

Review

# Hydrotalcites in Construction Materials

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**Featured Application:** In this contribution, the applications of the hydrotalcites and hydrotalcite-like materials in the building materials, cements, mortars, and concretes are summarized.

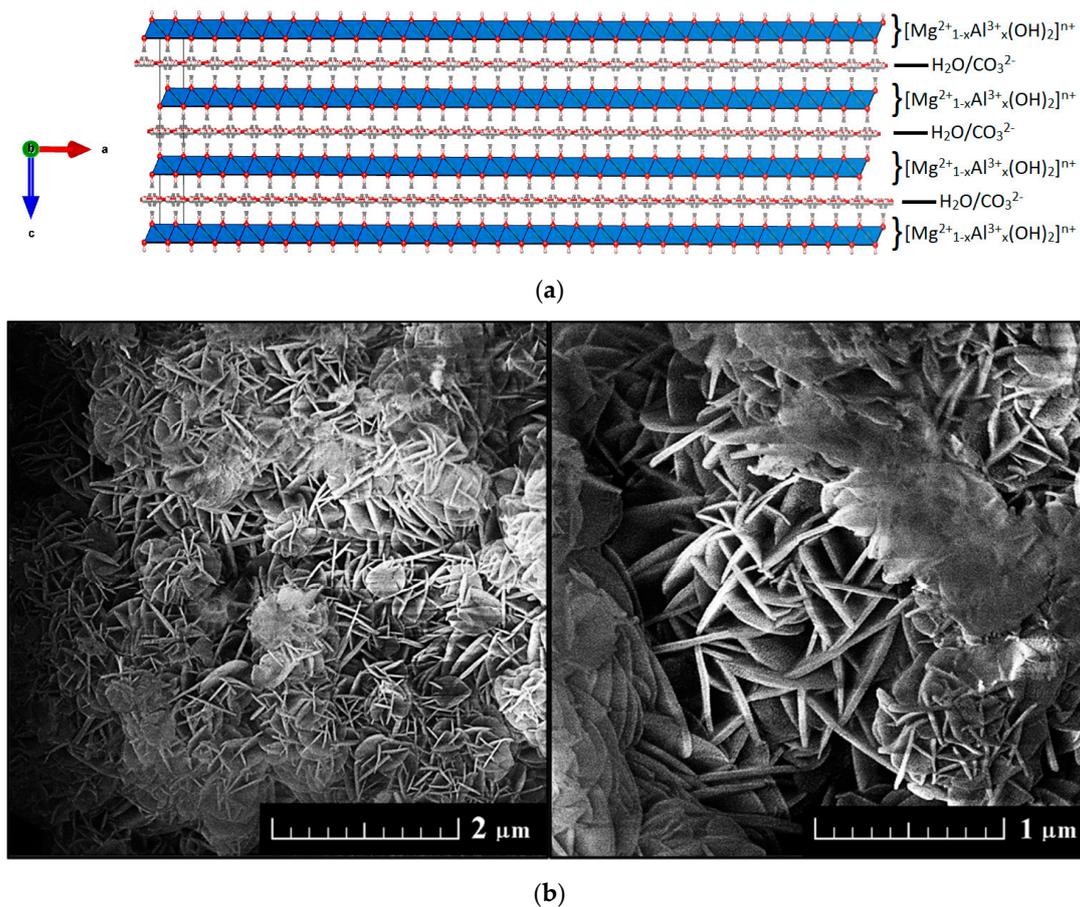
**Abstract:** Hydrotalcites are layered double hydroxides displaying a variety of stoichiometry caused by the different arrangement of the stacking of the layers, ordering of the metal cations, as well as the arrangement of anions and water molecules, in the interlayer galleries. The compounds of the hydrotalcite group show a wide range of the possible applications due to their specific properties, such as their large surface area, ion exchange ability, the insolubility in water and most of the organic sorbents, and others. Affordability, wide possibilities of manufacturing, and presence of sufficient natural deposits make hydrotalcites potentially very useful for the construction industry, as either a building material itself or an additive in mortars, concrete or in polymers composites used in constructions. Similar possible application of such material is in leakage control in a radioactive waste repository. The effect of use of these materials for ion exchange, anti-corrosion protection, radioactive ions containment, and similar purposes in building materials is examined in this review.

**Keywords:** hydrotalcite; layered double hydroxides; building materials; concrete; cement

## 1. Introduction

Hydrotalcites are layered minerals of both natural and synthetic origin, which are structurally derived from the brucite mineral-Mg(OH)<sub>2</sub>. Generally, they consist of three layers: two outer layers having a positive charge, and an inner intermediate layer which contains water molecules and replaceable anions (see Figure 1a). In brucite, the ions are octahedral, the central atoms are divalent magnesium cations, and the octahedral peaks fill the hydroxyl groups. Mineral hydrotalcite Mg<sub>6</sub>Al<sub>2</sub>(OH)<sub>16</sub>CO<sub>3</sub>·4H<sub>2</sub>O is the first discovered [1] and the best-known representative of layered double hydroxides (LDHs). The typical SEM microstructure is shown in Figure 1b.

The chemical composition of LDHs is not limited exclusively to Mg and Al atoms but can be described by the general formula, (M<sup>2+</sup><sub>1-x</sub>M<sup>3+</sup><sub>x</sub>(OH)<sub>2</sub>)<sup>x+</sup>(A<sup>n-</sup><sub>x/n</sub>yH<sub>2</sub>O)<sup>x-</sup>, where some divalent cations M<sup>2+</sup> are isomorphically replaced by trivalent M<sup>3+</sup> cations. Originally, electroneutral hydroxide layers thus obtain a positive charge, which is compensated by the charge of anions A<sup>n-</sup> located in the interlayer together with the molecules of crystalline water. The value of x in the above formula is equal to the molar ratio of trivalent cations in the hydroxide layers, M<sup>3+</sup>/(M<sup>2+</sup>+M<sup>3+</sup>), and usually, lies in the range between 0.20 and 0.33 [2].



**Figure 1.** Typical structure of hydrotalcites (a) and SEM images of the hydrotalcite sample with typical morphology (b).

Hydrotalcites have found the greatest practical use in the production and processing of polymers, especially as part of thermal stabilizing compounds for the processing of PVC, neutralizing additives, and flame retardants [3–5]. They find a very wide range of applications in heterogeneous catalysis, particularly as precursors for the synthesis of catalysts based on mixed oxides. They can be also used in sorption and decontamination processes or for the intercalation of various substances, including drugs. The most common minerals of the hydrotalcite group are listed in Table 1.

**Table 1.** Minerals of the hydrotalcite group.

Type of Hydrotalcite	Cations	Space Group	Reference
Hydrotalcite-2H	Mg <sup>2+</sup> , Al <sup>3+</sup>	P6 <sub>3</sub> /mmc	Bookin, A.S. et al. [6], Zhitova, E.S. et al. [7]
Hydrotalcite-3R	Mg <sup>2+</sup> , Al <sup>3+</sup>	R $\bar{3}$ m	Mills, S. et al. [8], Zhitova, E.S. et al. [7]
Desautelsite	Mg <sup>2+</sup> , Mn <sup>3+</sup>	R $\bar{3}$ m	Dunn, P.J. et al. [9]
Droninoite	Ni <sup>2+</sup> , Fe <sup>2+</sup>	R $\bar{3}$ m	Chukanov, N.V. et al. [10]
Iowaite	Mg <sup>2+</sup> , Fe <sup>2+</sup>	R $\bar{3}$ m	Braithwaite, R.S.W. et al. [11], Kohls, D.W. et al. [12]
Meixnerite	Mg <sup>2+</sup> , Al <sup>3+</sup>	R $\bar{3}$ m	Mills, S. et al. [8], Koritnig, S. et al. [13]
Pyroaurite-2H	Mg <sup>2+</sup> , Fe <sup>3+</sup>	P6 <sub>3</sub> /mmc	Ingram, L. et al. [14], Frondel, C. [15]
Pyroaurite-3R	Mg <sup>2+</sup> , Fe <sup>3+</sup>	R $\bar{3}$ m	Ingram, L. et al. [14]
Reevesite	Ni <sup>2+</sup> , Fe <sup>3+</sup>	R $\bar{3}$ m	Song, Y. et al. [16], Taylor, H. F. W. [17]
Stichtite-2H	Mg <sup>2+</sup> , Cr <sup>3+</sup>	P6 <sub>3</sub> /mmc	Mills, S. et al. [18], Ashwal, L.D. et al. [19]
Stichtite-3R	Mg <sup>2+</sup> , Cr <sup>3+</sup>	R $\bar{3}$ m	Mills, S. et al. [18], Ashwal, L.D. et al. [19]
Takovite	Ni <sup>2+</sup> , Al <sup>3+</sup>	R $\bar{3}$ m	Bish, D. L. [20], Mills, S. et al. [8], Maksimović, Z. [21]
UM1998-10-CO:CoHNi	Ni <sup>2+</sup> , Co <sup>3+</sup>	R $\bar{3}$ m	Song, Y. et al. [16]
UM2002-02-COH:FeNi	Ni <sup>2+</sup> , Fe <sup>3+</sup> , Fe <sup>2+</sup>	R $\bar{3}$ m	Smith, D.G.W. et al. [22], Maksimović, Z. et al. [23]
Woodallite	Mg <sup>2+</sup> , Cr <sup>3+</sup>	R $\bar{3}$ m	Mills, S. et al. [24]
Muskoxite	Mg <sup>2+</sup> , Fe <sup>3+</sup>	R $\bar{3}$ m	Jambor, J.L. [25]

## 2. Synthesis of Hydrotalcites

Hydrotalcite minerals with  $Mg^{2+}$ ,  $Fe^{2+}$ ,  $Mn^{2+}$ ,  $Zn^{2+}$ ,  $Cu^{2+}$ , and  $Ni^{2+}$  anions in the position of  $Mg^{2+}$  ions are often found in natural geological deposits [26] and therefore can be obtained from such sources. However, sometimes it is necessary to synthesize hydrotalcite group materials to achieve sufficient purity or other desired properties.

The most used method of hydrotalcite synthesis is a coprecipitation of two metal salts in alkaline solution at a constant pH value of approximately 10. A typical synthesis of a hydrotalcite employs a variation of a hydrothermal method, which often needs optimized reaction conditions to obtain desired product quality [27–31]. In addition, the hydrolysis reaction is often used [32, 33], as well as the sol-gel method [34,35]. The co-precipitation method is usually based on the simultaneous precipitation of inorganic salts from basic solutions at constant or at increasing pH [36]. The combustion method of synthesis can be advantageous because it involves a very rapid chemical process, the explosive decomposition of organic fuels, e.g., glycine or urea. Carbon dioxide and water formed during the combustion are employed in producing complexes with the metal ions [37,38]. Hydrotalcites obtained by the combustion process can be heated until the crystal lattice is destroyed, and subsequently recrystallized in a carbonate aqueous solution [39]. Sulphate-intercalated hydrotalcites for ammonia-nitrogen removal can be prepared using microwave-assisted synthesis. In this procedure the microwave irradiation contributes to the intercalation of sulphate anions to high crystallinity of the resulting material [40]. Another possibility is the preparation of hydrotalcites from industrial wastes. For example, the preparation of hydrotalcite from coal-combustion fly ash was explored. The synthesis was carried out as it is further described. Three molarity Three mol/L leaching solution of HCl, 30 min of aging at 65 °C and pH of 11.5, followed by crystallization for 12 h at 70 °C. Prepared hydrotalcite has a large external surface area with low microporosity, and it consists of sub-micron plate-like particles. The structural features of hydrotalcite prepared from fly ash were comparable to the hydrotalcites synthesized from pure raw chemicals with the exception of the presence of calcite [41]. The hydrotalcite-like compounds were also prepared by calcination and subsequent pozzolanic reaction from paper sludge from the newsprint company, using only recycled paper as a raw material [42]. Similarly, the hydrotalcite-like compounds were synthesized by the hydration reaction of activated art paper sludge-lime mixtures at 20 °C [43].

## 3. General Application of the Hydrotalcites

There are various possible applications of hydrotalcites. Due to the layered structure, hydrotalcites have a large surface area. They have great ion exchange ability and also, they are insoluble in water and most of the organic sorbents. Therefore, these materials, most commonly Mg-Al hydrotalcite, are suitable as basic heterogeneous catalysts for fine chemicals' synthesis. Due to their low reactivity, the as-prepared hydrotalcites must be activated before use as the catalysts. The first activation step consists of thermal decomposition of as-prepared Mg-Al hydrotalcite (see Figure 2).

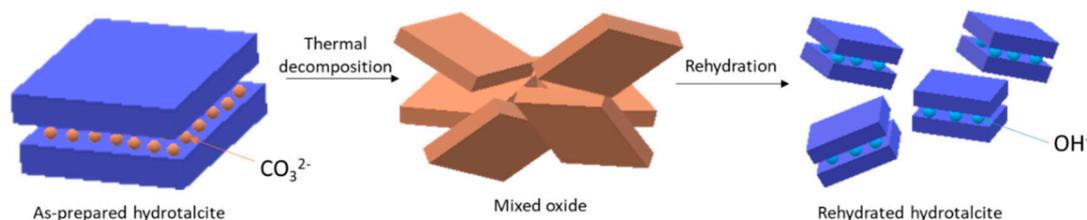


Figure 2. Hydrotalcite activation for catalysis.

Calcination of as-prepared Mg-Al hydrotalcite below 550 °C results in the formation of a well-dispersed mixture of magnesium and aluminum oxides (mixed oxide, MgAlO) with the presence of  $Mg^{2+}$ - $O^{2-}$  acid-base pairs (Lewis-type basicity). Mg-Al mixed oxide has a so-called “memory effect”, which allows the reconstruction of the original layered structure in contact with water.

Therefore, the second activation step consists of rehydration of mixed oxide leading to hydrotalcite intercalated with  $\text{OH}^-$  ion as compensating anions in the interlayer [44], which are Brønsted-type basic sites. These rehydrated hydrotalcites exhibit high activity in base-catalysed reactions which require Brønsted basicity, i.e., condensation reactions of aldehydes and ketones [45–51], Michael addition [52], Baeyer-Villiger oxidation [53,54], and others.

Hydrotalcites are also quite promising heterogeneous catalysts which enable the multistep processes, particularly when different and incompatible active sites are required [55]. Metal containing hydrotalcites are found to be very useful in many important industrial reactions, such as aldol condensation, followed by hydrogenation [56,57].

The complex structure of hydrotalcites, enabling complexation of intercalated anions with various charged species, can be used in wastewater cleaning applications. For example, Mg-Al hydrotalcites can be used as adsorbents for the removal of chloride ions from wastewater [58]. In addition, Fe-Mg-Al hydrotalcites with different intercalated anions were evaluated as selective adsorbents for capturing heavy metals from a complex aqueous environment [59]. Adsorbents derived from hydrotalcites can be used for the removal of acute toxic drug diclofenac in wastewater [60], while the usual treatment processes are not able to totally eliminate this compound. Moreover, Zn/Al layered double hydroxides can be used for the removal of nanoscale plastic debris from aqueous systems [61].

A medical application of hydrotalcite materials is also common, e.g., it can be used as an antacid. Usually, it is used together with aspirin without losing the medical effectiveness of both drugs [62]. Additionally, the hydrotalcites can be synthesized with bactericidal metal ions incorporated into the structure and used in cotton fibres for medical textile supplies [63]. Moreover, the hydrotalcites can be used as a raw material for various supplies, e.g., for black ceramic pigment [64].

Hydrotalcites also show very promising results in the ion-exchange technologies [65–68], where especially the anion exchange preference can be adjusted by different synthesis methods or anion occlusions [69]. The exchange properties are mainly affected by the interlayer anions [70].

Another very extensively studied problem, where the usage of hydrotalcites has great potential, is the  $\text{CO}_2$  and  $\text{H}_2\text{O}$  capture [71,72]. In contrast to the materials which are widely used for the  $\text{CO}_2$  capture, such as zeolites or activated carbon, hydrotalcites show stable adsorption capacities at high temperatures [73,74]. This is a very important property used in the steam reforming processes [75], but also for the removal of carbon dioxide from power plant fuel gases and for the reduction of total carbon dioxide emissions [76,77].

#### 4. Hydrotalcites as a Part of the Building Materials

Hydrotalcites as mineral substances, which are abundant and can be potentially easily produced on a large scale, came to consideration as a potential building material or appropriate additive in such materials. Because of their unique properties and low cost, they find usage especially as part of mortar and cement additives, where they prevent the chloride corrosion of concrete. Additionally, their sorption ability of dangerous material and usage in nuclear waste repositories, as well as application as a component of polymer composites, is very interesting, and it has been previously thoroughly studied [78–81]. The directions of application of hydrotalcites in the construction industry are described in the scheme (Figure 3).

##### 4.1. Application in Radioactive Waste Repository

The ion exchange ability of the hydrotalcites may be utilized in special materials designed for containing ions of dangerous elements. Thermally treated hydrotalcites were examined for the sorption of the radioactive isotopes of iodine in the anion form. In the sample calcined at 773 K with the destroyed crystal lattice, increased sorption of  $\text{I}^-$  anion was found, and hydrotalcite structure was reconstructed [82]. In addition, hydrotalcites are considered as possible material for anion scavengers for low-level radioactive waste repository backfills at near neutral pH for both  $\text{I}^-$  and  $\text{TcO}_4^-$  anions [83].

Since hydrotalcites may be incorporated into various building material mixtures, mortars, concretes, and backfills, their application as accessible and affordable materials is prospective.

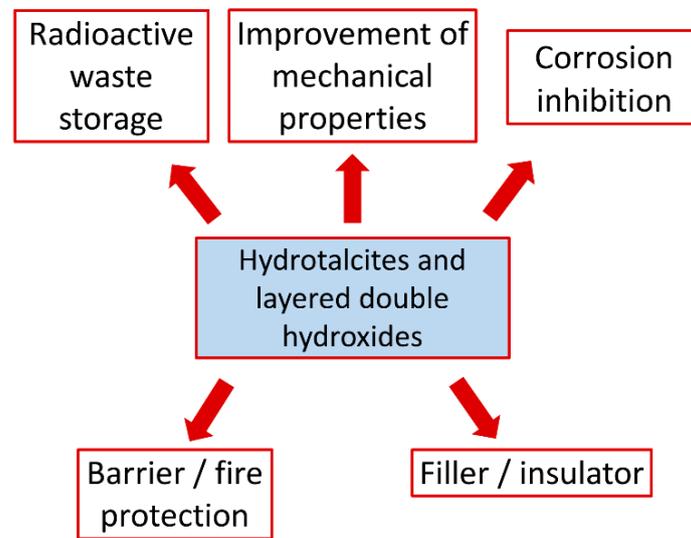


Figure 3. Scheme of application of hydrotalcites in building materials.

#### 4.2. Hydrotalcites in Reinforced Concrete as Corrosion Inhibitive Addition

The main problem in reinforced concrete structures is the corrosion of the steel reinforcement, especially in coastal environments [84–86]. Usually, the reinforcement steel in solid concrete structure is passivated due to the high pH and therefore does not tend to corrode (see Figure 4) [87]. Nevertheless, in the presence of a sufficient level of chloride ions, the inner steel reinforcement can be de-passivated so the corrosion of the structure may start. This may occur especially after the exposition of the material to the seawater or in general, in the near-sea environment. The overall expenses of corrosion of reinforcement are considerable, as it involves repairing, restoration, and the safety monitoring activities, as well as aesthetics of concrete structures and components. Hence, it is vital to project concrete resistant to chloride corrosion.

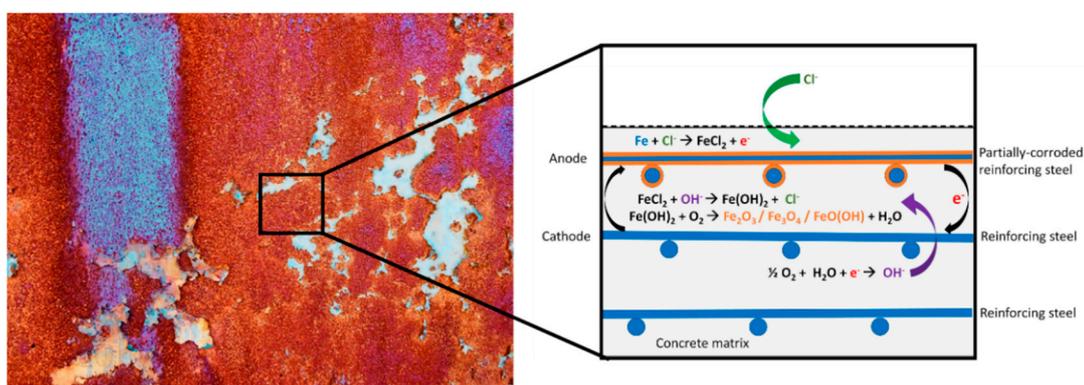


Figure 4. Scheme of the corrosion process.

In these days, there are several approaches of preventing the corrosion of concrete reinforcement. The first one is the use of surface coatings, which act as a barrier resistant to the chemical agents which can affect the concrete. The most common materials used for the coating are epoxy resin, polyurethane, acrylic coatings, polymer emulsion coatings, and chlorinated rubbers. The most important aspects which are being studied are their ability to adhere to the concrete, chloride permeability, the crack-brinding ability, and resistance to moisture and temperature variations. Previous studies showed high applicability

of epoxy resin, which exhibits a good adhesion to concrete, as well as quite high effectiveness in reducing the water absorption causing the reduction of damage of the concrete reinforcement [88–90]. Another way of the anti-corrosion protection of concrete and concrete reinforcement is use of admixtures. The possible materials which can be used as an admixture are waste materials, such as coal fly ash or polymers, in the form of particles, short fibers, or organic liquids. Such materials not only work as an anti-corrosion agents but also improve the mechanical properties [91,92]. Another possible sort of material which can be used as an admixture are the hydrotalcites.

Hydrotalcites can be easily applied in some strategies of corrosion protection of concrete steel reinforcements [93,94]. The results of experiments showed that some configurations of Al-Mg hydrotalcites, with carbonate anions in the raw hydrotalcites substituted by different anions (e.g., p-aminobenzoate, pAB), displayed better corrosion inhibiting result in extended time before the beginning of the corrosion. It also showed a higher chloride threshold in comparison to the same free ions in solution. The mechanism is based on ion exchange between free chloride ions and the intercalated inhibitive anions in hydrotalcite matrix and simultaneous release inhibitive anions. These anions act as active inhibitors, causing the increase of a threshold concentration of chloride ions. The anionic exchange capacity of hydrotalcites is quite high (2–4.5 miliequivalents/g), making the exchange of interlayer ions easily achieved [95,96]. In addition, the influence of the addition of the anticorrosion material on mechanical properties is important. This statement was already previously mentioned in the literature [97,98]. The addition of 5% of pAB hydrotalcite in mortar specimens resulted in considerably improved resistance of the material to the chloride diffusion, accompanied with small decrease of compressive and flexural strength in units of MPa. Another promising way, besides the mentioned 5% mass substitution of cement by pAB hydrotalcite in the bulk mortar, is the coating of reinforcing steel by part of cement paste coating 20% pAB/NO<sub>2</sub> [99]. Additionally, relative Mg-Al layered double hydroxides intercalated with hydroxide and organic anions can be applied as the corrosion inhibitors for carbon steel reinforced concrete [100]. However, the costs of hydrotalcite-like materials is quite high compared to the commonly used anti-corrosive materials.

#### 4.3. Hydrotalcites in Cements and Mortars

An interesting application of hydrotalcites is in isolation of dangerous heavy metals in cement prepared from industrial waste. Hydrotalcites can be used for double barrier technique effect in cement-based mortars prepared for the immobilization of lead contained in electric arc furnace dust. Double barrier mortars with dimercaptosuccinate in the interlayer of the hydrotalcite reduced lead release by approximately 50% in comparison with the conventional immobilization mortar [101].

Hydrotalcites often form in mortars and cements, where supplementary cementitious materials are used as a substitute for cement clinker in order to reduce the emissions of cement production. This replacement was studied recently by using magnesium oxide [57], metakaolin [102], or dolomite [103]. The presence of alumina from a raw aluminosilicate may improve the mechanical behavior of resulting mortars. It may interact with hydrated magnesium oxide and form a hydrotalcite-like phase [58], while effectively improving the mechanical properties by an enhanced bond with the cementitious matrix [59]. In addition, a material containing hydrate water molecules can decrease the materials' overall porosity [60].

Hydration kinetics can be also modified using hydrotalcites, since it is believed, that the addition of nanoparticles lowers the energy barrier of liquid-state precipitation reactions. Hydrotalcites also provide additional nucleation sites, which are essential for the cement hydration reactions [104,105].

The presence of hydrotalcites in concrete often affects the values of the compressive strength [106]. Many research studies show an increase of compressive strength, varying from units of percents to tens of percents [107,108]. Zou et al. referred to a 46,2% increase in compressive strength, which was attained by the addition of 1% LiAl layered double hydroxide [109]. However, it has been also reported that the presence of hydrotalcites leads to opposite effects [101]. Yang et al. used a 10% addition of a MgAl NO<sub>2</sub> layered double hydroxide, which led to the decrease of compressive strength by 14,2% and

the flexural strength by 19,1% [110]. Xiong et al. observed a 25% compressive strength decrease after the addition of 1–3% of MgAl layered double hydroxide [111]. The differences between the positive and negative influence on the compressive strength depend on the microstructure of both the hydrotalcites used and the concrete. The usage of inappropriate hydrotalcites may cause the increase of porosity, which can also lead to faster ingress of chlorides or other undesired substances [111]. This porosity is also growing with the amount of hydrotalcites added to the concrete [112]. Additionally, the presence of hydrotalcites can lead to an increase in the compressive strength [106].

Hydrotalcites were also studied as additives in slag cements, where their presence allows to control kinetics and formation of the C-A-S-H phase [113] and also helps binding chlorides [114]. Another approach to hydrotalcites in cementitious materials was shown in the study of Raki et al., where the hydrotalcites were used as hosts for intercalation of organic admixtures in nanocomposites [115].

Hydrotalcites are also present in non-cement binders, such as in ground granulated blast furnace slags, where they are beside the C-S-H and  $\text{Ca}(\text{OH})_2$  phase part of the reaction products, which define the strength of the material [116].

#### 4.4. Hydrotalcites in Polymer Composites

Polyurethane foam is one of the most important thermosets widely used as an insulating material in the building and construction industry. Previously, it was shown that rigid foam composites with polyether polyol, isocyanate, a fire-retarding agent, hydrotalcite, and a PET can be formed [117]. It was shown that a small amount of hydrotalcite increases the compressive stress, but excessive hydrotalcite adversely affects the compressive stress. Additionally, flexible polyurethane foam nanocomposites with modified layered double hydroxides were studied [118]. In another paper, the effect of the addition of a phosphorus-containing polyol (E560) and hydrotalcites or another layered double hydroxides (LDH) to flexible polyurethane foams synthesized with a castor oil based polyol (LB50) was reported [119]. Interesting flame-retardant properties were observed for composites formed from hydrotalcites and tris (1-chloro-2-propyl) phosphate and isocyanate-based polyimide foams [120]. Preparation of hydrotalcites and isostructural layered double hydroxides and also their applications as additives in polymers was also described in detail. The use of hydrotalcites as heat retention additives in horticultural plastic films and as flame retardants was described. Hydrotalcites and other LDH can be also used as stabilizing agents for PVC or other polymers and for the stabilization of pigments in polymers [121].

## 5. Conclusions

The possible applications of the group of double-layered mineral hydroxides, called hydrotalcites, in building materials was reviewed. Hydrotalcites are suitable for various applications in building materials, such as mortars, cements, and polymer-composite materials due to their unique properties, namely ion exchange ability, large surface area, the insolubility in water, and, last but not least, their availability. Ion exchange properties can be utilized in anticorrosion protection of steel in reinforced concrete structures and for the storage of dangerous leakages from various waste dumps, including radioactive waste repository. Its mineral nature enables their utilization as flame retardants in polymer composites. Low price and high-availability allow its application as a quality cement clinker, which may result in reducing the emissions of cement production.

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