonging to two non-structural classes (R1 $\geq$ 10 MPa and R2 $\geq$ 15 MPa) and one to structural class (R3 $\geq$ 25 MPa) were tested and compared both in the fresh and in the hardened state.

The obtained results demonstrates the possibility of manufacturing AAC mortars at room conditions which can cover three mechanical strength classes according to UNI EN 1504-3:2006 (R1 $\geq$ 10 MPa, R2 $\geq$ 15 MPa and R3 $\geq$ 25 MPa) by increasing the alkalinity of the activating solution [27–30]. In particular, the use of NaOH in alkali activated fly ash mortars caused efflorescence formation which was more pronounced with the increase of the compressive strength. The combined use of KOH instead of NaOH and calcium aluminate cement on fly ash weight removed the efflorescence phenomena [29].

Moreover, at the same mechanical strength class:

- the faster strength gaining of FA mortars prepared with KOH with respect to those prepared with NaOH is due to the faster and stronger incorporation of K^+ into the gel than the Na^+ ; in the same way, the faster strength development of MKK mortars with respect to FA ones is due to the faster polymerization of metakaolin with respect to fly ash;
- the dynamic modulus of elasticity of AAC mortars was at least 50% lower than that of traditional OPC mortars, respectively;
- AAC mortars showed lower restrained shrinkages than those of OPC mortars due to the lower modulus of elasticity;
- even if the higher alkalinity of AAC paste delayed the tendency to passivation of reinforcements, especially if galvanized, after one month of curing both bare and galvanized steel bars reached the same corrosion rates of those embedded in OPC mortars.

In addition, at the same mechanical strength class, many results depend strongly on pore distribution and total porosity of mortars since:

- the higher drying shrinkage of AAC mortars with respect to that of OPC mortars is due to the higher mesopores volume;
- the lower weight losses of FA mortars are due to the higher mesopores volume which has hindered the evaporation of water remained absorbed to the their surface;
- AAC mortars are more permeable to water vapour with respect to OPC mortars due to the higher presence of pores with large dimensions in FA mortars and the higher total porosity in MKK mortars;
- MKK mortars and FA mortars absorbed more and less water, respectively than OPC mortars since their total porosity was 60% higher and 40% lower than those of OPC mortars, respectively;

- FA mortars prepared with KOH and CAC showed an excellent resistance to sulphate attack thanks to their lowest porosities.

2.1.2 Chloride Induced Corrosion of Reinforcements in Alkali Activated Binders

In coastal zones, chlorides [39] promote the corrosion of embedded reinforcements and this phenomenon is considered one of the major cause of premature failure of reinforced concrete structures [35]. Methods proposed to mitigate reinforced concrete deterioration include the use of hydrophobic treatments, due to their ability to make concrete less susceptible to water saturation [6], corrosion inhibitors [5, 42], stainless steel rebars which are highly resistant to corrosion but very expensive, and galvanized reinforcements [2–4, 34]. In particular, galvanization of steel reinforcements is a cheaper prevention method against corrosion if compared to other anti-corrosion methods [4, 35, 41].

In the presence of $Ca(OH)_2$, as in the concrete pore solution, the protective layer is not only formed by zinc oxide and hydroxide, but mainly by a compact, protective, and highly chloride-resistant layer of calcium hydroxyzincate (CaHZn). The passivation of galvanized steel largely depends on the alkalinity of the environment and the concentration of Ca^{2+} ions. The passivation layer may be less protective due to the presence of chlorides or a high amount of soluble alkalis, whereas the formation process is favoured by oxygen availability [38].

It is well-known that steel reinforcements passivate in the alkaline environment of concrete pore solution, but in AAC, where NaOH or KOH concentrated solutions are used to activate the aluminosilicate powders, the alkalinity is much higher than in traditional OPC matrices. Alkalis are highly mobile in the pore system of AAC and this effect may significantly limit the durability of embedded reinforcements. Moreover, in AAC, the content of calcium, which contributes to the galvanized steel passivation, is much less than in OPC matrices.

The information provided by the literature on steel corrosion in AAC is limited and generally focused on simulated pore solution of alkali-activated concretes or fly ash or slag based mixtures, whereas there are no studies on metakaolin-based AAC.

Thus, the STM group investigated:

- the passivation capacity of bare and galvanized steel reinforcements in metakaolin (MK) and fly ash (FA) based AAC during the curing time [27, 43];
- the corrosion behaviour of bare and galvanized steel reinforcements in MK-and FA-based AAC in the presence of chlorides [27, 43];
- the passivation capacity and the chlorides induced corrosion behaviour of bare and galvanized steel reinforcements in FA-and MK-based AAC in comparison with that of cement based mortars, at the same strength class [27, 43].

To this aim the corrosion behaviour of both bare and galvanized steel reinforcements embedded in geopolymer and ordinary Portland cement-based mortars with

Fig. 3 Specimen for testing corrosion of reinforcing bars in AAC

three different strength classes (R1 ≥ 10 MPa, R2 ≥ 15 MPa and R3 ≥ 25 MPa) was investigated (Fig. 3) and compared both in the first month of curing and during 12 weekly wet-dry cycles in a 3.5% NaCl solution.

The main obtained results show that:

- during the first days after the cast, AAC prolong the active state of bare and galvanized steel reinforcements due to their initially very high alkalinity [30, 43]
- after 10 days of curing, polarization resistance increases for both bare and galvanized steel reinforcements in all types of mortars, indicating a clear tendency to-wards the passivation. This result is of particular importance for galvanized steel reinforcements in AAC mortars because the passivation process in these matrices occurs even in the absence of calcium, which is considered a fundamental element for galvanized steel passivation in common Portland cement-based materials
- during wet-dry cycles in a 3.5% NaCl solution, fly ash-based AAC ensure a higher protection to bare steel reinforcements compared to metakaolin-based AAC. Cementitious mortars protect bare steel reinforcements less than fly ash based AAC but better than metakaolin-based AAC. This is due to the higher total porosity of metakaolin-based matrix compared the other AAC matrices, which favours the ingress and thus the attack of chloride ions
- during wet-dry cycles in a 3.5% NaCl solution, galvanized steel reinforcements are strongly attacked in metakaolin-based AAC again due to the highest total porosity of metakaolin-based matrix

- for bare steel reinforcements, the higher alkalinity of the AACs matrix increases the minimum free chloride threshold necessary to induce the onset of corrosion in a chloride-rich environment compared to the cementitious matrix
- for galvanized steel reinforcements, the higher alkalinity of the fly ash based AAC matrix seems to decrease the minimum free chloride threshold necessary to induce the onset of corrosion in a chloride-rich environment compared to the cementitious matrix. However, pitting corrosion is not so penetrating to affect the Zn–Fe layer underneath pure zinc layer. Therefore, fly ash AAC are quite protective even for galvanized steel reinforcements.

2.2 *Multifunctional Building Materials from Biomass Waste*

Recent laws and directives are becoming stricter on energy consumption in buildings so more isolated and watertight structures are built to reduce the loss of heat. However, if adequate air changing is not guaranteed, the concentration of airborne pollutants and humidity in the indoor environment becomes a great problem. Reactive building materials offer an opportunity to provide indoor air cleaning with minimal energy use.

At this purpouse the STM group investigated the development of multifuncional building materials using as unconventional materials biomass and/or biomass wastes in order to also avoid wasting resources and raw materials supplied [22–24]. In particular, when the focus was on biomass wastes, the effect of using spruce sawdust shavings and biomass ashes as unconventional aggregates in lime-based mortar for indoor application has been investigated in terms of developed mechanical strength, permeability, capillary water absorption, moisture buffering ability and VOCs adsorption (Fig. 4) [40].

The effect of replacing sand volume with unconventional aggregates based on the different biomass wastes in lime-based mortars for indoor applications implies:

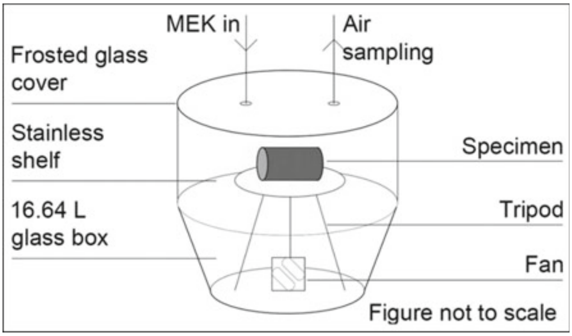

Fig. 4 Scheme of the test box for VOCs adsorption assesment

- A decrease of density of mortars: spruce saw dust and fly ash mortars can be classified as lightweight mortars
- A general decrease of compressive strength, but still acceptable for plastering/rendering applications. Instead, bottom biomass ashes increase the compressive strength of lime-based mortars of about 60%
- A general increase of capillary water absorption. However, bottom biomass ashes decrease the capillary water absorption of lime-based of about 50%, while the torrefaction process permits to 50% lower water absorption by capillarity action of mortars manufactured with spruce sawdust shavings
- An increase of water vapour permeability up to 50%
- An enhancement of indoor air quality in terms of up to three times higher MBV and up to 75% increased capacity to adsorb VOCs
- In this way, a better management of biomass waste and reduction of materials in landfill can also be achieved.

2.3 Smart Energy and Materials from Wastewater

During the last 50 years, the environmental policies and the social sensibility increased the attention to the pollutants types and concentrations in the treated wastewaters in order to obtain higher quality standard levels of the surface water.

The development of the technologies for wastewater during the time was fast and related to some specific objectives. For the nutrients reduction the traditional activated sludge treatment technique became the treatment of reference all over the world from 1930. From this technology still with full success, also several new and advanced processes based on the activated sludge principle were developed like membrane bioreactors, intermittent aeration units [33, 18, 19] and short cut via nitrite technologies. The biological unit in the time was optimized by increasing the possibility of completely reclaiming the existing structures in the full-scale plants, by introducing the feasibility of automatic control applications, by obtaining energy savings and by coupling the nutrients removals with the biological reduction of the sludge production [21, 44].

Phosphorus, essential element to life, represents the second nutrient in the wastewater with Nitrogen. Demand for rock phosphate is such that there is a global threat of phosphorus scarcity. In 2014, rock phosphate was added to the European commission list of critical raw materials. Scientific and technological options for Phosphorous removal and recovery from wastewater were already developed from 2000 years [1].

In the last 10 years, the WWTP have moved from the concept of waste treatment, to the concept of water reuse for beneficial purposes, such as agricultural and landscape irrigation, industrial processes, non-potable domestic use and groundwater replenishing [7]. On site energy recovery, particularly as biogas production, in WWTP is widely diffused as an alternative source of energy, for the recovery of thermal, electrical and mechanical energy, to be consumed either inside or outside the plant. In the

Fig. 5 Carbonera-Smartech 5-PHA and Struvite Recovery from anaerobic supernatants-years 2016 SMART-Plant Project (www.smart-plant.eu)—Coordinator Università Politecnica delle Marche; Referent Partner University of Verona

last years, two-step bioconversion comes into prominence as more value is derived to volatile fatty acid production before ending up to other end-products. Moreover, anaerobic processes offer much more than conventional wastewater treatment, recovering sustainable energy and valuable biochemical [20, 25].

Nutrient recovery and recycling takes an important role in wastewater valorisation. Recovered nutrients from the wastewater can be utilized as soil amendments or fertilizers for beneficial uses in agriculture. In particular, ammonia form is advantageous because it predominates in anaerobic reactor effluents and can be useful for fertigation purposes. Phosphorus recovery (i.e. in the form struvite or phosphorous salts) becomes essential for preventing eutrophication in the aquatic environment and for alleviating economic dependence on phosphate rocks.

The resources mentioned above are those most commonly recovered in WWTPs; in addition to them there are more innovative ones like cellulosic primary sludge [17] and polyhydroxyalkanoate.

This new approach is promoting and will recognize, probably for the next 50 years, the wastewater management as one of the main strategic economic sectors for resources valorisation and smart materials production in the effective innovative point of view of circular economy [37] (Fig. 5).

3 Closing the Loop for Construction Materials

Concrete is one of the most widely used materials in the world for manufacturing structural elements for building and infrastructures. Conventional concrete is not considered an environmentally friendly material because of the use of non-renewable natural resources, such as sand and gravel, and its high embodied energy.

In most cases, these concrete elements are demolished at the end of their useful life, generating what is known as construction and demolition waste (CDW). Using selective demolition techniques, very pure concrete waste with a high potential for recycling can be obtained. For this, all non-mineral dry building materials, such as plasterboards, wood, metals, plastics, glass should be removed and separated before the demolition of concrete elements. All these extra materials can be recycled as well to produce eco-friendly plaster and mortars such as wood chips [14], waste glass [15], waste plastic particles [13], bricks.

The possibility of reusing concrete waste particles after suitable treatment in recycling plants has been widely studied with encouraging results. In particular, concerning structural concrete manufacture, several papers showed the suitability of reusing up to 30% coarse recycled aggregate particles for concrete strength classes up to 40 MPa [11, 16, 36]. Moreover, a correlation between elastic modulus and compressive strength of recycled-aggregate concrete (RAC) was found in (8), showing that 15% lower elastic modulus is achieved by using 30% recycled aggregates, while tensile strength is reduced by 10% if the same concrete strength class is achieved by replacing 30% virgin aggregates with recycled concrete particles [16].

In terms of drying shrinkage, particularly if finer coarse recycled-concrete aggregate is used, lower shrinkage strains are detected especially for earlier curing times. This last aspect, when considered together with a lower elastic modulus, predicts a lower tendency to crack in the RAC. Concerning time-dependent characteristics, creep behavior is more influenced by the presence of recycled aggregates than shrinkage, although its variations are rather limited compared to what occurs in traditional concrete [12, 16].

Even if 100% replacement of virgin aggregate is carried out by using particles coming from treatment of CDW, structural concrete can be prepared due to the positive effect on compressive strength achieved by adding fly ash or silica fume to the mixture as a fine aggregate replacement with the aid of an acrylic-based superplasticizer. In this way, an adequate strength class value (30 MPa), as required for a wide range of common structural uses, can be reached both through virgin aggregate concrete and recycled concrete aggregate with fly ash, by suitably decreasing water/cement with the aid of a superplasticizing admixture in order to maintain the same workability.

Moreover, if fly ash is added to RAC, the pore structure is improved, and particularly the volume of macro pores is reduced, causing benefits in terms of mechanical performances such as compressive, tensile and bond strengths. The addition of fly ash proved to be very effective in reducing carbonation and chloride ion penetration depths in concrete, even in RAC.

Finally, on the basis of the results obtained through cyclic loading tests of beam–column joints made of either ordinary or RAC concrete, evaluated by means of parameters such as cracking patterns, supplied and dissipated energy, ductility and design values, the joint made of RAC showed adequate structural behaviour.

The previous encouraging results were obtained by using the only coarse recycled aggregate fraction, while, many authors found that in RAC the fine recycled aggregate fraction is particularly detrimental to both mechanical performances and durability of concrete. In addition, the presence in the recycled aggregate of materials other than concrete (that are mainly crushed bricks and tiles) reduces concrete performance due to their higher porosity and consequently their lower density and higher water absorption. For this reason the more recent approach is to recycle for concrete production only the coarse recycled fraction.

In short, the use of materials coming from C&D waste recycling instead of sand for the production of bedding mortars proved to be profitable not only for the obvious environmental advantages but also in terms of improvement of the mortar–brick interface, which is generally recognized as the weak chain link of the masonry assemblage. A further opportunity can be the reuse of the very fine fraction (Fig. 6) coming from recycling of CDW as filler for concrete, especially self-compacting concrete mixtures. Results obtained on the basis of the rheological tests on cement pastes showed that rubble powder proved to be more promising with respect to limestone powder and fly ash as mineral addition for self-compacting concrete. This indication was confirmed by the results obtained for fresh concrete: in fact, in the case of fly ash and ground limestone as mineral addition, a certain flow-segregation could be recorded on fresh mixtures. This was confirmed by the ultrasonic pulse velocity measurements related to hardened concrete specimens with different segregation.

In conclusion, an optimization of the self-compacting concrete mixture seems to be achievable by the simultaneous use of rubble powder and coarse recycled aggregate with improved fresh concrete performance and unchanged concrete mechanical strength.

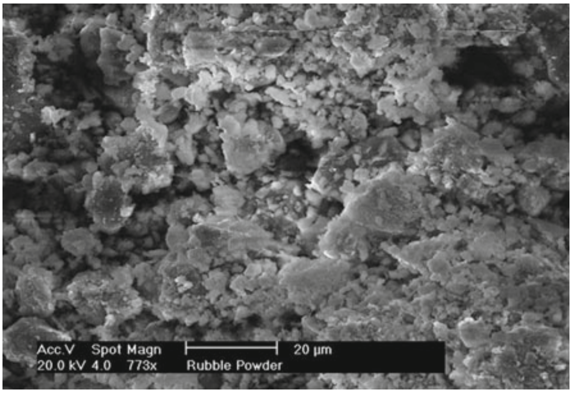

Fig. 6 SEM image of CDW powder at magnification of about 800×

4 A Tangible Vision: Beyond the Recycling Toward the Circular Economy

In recent years, the environmental and societal issues in the manufacturing, use, disposal and recycling of construction materials have been seriously considered. The demand for a more sustainable way of building and recovering the existing building stock is no longer a matter of personal choice, and actions are needed to increase knowledge and develop tools and regulations that enable maximum recovery of useful materials. This requires not only closing loops by reusing ‘waste’ and resources, but also slowing material loops by developing long lasting reusable products. In this way, the concept of Circular Economy (CE) can be effectively applied to the building sector, where innovation diffuses rather slowly, and where the focus has been on issues like energy use and energy efficiency more than recycling or reuse of materials at the end of life of buildings.

In this scenario, the research activities carried out by the group from the beginning of its life to date permit to laid solid foundations for creating a tangible vision concerning the future for next 50 years.

A strong link with the territory is consolidated and a wide network which ensures the global cross-fertilization is already created: now the exchange of ideas, innovation, research results, and researches themselves is a matter of fact and it is the key parameter which allows the cooperation and collaboration between academia and industry, locally and around the world.

The multidisciplinarity actually includes not only professors, but it also embraces students with different aptitudes and educations coming from different faculties who decide to approach the interaction between materials science and environment (Fig. 7).

In view of the multidisciplinarity and the network built by researchers, they are going to be increasingly included not only in the industrial context but also in the legislative one: an updated framework based on the needs of all stakeholders is

Fig. 7 New style of communication: from the group photo to the group selfie

the result of the group's research activity. The developed materials are not only sustainable but they also go beyond the simple concept of recycling:

- materials are designed to avoid the generation of waste and end-of-life products
- construction and demolition wastes are no longer treated as a minimum percentage, but they contribute to design new materials with added value
- new products are manufactured by using wastes/by-products originated from human activities and renewable natural sources to obtain smart energy and low CO_2 binders.

This perspective transforms wastes to secondary raw materials that are employed with a mindful design and used in suitable quantities, so as to reduce the impact of building materials.

Novel materials are designed with a view of multifunctionality to guarantee users' needs in a single solution. Newly developed materials are no more "passive" or "inert" with the surrounding environment, they are smart and self-sensing: they can be continually monitored in order to facilitate punctual interventions and guarantee not only the durability of structures, but also the health and safety of people that are in contact with them to live, work and play.

In this way initiatives, ideas as well as approaches are jointly related to fabricate more "circular buildings". The material intrinsic value is maintained and recovered as much as possible: today's products are resources for tomorrow.

Acknowledgements This work can not account of the most part of the scientific work of the people that in this 50 years have contributed to the activities of the research group. We have inevitably cut plenty of topics, not at all of minor interest or impact, but only because not easy ascribable under a homogeneous framework.

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