



Article Recycling Cigarette Butts in Ceramic Tiles

Qibin Yuan *[®], Abbas Mohajerani [®], Adrian Kristoforus [®], Halenur Kurmus [®], Urmila Chowdhury, Dilan Robert, Biplob Pramanik [®] and Phuong Tran [®]

School of Engineering, RMIT University, 376-392 Swanston Street, Melbourne, VIC 3001, Australia; abbas.mohajerani@rmit.edu.au (A.M.); adrian.kristof@yahoo.com (A.K.); s3432918@student.rmit.edu.au (H.K.); urmilachowdhury2000@gmail.com (U.C.); dilan.robert@rmit.edu.au (D.R.); biplob.pramanik@rmit.edu.au (B.P.); jonathan.tran@rmit.edu.au (P.T.)

* Correspondence: s3595781@student.rmit.edu.au

Abstract: Cigarettes are one of the favoured commodities on our planet. However, the annual consumption of 5.7 trillion cigarettes and 75% littering rate results in cigarette butts (CBs) being one of the most critical environmental issues. The leachate of heavy metals and toxic chemicals is polluting our ecosystem and threatening the wildlife species. Therefore, it is crucial to find effective and efficient recycling methods to solve the growing CB waste issue. In this study, unglazed fired ceramic tiles were manufactured with 0%, 0.5%, 1.0%, and 1.5% shredded CBs by dry mass to investigate the feasibility of the proposed sustainable recycling method. The chemical and mineralogical characterisation, density, shrinkage, bulk density, breaking strength, water absorption, and modulus of rupture were investigated and compared with the Australian Standards for ceramic tiles (AS 4459). The results revealed that tiles incorporating 0.5% CBs by mass demonstrated the greatest performance compared to the other mixtures. The water absorption for all tile-CB mixtures was found to be greater than 10%, with a positive growth tendency. The addition of 0.5% CBs by mass slightly improved flexural strength from 15.56 MPa for control samples to 16.63 MPa. Tiles containing 0.5% CBs by mass satisfied the modulus of rupture and water absorption limits for group III class according to the Australian Standards (AS 13006), and they may be suitable to be used as wall tiles. The result of a simulation equation predicts that an energy savings of up to 7.79% is achievable during the firing process for ceramic tiles incorporating 1% CBs by mass.

Keywords: cigarette butts; building materials; ceramic tiles; energy saving; recycling

1. Introduction (Literature Review)

Cigarettes have been one of the most common consumables in the world for a long time, from the past to the present. In 2016, 5.7 trillion cigarettes were consumed globally in a single year [1]. Studies have shown that cigarette consumption and daily smoking rates for both men and women have fallen in most countries over the last three decades; however, the number of smokers is still immense [2,3]. The absolute number of smokers increased from 720 million in 1980 to nearly 1 billion in 2012 [4]. Cigarette consumption continues to trend upwards in some developing countries, one being China, with more than 2.5 million metric tons of cigarettes consumed in 2013, surpassing the sum of the next 40 highest consuming countries [2]. Therefore, the number of cigarette butts (CBs) and associated waste generated cannot be overlooked.

Moreover, cigarettes are as harmful to the environment as they are to the human body. Researchers estimate that 75% of CBs are discarded carelessly into nature [5,6], and 95% of the cigarette filters are made of modified cellulose acetate polymer, which has poor biodegradability and can take more than 12 months to break down under inadequate environmental conditions [1,7]. The smoked CBs contain over 3500 chemicals, many of which are toxic and known to be carcinogenic, such as nicotine, polycyclic aromatic hydrocarbons, nitrosamines, benzene, cyanide, and cadmium [8–10]. The toxic chemicals



Citation: Yuan, Q.; Mohajerani, A.; Kristoforus, A.; Kurmus, H.; Chowdhury, U.; Robert, D.; Pramanik, B.; Tran, P. Recycling Cigarette Butts in Ceramic Tiles. *Buildings* **2022**, *12*, 17. https:// doi.org/10.3390/buildings12010017

Academic Editors: Ahmed B. Senouci and Walid Maherzi

Received: 15 November 2021 Accepted: 23 December 2021 Published: 28 December 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). and heavy metals will leach out, pollute the environment, and endanger wildlife. In the research paper "Toxicity of Cigarette Butts, and Their Chemical Components, to Marine and Freshwater Fish" [11], the experimental results demonstrated that both smoked and unused CBs are highly poisonous to the tested species of marine and freshwater fish. In addition, CBs can potentially release microfibres into our water bodies, reaching roughly 300,000 tons a year [6].

Due to the toxicity of CBs and their associated diverse contaminants, recycling may be a more desirable option for CB management than the conventional landfill and incineration methods [12]. With the aim to lighten the burden of waste management and to create some incentives to promote a more sustainable society, many researchers are investigating options for recycling and reusing waste materials. The potential applications that have already been investigated include recycling into the brick manufacturing process [13–16], recycling into asphalt for road pavement [17–19], conversion to cellulose pulp for paper production [20], use in sound-absorbing materials [21], and many more. Although many of the mentioned studies have encountered hurdles or consequent new environmental challenges during the investigation phases, they exhibit potential and therefore require further comprehensive research.

For the purpose of this study, ceramic tiles were selected to recycle and encapsulate CBs. Ceramic tiles are heterogeneous products and can be produced with various raw materials or even waste materials. One example would be the use of grog (crushed pre-fired clay products) in the raw material formulation. Moreover, the mineral constitutions of clay vary from region to region, but it is still feasible to make a competent tile by adjusting to a suitable recipe with an optimum production process [22]. In addition, porcelain tiles are the most recognised technological innovation on the basis of traditional porous ceramic tiles; they offer a superior end product with more refined raw materials and an optimal production process. For these reasons, the high tolerance of ceramic composition is validated and widely used, and changes in formulation and processes may even improve the tile performance.

The high demand for tiles also offers a potential future for the proposed sustainable approach. Statistics show that world tile production in 2017 demonstrated continual growth that reached 13.552 billion m² (2.2% higher than in 2016); tile consumption also increased by 2.2% to 13.3 billion m² [23]. Hence, the tile industry has the ability to recycle a large amount of waste material by only replacing a small percentage of the raw material. However, the economic recession, trade wars, and pandemics in recent years have led to a decline in world tile production to 12.673 billion m² in 2019 (a drop of 3.7% compared to 2018), in addition to tile consumption falling by 4% to 12.4 billion m² [24]. It must be highlighted, though, that for such a rigidly demanded building material, production and demand are still strong.

Long-term environmental impacts are critical considerations when recycling waste materials. Due to the high tolerance feature of ceramic bodies, approximately 300 papers have been published on waste recycling in ceramic tile making, with more than 56 types of waste studied for recycling in ceramic tiles [25]. Most studies have been conducted on non-hazardous waste, as the legal restrictions of recycling make the process complex and challenging [25], but some research findings have shown that the firing process can potentially immobilise the heavy metals in waste materials and encapsulate the hazardous leachate in fired tiles during the high temperatures [26]. In addition, municipal waste incineration fly ash tile studies have found that the tile glazing can further reduce the leaching of heavy metals by enclosing the surface porosity [27], which is commonly applied to tiles to improve the tile performance both aesthetically and practically.

Moreover, in 2020, Kurmus and Mohajerani [28] published a relevant journal article on the leachate analysis of heavy metals in fired clay bricks incorporating CBs. The leachate study was conducted according to the Australian Bottle Leaching Procedure (ABLP) for pH values 2.9, 5.0, and 9.2. The results revealed that littered CBs are highly prone to leaching heavy metals (Cu, Zn, Mn, Al, Fe, Ti, and Ba) in areas with highly acidic rain compared to the natural range of precipitation. Moreover, it was found that the firing process assisted in immobilising more than half the tested metals for the fired clay brick samples incorporating CBs [28]. The leachate results of the fired CB brick samples were found to be below the regulatory limits set by the local industrial waste guidelines, and therefore they can be classified as non-hazardous at the end of their service life [28]. Due to the similar production processes and high temperatures during the firing process of both fired clay bricks and ceramic tiles, it can essentially be assumed that similar leachate results will be achieved; therefore, tiles incorporating CBs can be considered non-hazardous waste. However, it is recommended that a comprehensive leachate analysis for heavy metals is conducted for the proposed recycling of CBs in tiles.

This paper is a preliminary research study on the incorporation of CBs into the manufacturing of ceramic tiles. In this research, tiles containing 0.5%, 1.0%, and 1.5% CBs by dry mass (roughly corresponding to 0 kg/m³, 10 kg/m³, 20 kg/m³, and 30 kg/m³ CBs, respectively) are manufactured, investigated, and compared to control samples. These percentages were selected based on the experiences and the large-volume feature of CB fibre. Numerous tests are conducted, including shrinkage, bulk density, water absorption, breaking strength, modulus of rupture, X-ray fluorescence (XRF) analysis, and X-ray diffraction (XRD) analysis. Moreover, the energy savings percentage during the firing process of tiles is presented as a function of CBs present in the mixtures.

2. Materials and Methods

2.1. Raw Materials

The cigarette is a product made up of several various elements (as shown in Figure 1). Although smoked cigarettes also have all the components as new cigarettes, the major constituents are the filter (mainly cellulose acetate polymer) and tipping paper. The filter retains the burning residue and chemicals of tobacco, cigarette paper, and additives, which makes it hazardous to our environment. In this project, the fresh CBs were supplied by Butt Out Australia Pty Ltd. (Melbourne, Australia); all the CBs are collected from their cigarette receptacles located in Australia's capital cities and major regional centres. Hence, the CB waste is composed of different brands with varying sizes. It is important to process the fresh CBs before adding them to the ceramic bodies. CBs were left in the oven at 105 °C for 24 h to remove the moisture content and disinfect the waste. Then, the oven-dried CBs were put into a blender, shredded into a flocculent form, and then stored in air-tight plastic bags to prevent moisture from the air. Due to the low density and fluffiness of CBs, the amount utilised in this study is trivial.

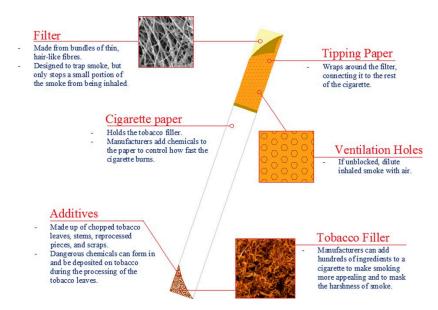


Figure 1. The compositional structure of a cigarette (from Kurmus and Mohajerani [1]).

The selection of ceramic raw materials was based on the three key elements of ceramics, which are plasticity provider, flux, and filler; sometimes, additives will be used to improve the overall performance [22]. The plasticity provider is the clay materials that accommodate other ingredients; it gives strength and plasticity to the unfired tiles. The flux is the feldspathic materials that promote densification during the liquid phase under high temperatures. The filler is a quartzous material that is similar to the bones in our body; it controls the shrinkage and pyroplastic deformation during firing. In this research, the formula was adapted and modified from relevant studies. The ceramic ingredients include kaolinite (Eckalite 2), feldspar (Potash 200 Mesh), quartz (Silica 200 Mesh), and bentonite. In addition, bentonite is an additive obtained from volcanic ash, and it is intended to improve the plasticity of eckalite clay with better dry strength [29]. All the ceramic raw materials were processed into a fine powdery form and purchased from Northcote Pottery Supplies Pty Ltd. (Melbourne, Australia).

2.2. Chemical and Mineralogical Characterisation

The major chemical constituents of experimental raw materials were determined through an XRF (S4 Pioneer, Bruker, Billerica, MA, USA). The crystalline mineral phases of the fired tiles were analysed through an XRD (D8 Endeavor, Bruker, Billerica, MA, USA). The fired tiles were crushed into powders below 75- μ m particle size. The analysis was done in the 2 θ = 6–90° range to detect the crystalline phases.

The chemical composition of the raw materials and CB samples was analysed by the XRF instrument, and the results are shown in Table 1. In Table 1, the results indicate that the clay body is mainly composed of SiO₂, Al₂O₃, K₂O, and Na₂O. As the CB does not exist in a powder form, the analysed results may not be accurate, and the sum of the analysed percentage is only 31.52%. The results of the CB sample show that it contains a wide range of chemicals, including CaO, K₂O, TiO₂, Cl, and SiO₂.

	Chemicals (%)										
	Na ₂ O	MgO	Al_2O_3	SiO ₂	P_2O_5	SO ₃	Cl	K ₂ O	CaO	TiO ₂	Fe ₂ O ₃
Kaolin	-	-	30.369	35.603	0.205	0.072	-	0.227	0.13	0.696	0.76
Feldspar	2.196	-	15.145	51.172	0.211	-	-	11.021	0.283	-	0.135
Quartz	-	-	0.459	82.311	0.237	-	-	0.01	0.137	0.018	0.031
Bentonite	1.299	1.325	13.055	39.948	0.224	0.344	-	0.465	1.394	0.221	3.846
Mixture	0.948	0.065	22.779	47.466	0.208	0.043		4.039	0.206	0.345	0.488
СВ	-	0.919	0.534	1.329	0.697	0.937	2.188	7.938	12.713	3.236	0.842

Table 1. The chemical composition (oxides) of the raw materials.

Note: mixture only stands for mixed control tile formula (no CBs added).

2.3. Preparation of Ceramic Tile Samples

The formulation of ceramic tiles is displayed in Table 2, and all ingredients are shown in percentage by dry mass. The amount of incorporated CBs varied from 0–1.5 wt. % with an increment of 0.5 wt. %. The preparation procedure of the CB–ceramic tiles includes (1) Mixing: CBs and ceramic raw materials are mixed in a 20-L Hobart mechanical mixer for a length of 15-min with 26 wt. % (of the total dry raw materials) of water that was added incrementally during the 15-min mixing process to make it into a slightly plastic and mouldable state. (2) Moulding: the mixtures were compacted in a cylindrical metal mould with a diameter of 100 mm by a Servopac Gyratory Compactor. All tiles were gyratory compacted 30 times with a pressure of 240 kPa, and the height of all samples was approximate 10 mm (Figure 2). (3) Drying: The specimens were air-dried inside the laboratory with a standard room ambient temperature for 5 days and then oven-dried at 105 °C for an hour before the firing process. The intention is to simulate the drying process of some manufacturers to reduce the possibility of deformation caused by oven-drying and lessen the energy used. (4) Firing: The firing process was done in the Thermoconcept electrical chamber furnace (Model KC220/13). Firing schedule was derived into 3 segments (shown in Table 3) from slow to fast heating rate, with a target firing temperature of 1200 °C. Once the target temperature was achieved, the furnace cooled down at a natural cooling rate and was ready for investigation of the properties of its contents.

Table 2. The p	ratios of raw	materials for	ceramic mixtures.
----------------	---------------	---------------	-------------------

	Control	0.5%	1.0%	1.5%
CBs	0.0	0.5	1.0	1.5
Kaolin	50.0	50.0	50.0	50.0
Feldspar	36.0	36.0	36.0	36.0
Silica	12.0	12.0	12.0	12.0
Bentonite	2.0	1.5	1.0	0.5

Note: materials were measured and presented by dry mass percentage (wt. %).



Figure 2. Pictures of ceramic tiles (left: freshly compacted; right: fired).

Segment	Rate (°C/min)	Time (h)	Temperature (°C)
0	0	0	20
1	1.5	2	200
2	2.5	2	500
3	3	3.89	1200

Table 3. The firing schedule.

2.4. Physical and Mechanical Tests

The shrinkage test was implemented by measuring the diameters of six replicate tiles for each batch, and then the diameters of different states of tiles were calculated and compared. The state of the tiles is in the order of moistened, air-dried, oven-dried, and fired tiles. The shrinkage rate (LS%) is calculated by the equation shown below:

$$LS = \frac{L_0 - L_1}{L_0} \times 100\%$$
 (1)

The water absorption was followed by the boiling method AS/NZS 4459.3-1999 (R2017). The fired CB–ceramic tiles were oven-dried at 105 °C to expel all the possible moisture, and then the constant dry mass (m_1) was measured before the water bath testing. Subsequently, the fired CB tiles were placed in the water bath apparatus located in the middle of 10-cm-deep water. The water was heated to 100 °C and held for 2 h before switching off. The wet mass ($m_{2(b,v)}$) of tiles was measured after the water cooled down

to room temperature. The water absorption $(E_{(b,v)})$ (Equation (2)) and bulk density (B) (Equation (3)) can be calculated as follows:

$$E_{(b,v)} = \frac{m_{2(b,v)} - m_1}{m_1} \times 100\%$$
⁽²⁾

$$B = \frac{m_1}{V}$$
(3)

The three-point bending test was performed in conjunction with AN/NZS 4459.4 by using a 50-kN Shimadzu universal testing machine. The breaking strength (S) (Equation (4)) and modulus of rupture (R) (Equation (5)) were determined by the following equations, respectively:

$$S = \frac{FL}{b}$$
(4)

$$R = \frac{3FL}{2bh^2}$$
(5)

where F represents the breaking load measured in newtons; L is the span between support rods measured in millimetres; b is the width (diameter) of the test specimen measured in millimetres; and h is the minimum thickness of the test specimen, also measured in millimetres.

2.5. Energy Saving

One of the main issues with the production of ceramic tiles is that it places a significant strain on the earth's natural resources, as substantial energy is consumed during the manufacturing process. It would be an incentive for tile manufacturers if adding CBs could save costs on energy consumption. Ceramic tiles are required to be fired at temperatures around or even above 1050 $^{\circ}$ C to achieve the vitrification stage to strengthen both the physical and chemical features of the ceramic. Furthermore, the decomposition of organic content in CBs will release more heat during the firing process, resulting in less energy being used to raise the temperature [30]. In the calculation method, the standard mean energy usage per kiln for ceramic tile firing was estimated to be 2.8 MJ/kg [31]. While the calorific value of the CBs was found to be 16.53 MJ/kg, this value is based on a laboratory study conducted by Mohajerani, Hui, Shen, Suntovski, Rodwell, Kurmus, Hana, and Rahman [16]. The process of partially replacing the ceramic tile mixture with CBs will allow the high-calorific-value material to act as an energy carrier during the firing phase and potentially lower the firing energy needed to manufacture the ceramic tiles. The energy savings were calculated by considering the mass of the control sample (m_1) , mass of CB-ceramic tile (m_2) , mass of CBs in the CB-ceramic tile (m_3) , calorific value of the CB (CV), and energy used for ceramic tile firing (q).

Energy saved,
$$\Delta E(\%) = \frac{(q \times m_2) - (q \times m_2 - CV \times m_3)}{q \times m_1} \times 100\%$$
 (6)

3. Results and Discussion

3.1. Characterisation of Fired Tiles

Figure 3 presents the XRD analysis results, exhibiting the crystalline phases of fired control tiles and tiles with CBs sintered at 1200 °C. The major crystalline phases presented in the ceramic body after firing are quartz (SiO₂) and syn–orthorhombic mullite (Al_{4.54}Si_{1.46}O_{9.73}). The introduction of CBs does not have a significant impact on the ceramic body in the mineralogical aspect, and the fired tiles with different percentages of CBs have similar results. The ceramic body did not develop new crystalline phases with the incorporation of CBs.

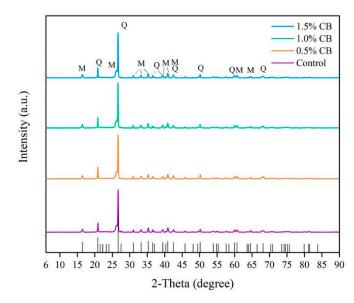


Figure 3. XRD analysis of tile samples sintered at 1200 °C (Q: quartz and M: mullite).

3.2. Physical and Mechanical Properties of Fired CB Tiles

3.2.1. Dimensions and Properties

The basic properties of fired CB tiles were measured with an electronic vernier caliper and electronic scale, and the average of five replicates for each percentage was calculated. As the tiles involve some hand processing steps, some non-uniformities occur on the height and edges. Several measurements were performed to reduce human error and increase the precision of the results. By comparing the properties of tiles with different percentages of CBs displayed in Table 4, there is an inverse proportional relationship between the amount of CBs and tile density, in which the density tends to become lower as the percentage of CBs increases.

	Control	0.5%	1.0%	1.5%
Mass (g)	115.32	114.82	114.54	114.22
Diameter (mm)	87.76	88.13	89.14	89.05
Height (mm)	9.10	9.35	9.13	9.19
Volume (mm ³)	55,017.71	57,013.60	57,000.65	57,214.93
Density (kg/m^3)	2096.16	2016.38	2009.17	1997.41

Table 4. The average basic properties of fired CB tile samples.

3.2.2. Shrinkage Test

The shrinkage test results presented are the average diameter of six replicate tiles per batch. The drying shrinkage is obtained from the comparison between the diameters of unfired dry tiles (by air-dry and oven-dry) to the 100-mm mould. The firing shrinkage is the shrinking ratio of the diameter of the fired tiles to dry green tiles. The total shrinkage is then calculated from the relationship between fired tiles and wet green tiles. From Figure 4, we can see that the shrinkage of CB-added tiles tends to increase compared to the sample tiles. The air-dry and oven-dry shrinkage values of the control tiles and the 0.5%-CB tiles are almost identical, but the oven-dry shrinkage is slightly higher for 1.0%- and 1.5%-CB tiles. However, CB components such as cellulose acetate can absorb and store moisture, so we can tell that a longer drying time is required with more CBs introduced. In addition, sufficient drying time ensures that the tiles do not deform during the firing process due to moisture release. The firing shrinkage of control tiles is 9.16%, and the firing shrinkage for all tiles that are incorporated with CB is around 9.42%, which does not depict visible differences compared with other shrinkages. The total shrinkage also has a similar pattern

to firing shrinkage, where the control is 11.76% and CB tiles are around 12.5% to 12.6%. In addition, the reduced drying shrinkage of the 1.5%-CB tile affects its total shrinkage, with both the drying shrinkage and the total shrinkage declining by around 0.1% compared to the 1.0%-CB tile. However, the 1.5% CB tiles also displayed a relatively low water absorption in the subsequent water absorption test, so this phenomenon is not due to human error in measurements. Since the addition of CBs is limited, there is no clear and sharp positive linear tendency in the shrinkage rate.

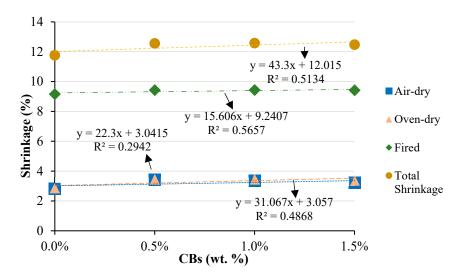


Figure 4. Shrinkage rate versus CB% by mass.

3.2.3. Water Absorption and Bulk Density

Water absorption is an essential physical characteristic of tiles which is related to open porosity and determines the category of potential applications of the tiles. Three replicates were used in this test. Figures 5 and 6 illustrate the water absorption and bulk density for the four batches of ceramic tiles. According to Australian standard AS 13006:2020, all tiles belong to the high water absorption group (>10%); hence, they can be categorised as group III for indoor wall tile purposes. The water absorption of the tiles with the addition of CBs has increased as opposed to the control tiles; this relationship matches the shrinkage rates. Overall, there is still a linear upward-sloping trend, but the average water absorption of 1.5%-CB tiles (14.39%) is lower than the 0.5%-CB tiles (15.29%). Although the small sample size may influence the average of water absorption, this result correlates with the shrinkage rates of 1.5%-CB tile. This may be due to the heterogeneity of CB components, such as the remaining tobacco and paper. A similar finding has been observed in related studies; Amin et al. [32] discovered that sewage sludge also increases water absorption with the increase in waste content, from roughly 12% for control tiles to more than 35% for tiles containing 30% sewage sludge content at a firing temperature of 1050 °C.

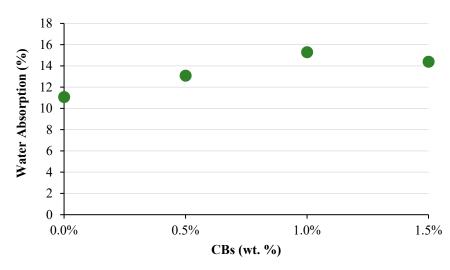


Figure 5. Water absorption versus CB% by mass.

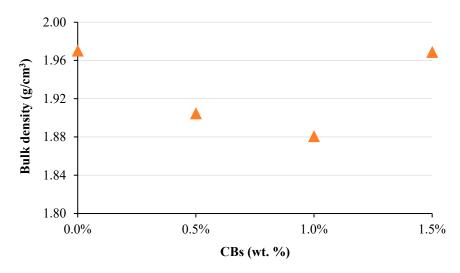


Figure 6. Bulk density versus CB% by mass.

3.2.4. Breaking Strength and Modulus of Rupture

The modulus of rupture is also known as the flexural strength; it is also one of the most crucial properties that indicate the quality of a ceramic tile. Three replicates were used in this test. Figures 7 and 8 reveal the breaking strength and modulus of rupture of ceramic tile. From the graphs, we can predict that increasing the incorporation amount of CBs is detrimental to the flexural strength, and we can clearly see that the tiles with 0.5% CBs have a slight improvement over the control tiles in Figures 7 and 8, which suggests that CBs have the ability to enhance or provide equivalent flexural strength to ceramic tiles. In another sewage sludge tile study, Jordán et al. [33] obtained similar data with a linearly decreasing trend in flexural strength; hence, the recovery amounts are restricted to comply with the strength requirements of tiles. As CB fibres are challenging to mix homogeneously with the powdered ceramic raw materials, the CB fibres will build up and create small voids internally, which will reduce the flexural strength. Subsequently, according to the guidelines of AS ISO 13006:2000, the minimum flexural strength of group III (water absorption > 10 %) must not be less than 15 MPa; therefore, only control tiles and 0.5%-CB tiles meet the requirements.

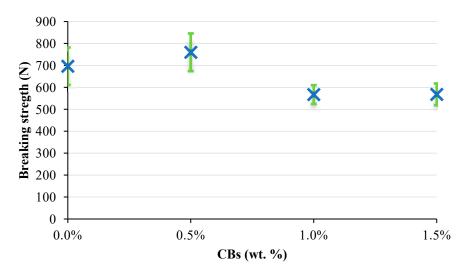


Figure 7. Breaking strength versus CB% by mass.

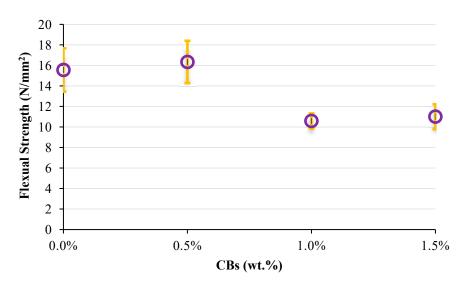


Figure 8. Modulus of rupture versus CB% by mass.

3.3. Energy Savings

Using Equation (6), the estimated energy savings during the firing process of 0.5%-, 1%-, and 1.5%-CB tiles were determined and are presented in Figure 9. The calculation results indicate that by firing tiles with 1% and 1.5% CBs, potential energy savings of 7.79% and 11.68% can be achieved in comparison to tiles containing no CBs. The organic content in CBs has a higher calorific value compared to the raw materials used in making tiles. Therefore, the partial replacement of bentonite with CBs results in further heat being released from the combustion of the waste during the firing process; this effect is produced by the decomposition of the organic matter in the CBs [34]. Relevant literature has also confirmed that the addition of CBs can save energy in the firing process. Mohajerani, Kadir, and Larobina [13] observed a 30.8% and 58.4% energy saving by recycling 2.5% and 5% CBs by mass, respectively, in fired clay bricks. Furthermore, Kurmus and Mohajerani [30] also used this calculation method and compared the estimation results with their laboratoryscale energy monitor outcomes; the comparison shows that these two methods only have a difference of around 1%, which provides a fairly close prediction. A number of factors can influence the actual production energy consumption, and the actual energy saving may differ from the simulated results. However, this method still gives us insight into the possible benefits of utilising CBs.

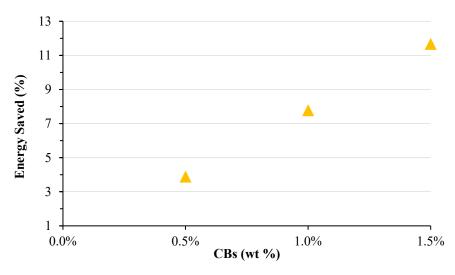


Figure 9. Calculated energy savings versus CB% by mass.

4. Conclusions

In this study, the addition of CBs to ceramic tiles was investigated, and the outcomes are promising for further exploration. Determination of the basic properties of ceramic tiles incorporated with 0.5%, 1.0%, and 1.5% CBs by total material dry mass was carried out. The experimental tests included chemical and mineralogical characterisation, density, shrinkage, bulk density, breaking strength, water absorption, and modulus of rupture, which were then compared with the Australian Standards for ceramic tiles.

The addition of CBs to ceramic tiles does not form new crystals inside the structure of tiles; nevertheless, other properties are sensitive to the CBs. Due to the fibrous nature of CBs, the drying shrinkage rises as the amount of CBs rises. However, firing shrinkage does not exhibit any apparent changes due to the low amount of CBs added. Although the effect on firing shrinkage is not noticeable, the density results show that the density decreases with increasing CB content. This density effect is probably due to the decomposition of CBs near the surface under high firing temperatures.

Water absorption is an important indicator to classify tiles. In general, water absorption will increase with the increasing CB content, as the cellulose acetate fibre combusts during the firing process. The water absorption of ceramic tiles increased from 11.07% for 0%-CB content tiles to 15.29% for 1.0%-CB content tiles, and all tested tiles satisfied the lowest requirement. The control tiles do not have a low water absorption feature, which is due to the variance to the formulation and manufacturing process. Hence, the CB tiles manufactured in this study can be categorised as group III-type tiles (water absorption > 10%).

The modulus of rupture also decreased with the increase in CB content due to increased porosity. However, the modulus of rupture of 0.5%-CB tiles is an outlier; it marginally enhances the flexural strength from 15.56 MPa to 16.33 MPa when compared with the control tiles. In contrast, the other tiles provide lower flexural strength compared with the control tiles.

The firing energy saved by incorporating 1% and 1.5% CBs into the tiles was estimated using the simulation equation. Estimated savings of 7.79% and 11.68% were found for tiles with 1% and 1.5% CBs by mass, respectively.

Ultimately, incorporating CBs into ceramic tiles has the potential to alleviate the waste management burden while providing suitable building materials. The addition of 0.5% CB in tiles can slightly improve flexural strength while qualifying for the group III classification of AS 13006 based on the water absorption and modulus of rupture, as well as being adequate to the requirements of wall tiles. Other aspects such as shrinkage are also in compliance with the typical tile shrinkage range.

In this project, we tried to replicate the industrial ram pressing method to produce tiles. Due to the lack of specialised machinery, many of the conditions and settings had to be compromised, such as the pressure in tile pressing having to be raised to around 40 MPa to obtain densified tiles. As the materials and manufacturing methods chosen do not provide outstanding performance like porcelain or stoneware tiles, more variance in experimental design and better manufacturing techniques should be undertaken to comprehend the effect of a better binder product in future experiments. The laboratory production should be as close as possible to industry processes and conditions rather than handmade standards. It is also recommended that the life cycle assessment, environmental impacts, and implementation guidelines be researched and developed in further investigations.

Author Contributions: Conceptualisation, Q.Y. and A.M.; methodology, Q.Y., A.K. and U.C.; validation, Q.Y., A.K. and U.C.; formal analysis, Q.Y.; investigation, Q.Y., A.K. and U.C.; data curation, Q.Y., A.K. and H.K.; writing—original draft preparation, Q.Y.; writing—review and editing, Q.Y., H.K., A.M., D.R., B.P. and P.T.; supervision, A.M., D.R., B.P. and P.T.; project administration, A.M. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Butt Out Australia Pty Ltd.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All data are presented in this article.

Acknowledgments: This work is part of an ongoing study on recycling waste in building materials. The authors would like to thank Butt Out Australia Pty Ltd, Visy Industries Australia Pty Ltd., the Australian Government Research Training Program (RTP) scholarship, RMIT University, the technical staff of the civil engineering laboratory at RMIT University, and Kris Coad from the School of Arts at RMIT University for their financial, in-kind, or technical support.

Conflicts of Interest: The authors declare no conflict of interest.

References

- 1. Kurmus, H.; Mohajerani, A. The toxicity and valorization options of cigarette butts. *Waste Manag.* 2020, *104*, 104–118. [CrossRef] [PubMed]
- Hoffman, S.J.; Mammone, J.; Rogers Van Katwyk, S.; Sritharan, L.; Tran, M.; Al-Khateeb, S.; Grjibovski, A.; Gunn, E.; Kamali-Anaraki, S.; Li, B.; et al. Cigarette consumption estimates for 71 countries from 1970 to 2015: Systematic collection of comparable data to facilitate quasi-experimental evaluations of national and global tobacco control interventions. *BMJ* 2019, 365, 12231. [CrossRef] [PubMed]
- Ng, M.; Freeman, M.K.; Fleming, T.D.; Robinson, M.; Dwyer-Lindgren, L.; Thomson, B.; Wollum, A.; Sanman, E.; Wulf, S.; Lopez, A.D.; et al. Smoking Prevalence and Cigarette Consumption in 187 Countries, 1980–2012. JAMA 2014, 311, 183–192. [CrossRef] [PubMed]
- Beaglehole, R.; Bonita, R.; Yach, D.; Mackay, J.; Reddy, K.S. A tobacco-free world: A call to action to phase out the sale of tobacco products by 2040. *Lancet* 2015, 385, 1011–1018. [CrossRef]
- Wallbank, L.A.; MacKenzie, R.; Beggs, P.J. Environmental impacts of tobacco product waste: International and Australian policy responses. *Ambio* 2017, 46, 361–370. [CrossRef] [PubMed]
- Belzagui, F.; Buscio, V.; Gutiérrez-Bouzán, C.; Vilaseca, M. Cigarette butts as a microfiber source with a microplastic level of concern. Sci. Total Environ. 2021, 762, 144165. [CrossRef] [PubMed]
- Puls, J.; Wilson, S.A.; Hölter, D. Degradation of Cellulose Acetate-Based Materials: A Review. J. Polym. Environ. 2011, 19, 152–165. [CrossRef]
- 8. Hoffmann, D.; Hoffmann, I.; El-Bayoumy, K. The less harmful cigarette: A controversial issue. A tribute to Ernst L. Wynder. *Chem. Res. Toxicol.* 2001, 14, 767–790. [CrossRef]
- 9. Novotny, T.E.; Slaughter, E. Tobacco Product Waste: An Environmental Approach to Reduce Tobacco Consumption. *Curr. Environ. Health Rep.* **2014**, *1*, 208–216. [CrossRef]
- 10. Hoffmann, D.; Hoffmann, I. The changing cigarette, 1950-1995. J. Toxicol. Environ. Health 1997, 50, 307–364. [CrossRef] [PubMed]
- 11. Slaughter, E.; Gersberg, R.M.; Watanabe, K.; Rudolph, J.; Stransky, C.; Novotny, T.E. Toxicity of cigarette butts, and their chemical components, to marine and freshwater fish. *Tob. Control.* **2011**, *20*, i25. [CrossRef] [PubMed]
- 12. Torkashvand, J.; Farzadkia, M. A systematic review on cigarette butt management as a hazardous waste and prevalent litter: Control and recycling. *Environ. Sci. Pollut. Res.* **2019**, *26*, 11618–11630. [CrossRef]
- 13. Mohajerani, A.; Kadir, A.A.; Larobina, L. A practical proposal for solving the world's cigarette butt problem: Recycling in fired clay bricks. *Waste Manag.* **2016**, *52*, 228–244. [CrossRef]

- 14. Kurmus, H.; Mohajerani, A. Recycling of cigarette butts in fired clay bricks: A new laboratory investigation. *Materials* **2020**, 13, 790. [CrossRef]
- 15. Kadir, A.A.; Mohajerani, A. Recycling cigarette butts in lightweight fired clay bricks. *Proc. Inst. Civ. Eng. Constr. Mater.* 2011, 164, 219–229. [CrossRef]
- Mohajerani, A.; Hui, S.Q.; Shen, C.; Suntovski, J.; Rodwell, G.; Kurmus, H.; Hana, M.; Rahman, M.T. Implementation of recycling cigarette butts in lightweight bricks and a proposal for ending the littering of cigarette butts in our cities. *Materials* 2020, 13, 4023. [CrossRef]
- 17. Rahman, M.T.; Mohajerani, A. Use of bitumen encapsulated cigarette butts in stone mastic asphalt. *Constr. Build. Mater.* **2020**, 261. [CrossRef]
- 18. Rahman, M.T.; Mohajerani, A.; Giustozzi, F. Possible use of cigarette butt fiber modified bitumen in stone mastic asphalt. *Constr. Build. Mater.* **2020**, *263*. [CrossRef]
- 19. Rahman, M.T.; Mohajerani, A.; Giustozzi, F. Possible recycling of cigarette butts as fiber modifier in bitumen for asphalt concrete. *Materials* **2020**, *13*, 734. [CrossRef] [PubMed]
- d'Heni Teixeira, M.B.; Duarte, M.A.B.; Raposo Garcez, L.; Camargo Rubim, J.; Hofmann Gatti, T.; Suarez, P.A.Z. Process development for cigarette butts recycling into cellulose pulp. *Waste Manag.* 2017, 60, 140–150. [CrossRef] [PubMed]
- Gómez Escobar, V.; Maderuelo-Sanz, R. Acoustical performance of samples prepared with cigarette butts. *Appl. Acoust.* 2017, 125, 166–172. [CrossRef]
- 22. Dondi, M.; Raimondo, M.; Zanelli, C. Clays and bodies for ceramic tiles: Reappraisal and technological classification. *Appl. Clay Sci.* **2014**, *96*, 91–109. [CrossRef]
- World Production and Consumption of Ceramic tiles. Available online: https://www.ceramicworldweb.it/cww-en/statisticsand-markets/world-production-and-consumption-of-ceramic-tiles-2017/ (accessed on 6 October 2021).
- 24. World Production and Consumption of Ceramic Tiles. The Figures for 2019 and Forecasts for 2020. Available online: https://www.ceramicworldweb.it/cww-en/statistics-and-markets/world-production-and-consumption-of-ceramic-tiles-the-figures-for-2019-and-forecasts-for-2020/ (accessed on 6 October 2021).
- Zanelli, C.; Conte, S.; Molinari, C.; Soldati, R.; Dondi, M. Waste recycling in ceramic tiles: A technological outlook. *Resour. Conserv. Recycl.* 2021, 168, 105289. [CrossRef]
- Yuan, Q.; Mohajerani, A.; Kurmus, H.; Smith, J.V. Possible Recycling Options Of Waste Materials In Manufacturing Ceramic Tiles. *Int. J.* 2021, 20, 73–80. [CrossRef]
- 27. Haiying, Z.; Youcai, Z.; Jingyu, Q. Study on use of MSWI fly ash in ceramic tile. J. Hazard. Mater. 2007, 141, 106–114. [CrossRef]
- 28. Kurmus, H.; Mohajerani, A. Leachate Analysis of Heavy Metals in Cigarette Butts and Bricks Incorporated with Cigarette Butts. *Materials* **2020**, *13*, 2843. [CrossRef]
- 29. Singer, F.; Singer, S.S. *Industrial Ceramics*; Springer Netherlands: Dordrecht, The Netherlands, 1963. [CrossRef]
- 30. Kurmus, H.; Mohajerani, A. Energy savings, thermal conductivity, micro and macro structural analysis of fired clay bricks incorporating cigarette butts. *Constr. Build. Mater.* **2021**, *283*, 122755. [CrossRef]
- Monfort, E.; Mezquita, A.; Granel, R.; Vaquer, E.; Escrig, A.; Miralles, A.; Zaera, V. Analysis of energy consumption and carbon dioxide emissions in ceramic tile manufacture. *Boletín Soc. Española Cerámica Vidr.* 2010, 49, 303–310.
- Amin, S.K.; Abdel Hamid, E.M.; El-Sherbiny, S.A.; Sibak, H.A.; Abadir, M.F. The use of sewage sludge in the production of ceramic floor tiles. *HBRC J.* 2018, 14, 309–315. [CrossRef]
- Jordán, M.M.; Almendro-Candel, M.B.; Romero, M.; Rincón, J.M. Application of sewage sludge in the manufacturing of ceramic tile bodies. *Appl. Clay Sci.* 2005, 30, 219–224. [CrossRef]
- 34. Gualtieri, M.L.; Gualtieri, A.F.; Gagliardi, S.; Ruffini, P.; Ferrari, R.; Hanuskova, M. Thermal conductivity of fired clays: Effects of mineralogical and physical properties of the raw materials. *Appl. Clay Sci.* **2010**, *49*, 269–275. [CrossRef]