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Empirical Study on Weather Resistance of White Artificial Stones in Subtropical Island Climate

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Abstract: White building components, whether decorative or the overall structure, are susceptible to extreme weathering conditions, which affect the durability of the building's external surface. In particular, white natural stone materials can provide buildings with beauty, durability, and sustainability, but this beauty is affected by key factors determining their weather resistance, namely physical properties such as porosity and acid and alkali resistances. As indicated by a past study, marble used as the exterior wall of a building in a similar environment will exhibit tarnishing after six months of weathering. Taiwan is a subtropical island, so the weather resistance of building materials to this environment is worthy of attention. As pointed out by the study, raw stone materials containing zirconium and silica can have greater weather resistance, do not easily change color, and possess good stress resistance properties. Focusing on this, this study attempted to identify stone materials with such compositions and sent samples to SGS (Societe Generale de Surveillance S.A., New Taipei City, Taiwan) for testing of weather resistance. This paper uses Spanish artificial stones for the study and observes the changes in the surface cleanliness on two buildings in Yilan County and Taipei in Taiwan after exposure to sun and rain. The experiments were conducted over two years. The study results showed that the artificial stones displayed no change in outward appearance under both rainy and acid rain environments. In Yilan, construction of an exterior wall was carried out in February 2018. The exterior wall used the white artificial stone directly from the manufacturer, and the black artificial stone with added paint for stone protection. After two months, the black artificial stone covered with paint exhibited a change in color and stains appeared on the surface, while the white artificial stone not covered with paint maintained the same cleanliness after two years. In Taipei, white Spanish artificial stone was used for the exterior wall of an entire building block in February 2020. After the frame was dismantled in September 2020, the color was found to be unaffected, being as pure white as it was when new. Therefore, this study selected Spanish artificial stone for the design of a building's exterior wall, as the cleanliness of the stone surface is not affected and it has good applicability in rainy and acid rain island environments.

Keywords: building façade; artificial stone; imitation stone; weather resistance; color change; natural stone; durability

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1. Introduction

The quarrying, transportation, and cutting processes for natural stone materials are quite difficult, which leads to the extension of the construction period and the increase in construction cost. Thus, the use of artificial stone can enhance the aesthetics of stone materials and control the cost [1]. With a controlled production process, artificial stones can successfully overcome many shortcomings in the design and installation of natural stones [2].

Structural reinforcement of buildings can extend their service life, reduce energy consumption, and reduce adverse effects on the environment. With the introduction of new products and/or the adoption of new building technologies, the durability and service life of a building can be predicted and hence its performance across the entire life cycle can be evaluated [3].

Natural stone materials tend to exhibit qualitative changes under changing environmental conditions [4–6]. Natural stone materials can be combined with other materials to meet the requirements of certain applications while maintaining the beauty that is usually attributed to natural stone materials [7]. Materials and products produced in such a combination are often referred to as artificial stones [4].

Artificial stones are advantageous because they exhibit consistent performance [1,4,7], but there have been very few studies of this in the literature [4]. Artificial stone has extremely low porosity, exhibits remarkable performance in construction applications, possesses consistent color and strength, and even has a maintenance-free surface (artificial stone does not require coating materials). As pointed out by a past study, tests of artificial stones conducted over 28 days will be required for long-term compatibility evaluation so as to observe the actual effects [2]. Therefore, it is worth conducting a study on the possibility of degradation of artificial stone under long-term use that changes the aesthetic values of its color and luster.

2. Materials

2.1. Composition of Artificial Stone Materials

Taiwan has a subtropical climate with warm, humid air currents and ocean currents. Therefore, the oceanic climate is a major factor in the weather resistance of a buildings' external surface. Taiwan has abundant rainfall, where the average annual rainfall can reach 2500 mm [8,9]. Since Taiwan has a humid climate, it is necessary to pay more attention to the problems of durability and permeability when choosing building materials. There are various types of artificial stone on the market; they have different compositions, qualities, and colors, and their properties depend on the characteristics of the stone materials [4]. Therefore, this study attempted to understand the requirements of weather resistance and then identify materials that meet these requirements for empirical analysis.

Zircon is a natural mineral with stable chemical properties. It can withstand temperatures of up to 3000 °C and possesses good durability [10–13]. Zirconium oxide was first introduced into biological materials because of its hardness, high density, and good abrasion resistance [14]. Zirconium oxide is an inorganic material that is chemically stable, non-toxic, and insoluble in water [15]. Portland cement with replacement by a fixed ratio of zirconium oxide possesses good radiopacity, compressive strength, setting time, water uptake, solubility, and sorption [14]. Another advantage of zirconium oxide is that it reduces the formation of plaques [16]. Recently, researchers have discovered that the micro-suspension and nano-suspension of modified silicon dioxide (SiO_2) or titanium dioxide (TiO_2) particles can improve the firmness and stain resistance of stone and grant the characteristics of self-cleaning and decontamination to the stone surface [17–20]. Due to its pozzolanic properties, the addition of nano-silica increases the compressive strength and reduces the overall permeability of hardened concrete. These effects can enhance the durability of concrete components and structures [21]. With the increase in nano-SiO_2 content, the compressive strengths of different mortars will increase accordingly [22]. The main cause of the increased strength and durability of concrete is the increase in crystalline $Ca(OH)_2$ content, especially the hydration in the early stage, which accelerates the formation of C-S-H (Calcium Silicate Hydrate) gel and reduces harmful pores as nano-fillers [23]. Thanks to these enhancements, nano-silica allows concrete to have greater density with the same fluidity. The anti-permeability grade of concrete is also greatly improved [23]. Using nano-SiO_2, TiO_2, Al_2O_3, and Fe_2O_3 as nano-fillers in the cement matrix, it has been found that the capillary pores are smaller and the total porosity is reduced [1–5,10–13,17,24]. The latest contribution to the development of building materials includes the addition of synthetic colloidal silica to concrete and cement mortar, so that the resulting product possesses improved aging performance in terms of increased strength, resistance to sulfate attack, and alkali–silica reaction [22].

The porous building materials used in historical buildings (e.g., natural stone materials, bricks, and ash mortar) are subjected to environmental and man-made destructive processes, leading to changes in the microstructural characteristics as well as physical and mechanical properties [25]. When $Ca(OH)_2$ reacts with carbon dioxide (CO_2) in the atmosphere, calcium carbonate ($CaCO_3$) will be generated. The newly formed $CaCO_3$ is chemically compatible with the calcareous matrix, so it can be combined with stone materials to strengthen them [26]. Heating the stone material to a high temperature is an artificial weathering method intended to induce a controllable microstructure [27]. Results have shown that the effectiveness of heating varies significantly with the carbonate content and porosity of heated stone; highly calcareous stone with very low porosity is the most sensitive lithotype [28]. The initial porosity of stone materials plays a very important role. Heating is a very promising method of artificial deterioration. The qualitative change depends on the initial porosity of the stone materials [25].

In view of the fact that Taiwan's natural stone materials are not weather-resistant, and summarizing the findings above, the elements of zirconium, silicon, and calcium play important roles in the weather resistance of stone materials.

2.2. Types and Sources of Stones Used in This Study

The artificial stones used in this empirical study are from the world's largest producer of quartz and slates in southeastern Spain (Figure 1). The company was established in Spain in 1980. In 2013, a materials science program was added and artificial stones that can be applied to exterior walls were introduced [29,30]. Therefore, Spanish artificial stones were selected for this study on weather resistance.

Figure 1. Southeastern Spain [31].

Composition analysis was conducted on a Spanish white artificial stone sample of 20 cm × 20 cm (Figure 2). From the test results, the elements of the Spanish artificial stone are calcium (Ca), magnesium (Mg), aluminum (Al), boron (B), barium (Ba), iron (Fe), potassium (K), sodium (Na), phosphorus (P), silicon (Si), strontium (Sr), zinc (Zn), and zirconium (Zr) (Table 1, Figure 3). The composition of zircon is the highest, accounting for approximately 10%.

Among the main compositions initially released by the Spanish manufacturer, the composition of the stone materials includes aluminum silicate, amorphous silica, crystalline silica, zircon, and inorganic pigments. In addition to the heat resistance due to the presence of zircon, these also increase the density and abrasion resistance of stone materials. Silica also provides stain resistance and compressive strength. As a compatible method, stone restoration based on inorganic binders in artificial stone has received much attention [2].

Figure 2. The Spanish artificial stone sample.

Table 1. Composition analysis of the Spanish artificial stone (Source: SGS Testing Center) [11–13].

Element	**Calcium (Ca)**	**Magnesium (Mg)**	**Aluminum (Al)**	**Boron (B)**	**Barium (Ba)**	**Iron (Fe)**
Accounted proportion in Dekton	3.21000%	2.34000%	0.00090%	0.04400%	0.09450%	0.00340%
Potassium (K)	**Sodium (Na)**	**Phosphorus (P)**	**Silicon (Si)**	**Strontium (Sr)**	**Zinc (Zn)**	**Zirconium (Zr)**
0.00032%	0.00990%	0.01550%	0.00234%	0.00050%	0.00040%	10.07000%

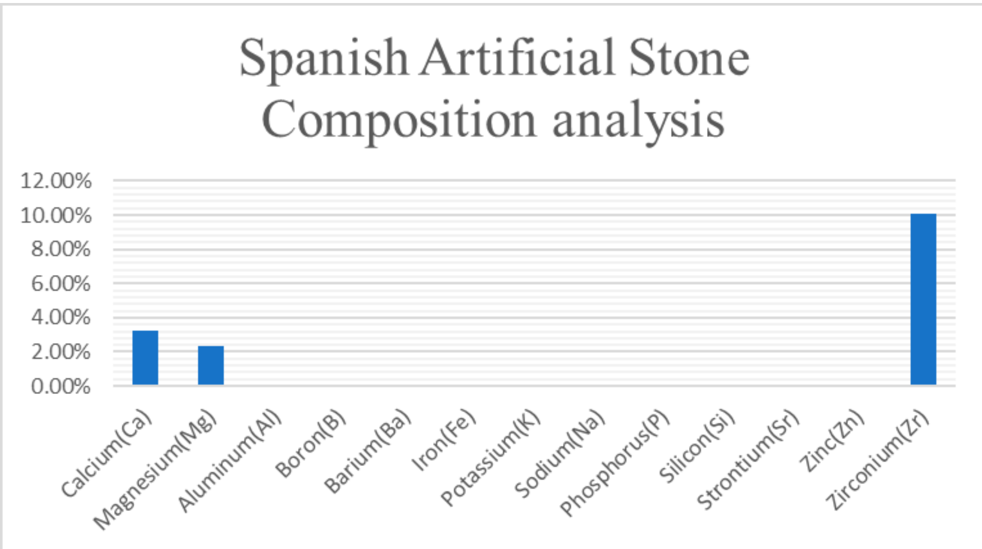


Figure 3. Bar chart of results of composition analysis of Spanish artificial stone [9–11].

3. Experimental Design

3.1. Weather Resistance Test in Laboratory

In this study, the Spanish artificial stones were subjected to experiments on their weather resistance, porosity, stain resistance, boiling water resistance, and compressive strength.

In the weather resistance study, the Spanish artificial stone (Figure 3) was tested to examine the weather resistance of the external surface using the ASTM G154-16 (Cycle) method. This is a testing method that examines ageing performance caused by exposure to sunlight, continuous humidity, and heating. With basic principles for operating fluorescent UV lamps and water equipment, techniques for specific exposure conditions are used in combination with methods evaluating changes in material properties. This method aims to reproduce weathering effects generated by exposure to the sun and moisture such as rain or dew in actual use. A UVA340 lamp was used to irradiate the sample with energy 0.89 W/m^2 at 340 nm; the operating temperature within eight hours was 60 ± 3 °C, and the operating temperature within four hours was 50 ± 4 °C [13]. The change in weather resistance of the external surface was observed after 3000 h. The experiment period was from 18 November 2016 to 14 April 2017. As attested by the experimental results, the external surface did not exhibit expansion or cracking (Table 2).

Table 2. Test results [13].

Test Item	Test Method/Test Content	Test Results
Weather resistance of outward appearance	ASTM G154-16(Cycle) [32]	No expansion or cracking phenomenon on outward appearance by visual inspection
Conditions of weather resistance		
Lamp tube	UVA340	
Illumination energy	0.89 W/m^2 at 340 nm	
Conditions of each cycle		
8-h light-illumination	Blackboard temperature: 60 ± 3 °C	
4-h condensation	Blackboard temperature: 50 ± 4 °C	
Testing time	3000 h	

(Data source: SGS Testing Center).

In a study on weathering of stone materials [33], both visual observation and SEM observation were used to evaluate the changes in color, weight, porosity, salt content, and dissolved calcium. This present study adopted the same research methodology (Table 3). The CNS 11367 method (1985) was adopted to observe some physical changes produced in Spanish artificial stones. First, the resistance to boiling water of stone materials was observed. The experimental results showed that there is no expansion or foaming on the external surface. The thickness expansion rate increased by only 0.05%, the impact was minimal, and the mass change rate was unchanged. Second, good results were obtained for impact resistance; the stone material had no ruptures, dents, or other defects. Moreover, the Spanish artificial stone was subjected to tests of stain resistance for tea, coffee, milk, vinegar, citric acid, ethanol, and acetone. Experimental results show that all had no impact on the Spanish artificial stone, demonstrating its good stain resistance. Using direct burning of cigarettes to test the smoke resistance of the surface showed that there were no burn marks that could not be cleaned and no color fading, cracks, or other defects on the stone material.

Using the ASTM C170/C170M-09 method for compressive strength, the compressive strength was found to be 4563 kgf/cm^2, which is higher than the standard compressive strength of granite, 1336 kgf/cm^2, and that of marble, 530 kgf/cm^2, showing that this Spanish artificial stone has good compressive strength. Using the ASTM D792-08 Method A, the density of this stone material is 2.49 g/cm^3. The ASTM D570-98 (2010) e1 is used for water absorption. The results show the water absorption rate of this stone material is only 0.03%. These results are consistent with the results of the above-mentioned anti-pollution experiments. Therefore, these stones will resist chemical changes triggered by rain when used as exterior walls.

In terms of chemical resistance (23 °C, 30 days), the reactions generated on an exterior wall by climate change or other external factors were also examined. The CNS 4447 (1992) method was used with 5% KOH solution, 5% HCl, and 5% H_2SO_4, and the results demonstrated that there were no abnormal effects on the external surface of the stone material in terms of chemical resistance.

Table 3. Details of the test.

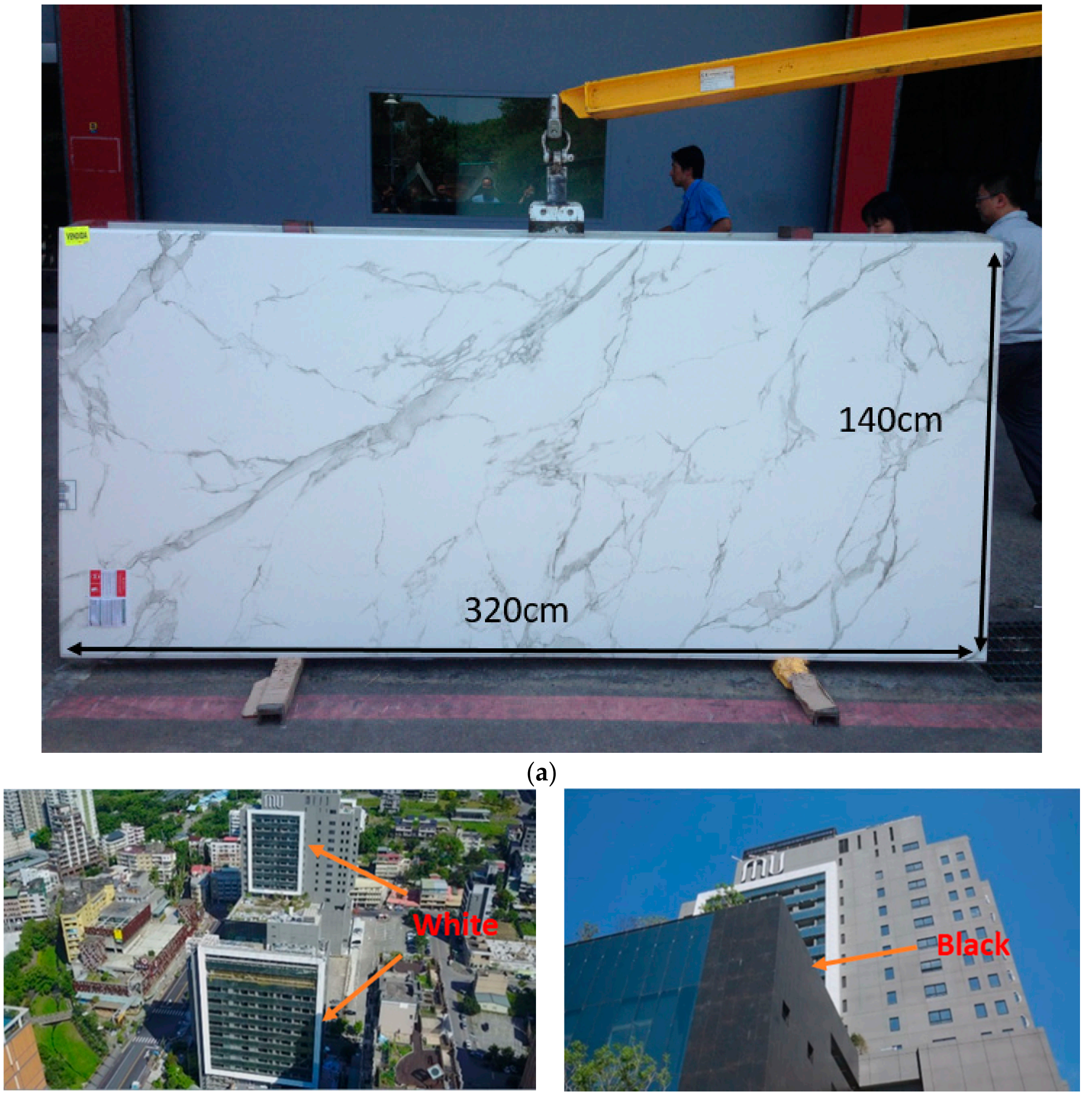
Test Item	Test Method	Test Results
Boiling water resistance	Referring to CNS 11367 (1985) [34]	No expansion and foaming phenomenon on outward appearance
a. Thickness expansion rate (%)		0.05
b. Mass change rate (%)		0
Impact resistance (28.4 g, 900 mm)		No rupture or dent and other defects
Stain resistance		
a. Tea		No impact by visual inspection
b. Coffee		No impact by visual inspection
c. Milk		No impact by visual inspection
d. Vinegar		No impact by visual inspection
e. 10% Citrus acid		No impact by visual inspection
f. 95% Ethanol		No impact by visual inspection
g. Acetone		No impact by visual inspection
Smoke resistance (direct burning of cigarettes)		No burn marks that cannot be cleaned, color fading, cracks, or other defects on the surface
Compressive strength (kgf/cm^2)	Referring to ASTM C170/C170M-09 [35]	4563
Density (23 °C) (g/cm^3)	Referring to ASTM D792-08 Method A [36]	2.49
Water absorption (%)	Referring to ASTM D570-98 (2010) el [37]	0.03
Chemical resistance (23 °C, 30 days)	Referring to CNS 4447 (1992) [38]	
a.5%KOH solution		No abnormal state
a.5%HCL		No abnormal state
a.5%H_2SO_4		No abnormal state

Note: Standard compression strength of granite: 1336 kgf/cm^2; standard compression strength of marble: 530 kgf/cm^2.

3.2. Case One—Mu Jiao Xi Hotel in Yilan; Experiment Period: 2 Years

The Mu Jiao Xi Hotel in Yilan was built in May 2015. The buildings are located on both the north and south sides of Jiankang Road. The building areas are 2594 m^2 and 5120 m^2, respectively. Respectively, there are eleven and fourteen floors above ground and the building heights are 49.45 m and 63 m. The two buildings are located over two street blocks. In February 2017, Spanish artificial stones of both the white (Figure 4a,b) and black (Figure 4c) types were used as exterior wall materials for the buildings. The Spanish artificial stones had a standard size of 320 cm × 140 cm and were cut into pieces with typical sizes of 145 cm × 80 cm and 120 cm × 140 cm. After the completion of the exterior walls, the changes in the weather resistance of the external surface were tracked until September 2020.

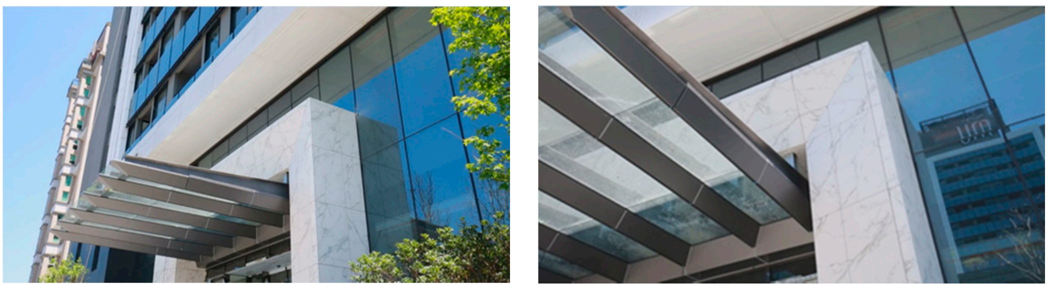
The exterior wall project for Mu Jiao Xi Hotel was completed in February 2018. As of September 2020, no color change has yet been observed (Figure 5a,b and Figure 6). In addition to the confirmation of weather resistance obtained by the laboratory indoor weather resistance test, this shows that the artificial stone of an exterior wall shows no changes after two years of outdoor exposure. The paper proposed a new protective coating for use on building stone materials that can prevent the stone from changing color and improve the hydrophobicity [39]. Therefore, in this study, an oily stone material protective agent was used on the black artificial stone. Over time, interactions between the coating and the surrounding environment generated a matte surface on the stone materials, and dirt and stains due to weathering were observed (Figure 7). Therefore, it is not advisable to add a stone material protective agent to Spanish artificial stones that already possess natural compositions and weather resistance. Under typical conditions, Spanish artificial stones display sufficient weather resistance.



(**a**)

(**b**) (**c**)

Figure 4. Mu Jiao Xi Hotel in Yilan. Both buildings use Spanish artificial stones as decorative materials: (**a**) white, (**b**) white, and (**c**) black.



(**a**) (**b**)

Figure 5. Photos of the completed building façade with white artificial stone showing no observed color changes: (**a**,**b**).

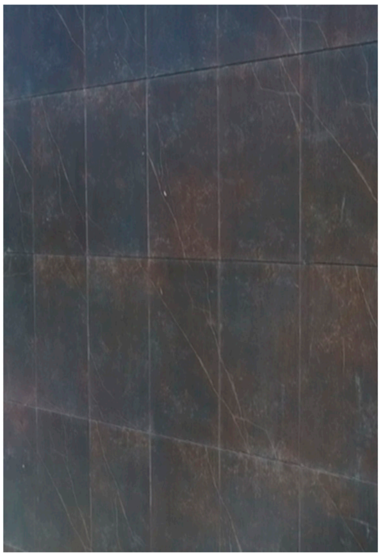
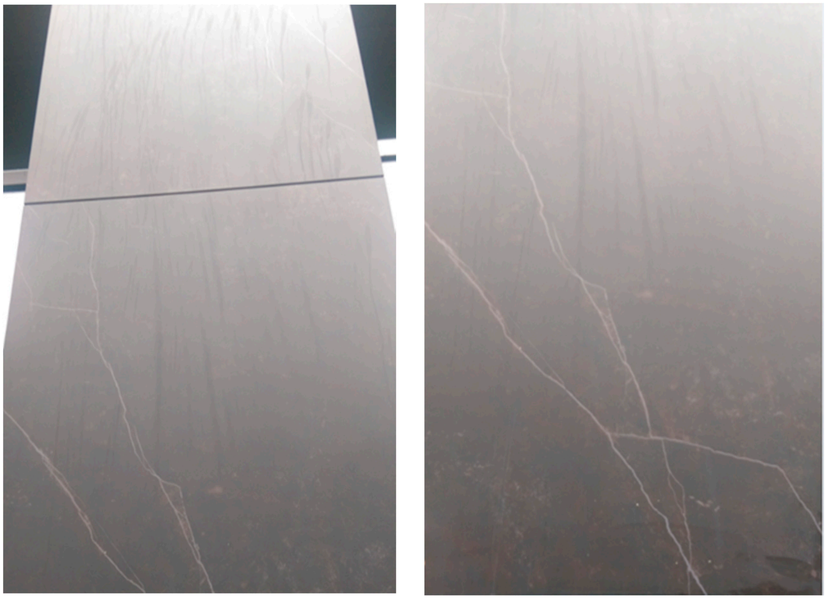


Figure 6. The completed building façade with black artificial stone, with no color change observed.



(**a**) (**b**)

Figure 7. Appearance of dirt on black artificial stones with oily stone material protective agent: (**a**,**b**).

3.3. Case Two—Culture, Leisure, and Social Education Center in Taipei

The second case is the Culture, Leisure and Social Education Center for Taipei City, located next to the Miramar Shopping Center in Taipei. It has eight floors above ground and four levels of basements, with a building area of 18,996 m^2. It can accommodate 2300 people and includes bookstores, offices, wedding halls, multi-functional assembly halls, and dormitories. White Spanish artificial stone pieces of size 320 cm $\times$ 140 cm were cut into various sizes for use, as shown in Figure 8.

The installation of stone material in the exterior wall was completed on 14 February 2020, as shown in Figure 8. On 15 September 2020, it underwent various inspections, including measuring widths of seams between segments and the flatness of the cuts, and finally a waterproofing test on the exterior wall. The building at the time is shown in Figure 9.

(**a**)

(**b**) (**c**)

Figure 8. Photos of construction on 14 February 2020: (**a**) moving, (**b**) installing, and (**c**) installed.

(**a**) (**b**)

Figure 9. The building of the Culture, Leisure, and Social Education Center, with no observed impact on the color of the external surface of the Spanish white artificial stones: (**a**) view 1 and (**b**) view 2 (source: photographed for this study on 15 September 2020).

4. Experimental Results

In this study, outdoor empirical analysis was conducted for the two cases. Case One is located in Yilan with a duration of two years from February 2018 to September 2020. Case Two is located in Taipei with a duration from February 2020 to September 2020. After half a year and the dismantling of the frame, all the Spanish artificial stones show no change in outward appearance and were still as pure white as new. As indicated by a past study, in Taiwan's humid and rainy climate, natural marble will undergo yellowing within half a year and this was found to be a result of heavy rain and acid rain [6]. Therefore, this study also investigated the effects of rainfall and pH value.

4.1. Rainfall and pH Value during the Experiment Period

In this study, the rainfall and pH values for Yilan from January 2015 to September 2020 were collected. The graph in Figure 10 shows the average annual rainfall in Yilan can reach 229 mm; compared with other regions of Taiwan, the rainfall is relatively high. The average annual pH values from 2015 to 2017 were 5.42, 5.57, and 5.21, respectively; the average pH value for these years is 5.4, which is classed as acid rain. For the rainfall and pH values of Taipei from January 2015 to September 2020, the graph in Figure 11 shows the average rainfall has had a declining trend in recent years, and the average rainfall during this period was 182.29 mm. The average annual pH values from 2015 to 2017 were 5.23, 5.51, and 5.32, respectively; the average pH value for these years is 5.35, which is classed as acid rain.

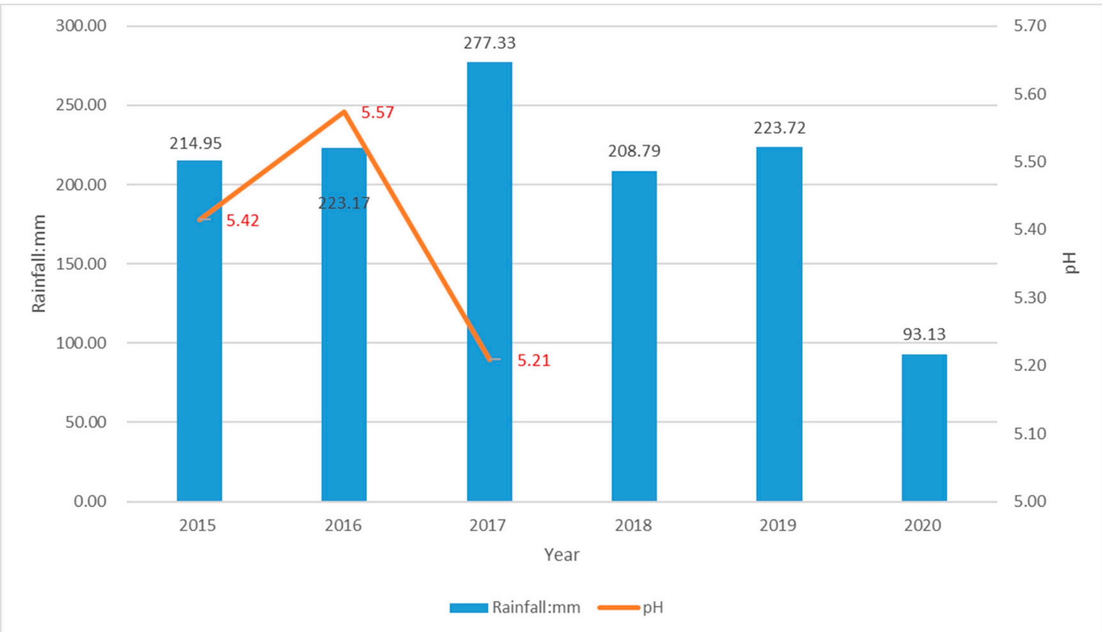


Figure 10. Dual-axis graph for rainfall and pH value in Yilan during the study period (1.2015–9.2020). Note: The Environmental Protection Administration only provided the pH value up to 2017.

Water is the main factor in natural weathering. Rain penetrates stone materials through their porosity and the salt content of the rainwater and pollutants in the air cause chemical reactions. With the added effects of changes in temperature and other factors, the internal structure of stone materials may be damaged [39–41]. The amount of rainfall is very important for the deposition of dry pollutants on the building façade, especially in the dry season of the Mediterranean climate, and the porosity and shape of stone materials, as well as the exposure conditions are also important factors affecting the weathering of stone materials [41]. For Yilan, the average annual rainfall for the past five years is 229 mm and its pH value is approximately 5.4, and for the Taipei basin, the average annual rainfall for the past five years is 188 mm and its pH value is approximately 5.3, which are close to the level of acid rain. Under such conditions, the exposed artificial stones in the exterior wall will not show degradation after weathering, but instead maintain their original state (Figure 9).

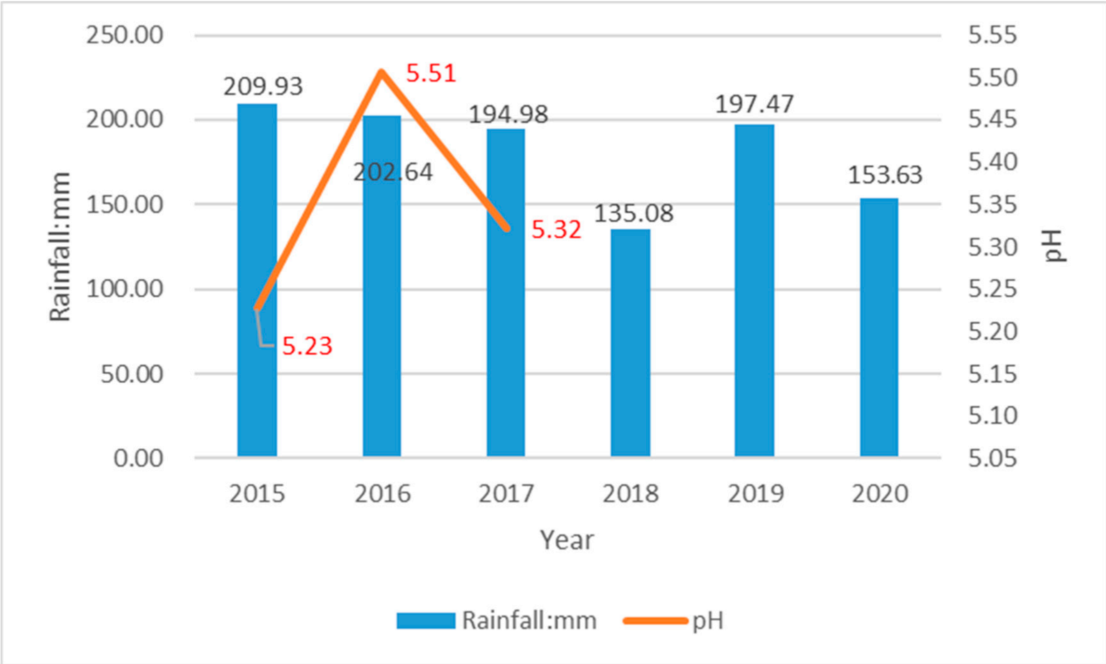


Figure 11. Dual-axis graph for rainfall and pH value in Taipei during the study period (1.2015–9.2020). Note: The Environmental Protection Administration only provided the pH value up to 2017 [42].

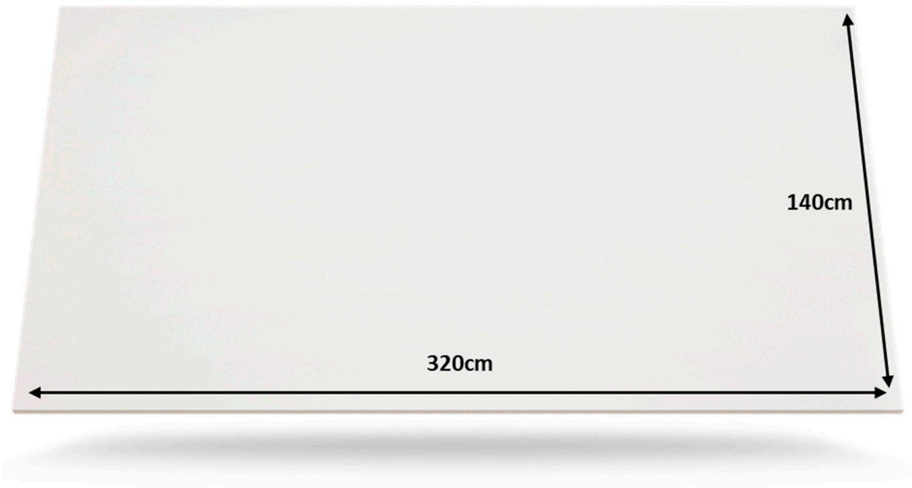
4.2. *Experimental Results of White and Black Spanish Artificial Stones from the Original Manufacturer*

A series of tests on the original Spanish artificial stones were conducted by the manufacturer in a weathering chamber using a xenon arc lamp to test the stability of the ultra-compact surfaces against UV light. Both the white (Figure 12a) and black (Figure 12b) artificial stones were tested as they represent the full range of colors and they were expected to be the most sensitive to the experimental conditions.

The tests were conducted using the Q-Sun tester system (Q-SUN-XE-3-HS) equipped with daylight filters with an irradiance of 0.51 W/m^2 at 340 nm. Typical 102/18 cycles based on the ISO 113 41:2004 standard were adopted and the following test parameters were used: black panel temperature 63 °C, air chamber temperature 43 °C, relative humidity 30%, 1:42 h light/0:18 h light and spray [10].

The samples were measured and compared after more than 5000 h of exposure. The change in color was determined using the unequivocal parameter $\triangle E^*$ of the CIELab chromatic space, representing the difference between two colors. When $\triangle E^* < 1$, both colors are considered equal. When $\triangle E^* > 1$, the color difference can be noticed by the naked eye. The results are provided in Table 4.

Observations of the white and black Spanish artificial stones after being exposed on the exterior walls in the outdoor empirical studies, as well as the reported results from domestic and foreign laboratory studies, have yielded consistent results for weather resistance.



(**a**) White.

(**b**) Black.

Figure 12. Spanish artificial stones (**a**) white, (**b**) black used in this study.

Table 4. Results of original experiment.

Color	Exposition Time, h	△E *
Black	>5.000	<1
White	>5.000	<1

Note 1: Data source: Study in Cantoria (Almeria), Spain, on 2 May 2014. Note 2: These display values indicate that the products are not affected by continuous UV irradiation. Therefore, the product is suitable for its intended use as external applications.

5. Conclusions and Future Developments

Taiwan has a subtropical island climate with high humidity and dense water vapor in the air. Therefore, it is particularly important for the stone materials used in the façade to pass weather resistance tests in Taiwan. Such tests should not be limited to the laboratory [12,33], but should be developed for long-term observation under outdoor conditions.

This study examined two cases. The first is in Yilan County, Taiwan, where the changes in black and white Spanish artificial stones after weathering of exterior walls of buildings were observed from February 2018 to September 2020. The average annual rainfall for the past five years in Yilan is 229 mm and its pH value is approximately 5.4. The results showed the Spanish artificial stones under these environmental conditions did not exhibit weathering after more than two years.

The second case is in Taipei. A building covered entirely with white Spanish artificial stones was observed from February 2020 to September 2020. The average annual rainfall for the past five years in Taipei is 182 mm and its pH value is approximately 5.3. After a period of natural weathering for 6 months, the building's external surface looks as pure white as new.

Comparing these two cases with the qualitative changes in natural marble generated after weathering over half a year in Taipei [6], different trends can be clearly observed for natural stone and artificial stone in the weather resistance studies. A past study indicated that the porosity of stone material will affect the qualitative change in the stone material [41]. As shown by the experimental results of this study both inside and outside the laboratory, the water absorption rate of Spanish artificial stone is only 0.3%. The strong stain resistance of the artificial stones can be observed through the actual weathering of the exterior walls. Attributing the results to the composition of the artificial stone, zirconium oxide can reduce the formation of plaque [16] and nano-silica was found to be a very effective concrete additive that can improve strength, ductility, and durability. It can also be used as an additive to improve the workability as well as the strength of high-performance and self-consolidating concretes [3]. Adding synthetic colloidal silica to concrete and cement mortar and thereby enhancing the strength produces improved aging performance in terms of resistance to sulfate attack and alkali–silica reaction [1]. Therefore, the Spanish artificial stone has good applicability for a building's external surface in subtropical island climates.

Author Contributions: Y.-C.C. Chiu collected data and wrote the manuscript; P.-H.C. Chen provided guidance on writing and structure of the manuscript; W.-C.L. Liao provided advice on the tested materials. All authors have read and agreed to the published version of the manuscript.

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