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A Review on Emerging Applications of Natural Sericite and its Composites

[1]D. Harikishore Kumar Reddy, [1]Seung-Mok Lee, [2]Jong-Oh Kim

[1]Department of Environmental Engineering,
Kwandong University, Gangneung, Gangwondo, Republic of Korea-210 701
[2]Department of Civil Engineering, Gangneung-Wonju National University,
Gangneung, Republic of Korea, 210-702

Abstract: In the rapidly evolving field of material engineering, clay minerals represent the key constituents to explore and harvest the potential of economically-viable and environmentally-friendly hybrid composites. In light of recent developments for the preparation of pigments, flame retardants, polymer composites, health care products and materials for wastewater treatment, sericite and its composites have been widely studied. Sericite is a clay mineral that is abundant on alarge scale in nature. Its abundance and low cost mean that sericite is can be utilized as an effective raw material for the production of various industrial products. Thus, in this review, an overview of the current state of sericite and its composite materials, the properties of sericite and intended applications were presented. In addition, this review outlines research trends and prospects for the future are briefly discussed.

Key words: Clays · Sericite · Polymer composites · Pigments · Wastewater treatment

INTRODUCTION

The increase of population and rapid industrialization demand various materials for industry, infrastructure, transportation and human needs. However, economic imbalance throughout the world leads to the search for low-cost and easily available materials. Naturally available materials always attract special attention due to their ease of availability, abundance in nature, environmentally-friendly properties and are economic. Among other natural resources available, clay materials have attracted industry and academia. Over the last few years, the science of clay minerals has generated great interest in a variety of scientific communities, ranging from chemical, materials, environmental science to engineering. A literature survey has been carried out using the Scopus© database from the years 2001-2011, with the keyword "clay" used in the search for results. A linear growth rate was observed in the number of articles published over the past three years (Figure 1).

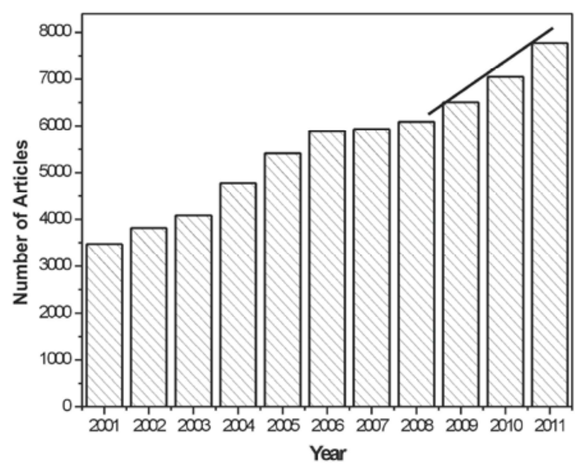

Fig. 1: A Scopus database literature survey of annual published journal papers with the term "Clay" published from 2001 to 2011.

Clays are nature's gifts-wide spread, highly abundant and filled with low-cost chemical substances. They are important groups of minerals because they are among the most common products of the chemical weathering of rocks. Clays are well known and very familiar to mankind from the earliest days of civilization, as they were the raw materials used to make china and porcelain. Clay minerals are phyllosilicates which usually form as a result of

Corresponding Authors: Seung-Mok Lee, Department of Environmental Engineering, Kwandong University, Gangneung, Gangwondo, Republic of Korea-210 701. Tel: +82 33 649 7277.

chemical weathering of other silicate minerals atthe surface of the Earth. The composition and morphology of natural clays may vary from one location to another, depending on their geological origin [1]. These materials are used for numerous applications, such as adsorption, healthcare, catalysis [2-6] and many others. Clay minerals with negative charges gained special attention because they can be neutralized by inorganic exchange cations, replacement of these inorganic exchange cations with quaternary ammonium cations by simple ion exchange reactions results organo-clays with organophilic clay surfaces [7-8]. These organo-clays were used as sorbents to remove various toxic contaminants from aqueous solutions [9-10]. In addition, the pillared interlayer spaces of clay minerals by inorganic or organic species can also act as carriers to immobilize the host reactions, acting as adsorbents for remediation of pollutants or nuclear wastes [11], catalysts [12] and enzyme electrodes [13]. In recent years, clay polymer nanocomposites have received great academic and industrial attention and are popular research topics [14,15]. This is due to the fact that inorganic particles in a polymer matrix could exhibit improved physical and mechanical properties of base polymers [16]. Both in their native states and in numerous modified forms, clays are versatile materials as they can be molded into any shape. Due to the aforementioned reasons, clays and their composites have gained renewed interest from science and academia, as well as industry, for their unique and interesting properties, providing novel functional organic/inorganic hybrid materials [17].

Sericite is a clay mineral of special interest because of its smooth molecular surface, acid-resistance, fireproofing and electrical insulating properties [18]. As a subspecies of muscovite occurring in large amounts in natural deposits, sericite has attracted many researchers working in different areas. Recently, sericite-based hybrid materials have attracted extensive attentions for both theoretical and practical purposes because of their environmental friendliness, weather durability and low cost compared to other materials. The unique class of these sericite-based hybrid materials can retain not only beneficial features of both inorganic and organic components, but can also allow researchersto systematically tune the properties of the hybrid materials through the combination of appropriate functional components.

Based on the above consideration, in this review, we describe the brief history and structural characteristics of sericite. We then discuss the development of sericite-based materials in various fields. The feasibility of using natural sericite and its composites in wastewater treatment is reviewed, along with the preparation of polymer composites and pigments. In addition the problems or major challenges that remain to be solved and future direction for future researchwas also discussed.

Sericite: Sericite is among the oldest known clay minerals and also one of the most abundant. Sericite is full of minerals, being used long ago as medicine in the East Asia. The use of "sericite" and description of its curative properties are recorded in ancient Korean and Chinese medical books. Sericite is a clay mineral in the muscovite family (it is the cryptocrystalline form of muscovite) is a lustrous, layered powder. Sericite is a petrographic term used to indicate highly refractive and fine-grained mica, found in hydrothermallyaltered rocks [19]. Sericite is a kind of phyllosilicate native mica mineral, usually white or near-white in color and it has similar characteristics to bentonite. Typically, sericite has a three-layer unit cell, consisting of two silicon-oxygen tetrahedrons and one aluminum/magnesium-oxygen octahedron. Between the layers, potassium ionic bonds hold the layers together. However, depending on its formation and location, sericite exhibits significant differences in composition. The elemental composition of sericite from various studies appears in Table 1.

Sericite has peculiar characteristics when compared with other clay minerals. Sericite is one of the rare, naturally swelling layered minerals that expand its volume when immersed in polar liquids and it possess a high degree of negative charges on its platy surfaces. The powder of sericite has some particular properties, such as acid- and alkali-resistance and chemical stability.

Table 1: Elemental composition of the sericite reported in literature

	SiO_2	Al_2O_3	K_2O	Li_2O	Na_2O	MgO	CaO	Fe_2O_3	TiO_2	SO_3	MnO	H_2O	Reference
wt.%	52.04	30.02	5.68	0.01	0.60	0.27	0.03	1.26	-	-	-	-	18
Concentration	69.5-79.9	12.0-15.6	3.29-5.55	-	0.10-2.00	0.15-0.66	0.07-2.22	1.68-3.22	0.12-0.26	0.00-0.08	<0.05	3.0-5.0	34
Actual-water washed sericite %	53.41	30.70	7.06		0.19	0.30	0.03	1.95					48
Composition%	70.12	17.97	6.07	-	0.14	1.36	0.27	0.71	-	-	-	-	73

When sericite is used as coating or filler for plastics, oil-based paint, rubber and so on [20-22], the mechanical intensity, tenacity, adhesion and anti-aging performance of the matrix are improved. Sericite powder particle contains layers (plates) of an expandable-layered structure. The flat surface of the sericite layer carries negative charges, while the edges are positively charged. It was reported that it is difficult to distinguish the sericite from kaolin and therefore it is often wrongly identified as kaolin or white clay, however, few characteristics of sericite likefineness and fat feeling, similar to talc- qualities that kaolin does not possess [23].

Applications of Sericite and its Composites

Polymer Composites: In the past decades, polymer composites have become a focus of research [24]. Among all the studied polymer composites, clay and layered silicates are the most widely used precursors [25-26]. This is due to the fact that clay materials are easily available and their intercalation chemistry has been studied for a long time [27,28]. For example, lamellar fillers, such as mica and clay, exhibit excellent abilities to improve the flexural modulus, heat deflection temperature, warping and gas barrier performance and theyreduce the product cost [29,30]. Functionalizing polyolefin to improve its compatibility with inorganic fillers or engineering plastics has been intensively studied to prepare high-performance polyolefin composites [31-33].

Accordingly, several researchers have prepared high-density polyethylene (HDPE)/sericite composites. In the perspective an attempt was made to prepare HDPE/sericite composites using a co-extrusion of HDPE with sericite and then study its oxidation profile [34]. The authors are highly credited for their studies on the effects of coupling agents on the prepared composite and also for their accurate characterization of these composites. A high degree of oxidation was observed with the HDPE/sericite composite. Also, in the natural aging of the HDPE/sericite composites, sericite acted like an oxidation catalyst. The authors also claimed that HDPE/sericite behaved as a sensitizer for the photo-oxidation reactions. In a similar work, the interfacial interaction of the HDPE composite with sericite was studied [35]. Ultraviolet (UV) irradiated HDPE was used in the preparation of HDPE/sericite composite. The UV-irradiated HDPE/sericite composite improved the dispersion and the interfacial interaction of sericite with the matrix; in contrast, the irradiated HDPE/sericite composites significantly increased tensile yield strength (27.1-30.6 Mpa). To advance polymer-clay nanocomposite science in 2008, a new polymer nanocomposite using potassium-sericite (K-SE) was synthesized [36]. The beauty of this research was exfoliation of the mica layer. The authors claimed there was a considerable exfoliation of the organically modified sericite. A year later, the same group prepared six exfoliated mica-polyamidenano composites using sericite [37]. The morphology of the nanocomposite revealed mica nanolayers with very high aspect ratios; that is, at levels about two times greater than that of conventional exfoliated clay-polymer nanocomposites. The authors concluded that the degree of exfoliation, aspect ratios and dispersion homogeneity of silicate platelets will play important roles in the development of high-performance nanocomposites. Wu, *et al*. [38] reported a novel starch-graft-polyacrylamide/clay superabsorbent in 2000. The composite was synthesized by the graft copolymerization reaction of acrylamide, potato starch and clay mineral, followed by hydrolysis with sodium hydroxide. However, lower water absorbency was observed for sericite compared with bentonite and kaolinite.

In a recent study, sericite/acrylonitrile butadiene styrene plastic composite material was prepared [39]. This polymeric composite was prepared with different amounts of silane-modified sericite, mechanism and properties of the resulting composite were evaluated. In another study hetero-aggregation of two minerals, chalcocite and sericite, using two anionic polymeric dispersants, pulp interfacial chemistry and mediate sericite-chalcocite particle interactions in aqueous dispersions, was studied [40]. Both polymers exhibited greater adsorption onto chalcocite and the adsorbed polymer layer dominated the interfacial chemistry, which showed subtle mineral phase and pH dependency. In a recent study, Chen, *et al*. [41] investigated the biodegradable properties of polylactide/sericite powder nanocomposites in a batch foaming method. They have extensively studied the various properties (thermal, rheological, particle dispersal) and compared them with microcellular polyactic acid/organic modified montmorillonite.

Pigments and Coatings: Recently, substrate-based inorganic pigments have attracted more attention because of their wide range of applications, including optical filters, cosmetics, plastics, inks and paints [42-45]. The substrate-based inorganic pigment, mica-titania is the most widely used pearlescent pigment [46], because coating of TiO_2 on the mica surface provides excellent chemical stability [47] and also resistance to weathering, sunlight and heat. In general, muscovite was conventionally used as a flat substrate in the preparation of the mica-titania pigmentmentionedabove. Unfortunately, natural muscovite depositsare limited and expensive.

Sericite is a low-cost material subspecies of muscovite, occurring in large amounts of natural deposits, thusit has attracted widespread attention in the preparation of pigments. Naturally abundant sericite clay has a platy structure, as well as swelling and delaminating characteristics. The pigments prepared from sericite have greater surface strength, faster ink drying times and improved opacity [48]. Based on the above considerations, sericite has been widely used in pigments byitself and with other materials. Therefore, preparation of mica-titania-based inorganic colored pigments by using rutile TiO_2-coated sericite has been extensively studied in recent years.

Ren and colleagues [47, 49] have extensively studied the preparation and characterization of various sericite-based inorganic pigments. Accordingly, in 2007 they endeavored to advocate an initial study examining the optical properties of sericite-titania composite. They prepared the composite by depositing TiO_2 nanoparticles on lamellar sericite. The authors reported that island-like structured rutile TiO_2 aggregates were strongly anchored and well dispersed on the flat surface of sericite. The whiteness, lightness and reflectance of TiO_2-coated lamellar sericite increased, along with increasing TiO_2 loading. One year later, the same group studied the pigment properties of TiO_2-coated sericite in the presence of La^{3+} cations [49]. TiO_2-coating of sericite was achieved by chemical deposition. According to the authors, the light scattering indexes of the TiO_2-coated lamellar sericite powders were dozens of times higher than that of the naked lamellar sericite powders. Also, TiO_2-coated lamellar sericite powders prepared in the presence of La^{3+} had a higher light scattering index than that prepared in the absence of La^{3+}.The presence of La^{3+} causes the formation of Si-O-La and Al-O-La bonds on the sericite surface and Ti-O-La bonding on the surface of TiO_2 coating layers. The yellowness of the TiO_2-coated sericite powders obviously increased and the brightness slightly decreased. In a similar manner, the pigment properties of the rutile TiO_2 coating layers on the lamellar sericite surface induced by Sn^{4+}were studied [50]. Based on XPS analysis, they found that Sn^{4+} cations were anchored tothe sericite surfaces by formation of Al-O-Sn and Si-O-Sn bonds and the aluminum and silicon sites on the sericite surfaces were dominantly occupied by Sn^{4+} cations. The presence of Sn^{4+} induced the phase transformation of amorphous to rutile TiO_2 and the brightness, whiteness and light scattering indexes of the rutile TiO_2-coated sericite powders were slightly higher than those of the anatase TiO_2-coated sericite powders.

In 2011 they continued their studies of lamellar sericite. They prepared three inorganic composite pigments by coating Fe_2O_3, Bi_2O_3 and $CoAl_2O_4$ on the sericite surface by the homogeneous hydrolysis of metal salts [51]. They achieved red, yellow and blue colored composites by coating Fe_2O_3, Bi_2O_3 and $CoAl_2O_4$ on the sericite surface. The authors reported that the inorganic coating layers were uniformly and tightly anchored to sericite surfaces, via chemical bonding at the interface between the coating layer and the sericite substrate. Furthermore, they prepared the inorganic colored composite pigments by hydrolysis of $FeCl_3$, $Bi(NO_3)_3$ and$Co(NO_3)_2/Al(NO_3)_3$ in the presence of a mica-titania substrate and calcination at different temperatures [52]. Red, yellow and blue colored pigments were achieved by coating Fe_2O_3, $Bi_4Ti_3O_{12}$ and $CoAl_2O_4$ on the mica-titania substrate surface. The color and color purity were tuned by changing the type, morphology and loading of the metal oxide coating layers.

The formation of a phospholipid vesicle coating on the surface of inorganic sericite with Mg^{2+} cation bridged phosphatidylcholine has been studied [53]. The stabilization properties of phospholipid vesicles on the sericite surface were demonstrated by the various concentrations of magnesium cation. The presence of magnesium was found to have a much more pronounced influence on the lipid deposition process. In another study, the authors described the formation of phospholipid vesicles on an inorganic sericite surface, characterized by electron microscopy (FE-SEM, TEM) and evaluated qualitatively by XPS as a function of the etching time [54]. The vesicle mobility on the surface was restrained through a chelation effect using magnesium cations; the degree of stabilization demonstrated at various concentrations of the magnesium cation showed that it had a much more pronounced influence on the lipid deposition process. Sericite was also studied as pigment for coating paper. In 2006, sericite-based pigment was prepared and the performance was evaluated [48]. The authors extensively studied its performance and they reported a notable water retention with substituted sericite. Another study examined partially or totally delaminated clay in a lightweight coating formulation with sericite [55]. The authors claimed that among the coated paper properties, the addition of sericite raised the opacity and brightness of the coated sheets, which decreased linearly with increasing sericite substitution. A notable swelling and delamination effect during preparation of the coating color was observed with sericite.

Wastewater Treatment: Heavy metal ions, aromatic compounds, pharmaceuticals and dyes are often found in wastewater as a result of their widespread industrial use [56-59]. These pollutants will cause health problems when they exceed the tolerance limits in water [60-61]. Hence it is essential to eliminate these contaminants before they discharge into soil and ground water. Several techniques have been employed for the treatment of wastewater; however, adsorption is one of the most commonly employed and efficient techniques for the removal of toxic pollutants using various kinds of adsorbents [62-63].

However, more attention has been focused on the development of low-cost and environmentally-friendly adsorbents for water treatment [64]. In this regard, the application of natural clays as alternative adsorbents in wastewater treatment has been widely studied due to various advantages: Low-cost, copious availability in nature, non-toxicity, a high specific surface area and a high potential of ion exchange for charged pollutants [65-67]. Sericite is a naturally available, low-cost material and the presence of hydroxyl groups on the surface of sericite made it an attractive material for the removal of various pollutants from water.

For instance, sericite was used as an adsorbent for the removal of Cu(II) and Pb(II) from aqueous solutions by using batch and column experiments [68]. Various influencing parameters like pH, contact time and ionic strength for the removal of metal ions were investigated. The adsorption equilibrium data was better fitted to the Langmuir isotherm model and the sorption capacities were found to be 1.674 mg g^{-1} for Cu (II) and 4.697 mg g^{-1} for Pb(II). The authors reported that the presence of hydroxyl groups on sillanol and aluminol in sericite was responsible for the adsorption of metal ions. In another study, an inorgano-organo sericite was prepared by using hexadecyltrimethylammonium bromide (HDTMA) and alkyldimethylbenzylammonium chloride (AMBA). It employed these materials for As (III) and As(V) removal [69]. Newly-prepared hybrid materials were characterized by using FTIR, SEM and XRD studies. From the sorption experiments, it was reported that adsorption efficiency of both hybrid materials was higher than sericite.

In a recent study, Kwon and Jeon [70] studied the removal of Ni(II) from an aqueous solution using sericite. A high adsorption capacity of about 44 mg/g at pH 7.5 was reported for the removal of Ni(II) using sericite. They also reported that sericite had higher removal efficiency compared with Amberlite IR 120 plus resin. Furthermore, in a recent study using sericite,cesium was removed from an aqueous solution [71]. The authors reported a maximum adsorption capacity of 6.68 mg/g. In another study, adsorption of Mn^{2+} into manganese-coated sericite (MCSe) was investigated [72]. The amount of Mn^{2+} adsorbed by manganese-coated solids increased as the solution pH increased from 4.0 to 11.0 pH. The data fallowed the Freundlich isotherm. Mn^{2+} breakthrough time in the effluent from both MCS and MCSe columns in the presence of hypochlorite was greatly enhanced. According to the authors, this adsorbent is better for removal of Mn^{2+} contaminated water. Removal properties of As(III) and As(V) by several metal oxides having different mineral types, including sericite,were investigated in a recent study [73]. The authors reported a negligible adsorption of As(III) and As(V) by sericite.

The adsorption of Ba^{2+}, Pb^{2+}, Cu^{2+} and Zn^{2+} on layer silicates was studied [74]. For this purpose, in an initial experiment, the authors studied the external surface charge of materials, including sericite, using X-ray photoelectron spectroscopy. They found a low outer surface charge for sericite compared with all other mica and illites. Due to this result, they have not studied the application of sericite for the adsorption of the aforementionedmetal ions. Three organo-modified minerals, including sericite as an adsorbent for the removal of organic contaminants, such as benzene, phenol and toluene, were studied [75]. Adsorption equilibrium was described by the well-known Langmuir isotherm model. Sericite-based materials exhibited a lower adsorption compared with montmorillonite-and zeolite-based materials.

It was observed that sericite and its composites exhibited comparable adsorption capacities for different kinds of pollutants. A comparison of efficiency thatsericite exhibited withvarious pollutants are summarized in Table 2. The complex adsorption mechanism involved in the removal of pollutants with sericite was due to the surface silinol and aluminol hydroxyl groups and also due to the ion-exchange mechanism.Sericite has two different surface-active groups:(i)=S_1-OH of the silanol group; (ii)=S_2-OH representing thealuminol group [68, 69]. The deprotonated

Table 2: Summary of the advances in application of sericite based materials in removing different contaminants from water 1

Contaminants in water	Sericite used and modification	Effecting variables	Efficiency	Reference
Copper(II)	Sericite without modification	pH, time, Initial concentration and ionic strength	1.674 mg g^{-1}	68
Lead(II)		pH, time, initial concentration, and ionic strength	4.697 mg g^{-1}	
Arsenic(III)	Hexadecyltrimethylammonium bromide (HDTMA) and Alkyldimethylbenzylammonium chloride (AMBA) modified sericite named as AA and AH	pH, initial concentration,background electrolyte	0.338 for AA and 0.433 mg/g for AH	69
Arsenic(V)		pH, initial concentration, background electrolyte	0.541 mg/gfor AA and 0.852 mg/g for AH	
Nickel(II)	Natural sericite	pH, dose, Time, temperature,	44 mg/g	70
Cesium(I)	Natural sericite	pH, concentration, Time,	6.68 mg/g	71
Manganese(II)	MCSe	pH, concentration, temperature	13.83 g/Kg	72

surface $=SO^-$ behaves as a Lewis base, which bindswith the metal ion. This adsorption of metal ions can be interpreted as a competitive complex formation. The metal and sericite interactions aregiven as follows:

$$\equiv S_1 - OH + M^{2+} \rightarrow \ \equiv S_1 - O - M^+ + H^+ \quad (1)$$

$$\equiv S_2 - OH + M^{2+} \rightarrow \ \equiv S_2 - O - M^+ + H^+ \quad (2)$$

Where M = Cu(II), Pb(II) and Ni(II)

Biological Applications: The use of clay minerals for medicinal purposes is as old as civilization [76]. The use of sericite for medicinal purposes is almost as old as the recorded history of the ancient Chinese and Koreans. Traditionally, sericite has been used in alleviating pain in the reproductive organs of Korean women [77].

Since the properties of far-infrared (FIR) rays emitted from sericite ceramics are known, they have been used in the medical field to promote health; there are numerous reports that beds made from sericite ceramics help relieve dysmenorrheic pain [78]. In a recent study, the efficacy of a FIR-emitting sericite belt in the relief of dysmenorrhea and in the reduction of an analgesic has been successfully applied [78]. The authors claimed that hot packs were used to heat the ceramics. Slight pain relief in both groups was reported and the FIR-emitting sericite belt with a hot pack was an effective and safe treatment option,offering a prolonged effect for the management of primary dysmenorrhea. Recently, Park, *et al*. [79] studied the antibacterial properties of a mix of minerals, including sericite, talc and halloysite, all from Korea. The antibacterial effects onseveral gram-positive and gram-negative bacteria were examined, using liquid growth inhibition assays and a colony-counting method. The ore mixture showed a reduction in the growth of all bacteria; however, the mechanism was not investigated.

Other Applications: Apart from the aforementioned applications, few other studies haveendeavored to explorethe properties and various applications of sericite. In the past decade, clay composites for use as flame retardants have gained increasing interest [80, 81]. In view the importance of clays as fire retardants, Chou, *et al*. [82] studied the preparation and characterization of the intumescent fire retardant coating. They have compared the flame-retardant properties of sericite with the graphite/sericite mixture. They reported that sericite can be useful as natural flame-retardant filler when obtained from the simple and conventional process of mining.

CONCLUSIONS

Many advantages are associated with sericite and the major advantage is its availability in nature on alarge scale, thus the cost of sericite is low. Its medicinal properties, environmentally-friendly nature, low cost and the possibility to modify it into different composites make it a core material for several industrial applications. The low-cost nature of this material will attract more scientists working in the field of wastewater treatment. It is interesting to note the naturally available mineral Sericite is a promising material for a number of commercial applications. The use of sericite for pigments and coatings, for the adsorption of pollutants, for biological applications and for flame retardants is widespread andvery attractive. In addition, the environmental applications of sericite-based materials seem to be endless. The low-cost and environmentally-friendly sericite materials will gain more interest through processes that are cleaner rather than environmentally-damaging.

In addition few disadvantages are associated with sericite; the material has not been studied enough in order tofully understand its structural and mechanical features. The modification process also seems difficult, according to various reports. Most of the recent achievements are still based on laboratory-level tests. Many issues may need to be solved for material production and field applications. In addition, further insights into the mechanism and modification of sericite are required for further enhancement of sericite-based materials. These limitations have to be overcome through future studies in order to increase and improve the applications of this material.

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