Assessing the use of Biochar as a Cementitious Replacement in Structural Concrete

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Dissertation submitted to
the Faculty of Science & Engineering
University of Limerick

In partial fulfilment of the requirement for the
Degree of Masters of Engineering in
Civil Engineering

March 2023

It is hereby declared that this report is entirely my own work, unless otherwise stated, and that all sources of information have been properly acknowledged and referenced. It is also declared that this report has not previously been submitted, in whole or in part, as part fulfilment of any module assessment requirement.

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Abstract
Cement production accounts for between 4% - 8% of total global anthropogenic CO_2 emissions. The use of supplementary cementitious materials is seen as a way to reduce the environmental burdens of cement production. The Irish Government's 2023 Climate Action Plan mandates the use of low-carbon cement on all public body construction projects as well as supporting the research and development of novel binders and fillers in low-carbon cement. This paper investigates the use of locally produced biochar, a porous carbon-based material derived from the thermal decomposition of biomass, as a partial cement replacement in structural concrete. A "Cradle to Gate" life cycle assessment undertaken found that a carbon-neutral can be achieved with 20% by weight biochar addition. The physical and chemical properties of biochar from local Irish Timber and Juncus waste biomass sources were characterised using elemental analysis, scanning electron microscopy, and thermogravimetric analysis before being added to the concrete at varying cement replacement levels from 0% - 6%. The effects of its introduction on the mechanical properties and durability of concrete were investigated through experimental analysis. Biochar was found to reduce the workability of concrete significantly. Both sources of biochar caused reductions in compressive strength when compared to the control, however, the addition of 4% Juncus biochar led to an increase in flexural and split tensile strength. It was also found that both the Timber and Juncus biochar increased the permeability of the concrete by 45% and 20% respectively.

Keywords: Concrete, Sustainability, Biochar, Strength, Permeability

1. Introduction

Climate change poses a great threat to human life and the earth's ecosystems (Rama *et al.* 2022), however, mitigating its effects is possible through the decarbonisation of the world's major industries. Cement manufacturing alone is responsible for a quarter of all industry CO_2 emissions (Andrew 2018), and between 4% - 8% of the world's production of anthropogenic CO_2 emissions (Amran et al. 2021). Reducing these emissions will be key to achieving the goal of carbon neutrality by 2050 (UN 2015).

Concrete is the second most used material in the world after water, with over 25 billion tonnes produced annually, exceeding 3t for every person on the planet (Akhtar and Sarmah 2018). The fuel burning and chemical processes in the production of ordinary Portland cement (OPC) lead to high levels of CO_2 being released into the atmosphere. While cement typically accounts for just 20% of the total concrete volume, it is responsible for approximately 90% of the total emission of CO_2 (Yang *et al.* 2015). Approximately 1 tonne of CO_2 is released into the atmosphere during the production of 1 tonne of OPC (Campos *et al.* 2020). Such release of CO_2 causes serious environmental hazards such as ozone depletion and global warming (Danish *et al.* 2021).

With the increasing scarcity of natural resources and the environmental effect of OPC production, significant research has focused on the replacement or partial replacement of OPC with supplementary cementitious materials (SCMs) (Danish *et al.* 2021). Using circular economy principles, recycled and waste materials such as ground granulated blast furnace slag (GGBS) and fly ash have been used successfully as SCMs in blended cement (Akhtar and Sarmah 2018). As a result, more than 80% of the cement used in Ireland today is blended (CMI 2014). As part of the 2023 Climate Action Plan, the Irish Government have outlined the mandatory use of low-carbon cement on all public body construction projects from 2023 onwards, with the aim of reducing the embodied carbon in construction materials produced and used in Ireland by 10% by 2025. The government have also pledged to support the research and development of low-carbon cement through the introduction of novel binders and fillers (Department of the Environment 2023)..

There has been a recent drive within the construction material research community regarding the use of natural or biobased materials in concrete. These materials have the added benefit of sequestering biogenic carbon within the concrete by preventing its release back into the atmosphere (Mensah *et al.* 2021). One material being considered is biochar. Biochar (BC) is a porous carbon-based material derived from biomass. It is produced through a process called pyrolysis. Its production is managed to ensure that the contained carbon is stored as a long-term carbon sink, and it is not intended to be burnt for energy (EBC 2022). Pyrolysis is a process that involves the thermochemical decomposition of organic material at high temperatures in the absence of oxygen (Weber and Quicker 2018). This process releases three products: syngas, bio-oil, and biochar (Legan *et al.* 2022).

In principle, biochar can be produced from all sources of organic matter, with each source providing a product of unique properties. The porosity, surface area, and chemical composition of each batch of biochar differ depending on the source and the conditions of pyrolysis (Weber and Quicker 2018).

The global carbon cycle is comprised of various flows and pools of carbon that are part of the Earth's ecosystem. The carbon within these pools has different lifecycles, which

allows for flow between them. To decrease the amount of carbon in the atmosphere, the carbon in the active pool should be transferred to a passive pool comprising of inert or stable carbon (Kwapinski *et al.* 2010). The production of biochar can facilitate the flow of carbon from the active to the passive pool. Chen *et al.* (2022) carried out a cradle-to-gate LCA on concrete blocks containing different concentrations of BC and recycled aggregates (RCA). It was found that a 10% by-weight replacement of OPC with BC sequestered 119 kg CO_2e per tonne of concrete. It was also found that a carbon-negative block could be produced with a 30% dosage of biochar.

Of the biomass sources reviewed in the literature, it was found that biochar derived from wood waste performs best in terms of enhancing the concrete's mechanical and durability properties (S. Gupta *et al.* 2018; Liu *et al.* 2022). Wood waste has a well-established waste stream, so it is important to identify alternative waste organic materials that are readily available while not negatively affecting the mechanical and durability properties of the concrete. Juncus, more commonly known as rushes, has been identified as an indigenous biomass source with a high abundance and no current waste stream.

1.1. Aims and Objectives

The aim of the research presented in this paper is to investigate the use of locally produced biochar as a cementitious replacement and assess its effect on the mechanical and durability properties of concrete. To reach this aim, the following objectives were set:

- Assess biochar's ability in reducing the global warming potential of cement.
- Procure locally produced Timber and Juncus biochar and assess their thermal stability, microstructure, and elemental composition.
- Develop a representative concrete mix to act as a control, and several blended mixes by varying the amount of biochar used as a cement replacement.
- Assess the effect biochar has on concrete's morphology, workability, compressive, flexural, and split tensile strength as well as its effect on concrete's resistance to chloride ion penetration.

1.2. Life Cycle Analysis

An A1-A3 (Cradle to Gate) life cycle analysis (LCA) was conducted to assess the biochar's performance in reducing the global warming potential (GWP) of blended cement. This LCA is concerned with the carbon emissions (kgCO_2e) released during raw material extraction, processing, manufacturing, and transportation of materials between these processes, focusing on the addition of Timber biochar to CEM II/A. A GWP value for the cement was obtained from an Environmental Product Declaration developed by Cement Manufacturers Ireland (CMI) (CMI 2022b). A GWP value for the Timber biochar was obtained from an LCA conducted by Greenbelt (Timber BC producers) as per the European Biochar Commission Guidelines. The addition of 2%, 4%, and 6% BC showed a reduction in net carbon emissions of 8%, 17,% and 26% respectively. It was also found that a carbon-neutral cement could be achieved with a 20% by-weight BC replacement (Fig. 1).

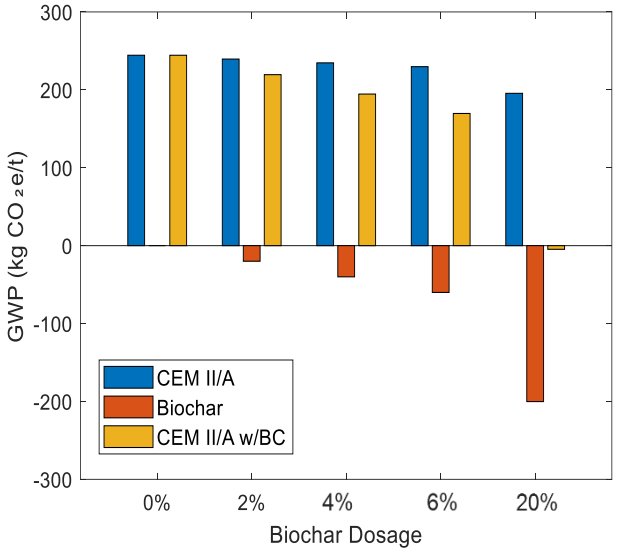

Figure 1: Life Cycle Assessment for cement with varying Timber BC replacement levels

2. Literature Review

This literature review concentrates on the findings of several studies assessing the fresh, mechanical and durability properties of biochar-added concrete.

Many of the mix designs reviewed in the literature presented mixes of unconventional proportions. For example, Sirico *et al.* (2021) investigated a mix with a ratio of fine to coarse aggregates of 2:1 (Appendix A). This can create issues regarding the workability of the concrete.

2.1. Fresh Properties

2.1.1. Workability

The workability of concrete with biochar as a SCM has been highlighted as a common issue across the literature. Multiple studies recommended limits of 5% cement replacement due to workability concerns. Biochar reduces the workability of concrete due to its porous nature and high water absorption (Akinyemi and Adesina 2020). The rate of decrease increases with increasing biochar content. (Liu *et al.* 2022). The high carbon content of biochar has also been reported to result in a higher demand for water to achieve good workability (Akinyemi and Adesina 2020). The reduction in workability is also related to the pyrolysis temperature in which the biochar was manufactured and the particle size of the biochar. The workability reduces in the presence of finer biochar particles produced at higher temperatures (Chen *et al.* 2020). Souradeep Gupta *et al.* (2018a) found that the average fluidity of BC concrete was reduced by 10 - 30% depending on the type of BC and the percentage of cement being replaced. This reduction in workability has been overcome by either increasing the water/cement (w/c) ratio, using an optimal superplasticizer dosage, or pre-saturating the biochar (Danish *et al.* 2021).

A study by Maljaee *et al.* (2021) discovered that in order to provide enough free water in a concrete mix, the w/c ratio should be increased from 0.4 to 0.48 when just 1.5% of the cement is replaced with biochar. The downside to this solution is the reduction in the strength of concrete.

Superplasticizers can also be used to limit the negative effects of BC on workability with the added benefit of maintaining concrete's mechanical properties. (Sirico *et al.* 2021). One study by S. Gupta *et al.* (2018) reported a 26% and 14% increase in the superplasticizer needed to maintain the flow at 5% biochar addition made from mixed wood and food waste respectively. However, there are conflicting results in the literature on the success of pre-saturating biochar before adding it to the concrete mix. Therefore, more research is needed in this area before it can be characterized as a method to improve the workability of biochar-added concrete.

2.1.2. Air Content & Setting Time

The addition of BC reduces the fresh density of mortars and increases the air content. This is because BC has a lower specific gravity than common mineral admixtures and has pores that hold air or water (Tan *et al.* 2021).

The addition of biochar in cementitious composites has been found to reduce both initial and final setting times (Souradeep Gupta *et al.* 2018a). This is because the biochar's filler effect and free water reduction contribute to the overall cohesiveness of the mixture. The accelerated hydration reaction also contributes to the reduction in setting time (Akinyemi and Adesina 2020). Souradeep Gupta *et al.* (2018a) found that the addition of 2% BC reduced setting time by 66% as the water retention capacity and filling effect of the biochar reduced leakage and increased the aggregation of mortar.

2.2. Hardened/Mechanical Properties

2.2.1. Compressive Strength

The compressive strength of biochar-added concrete varies according to biochar type, content and pyrolyzing temperature. The trend in the literature for the optimal biochar addition for compressive strength was therefore uneven (Mensah *et al.* 2021). The w/c ratio and curing conditions also affect the compressive strength as is the case with standard concrete. There is consensus in the literature that optimizing the BC dosage can enhance the compressive strength of a concrete mix. According to Souradeep Gupta *et al.* (2018b) this is due to the fine size of the biochar particles in comparison to the mixed cement, sand and aggregate grains. Such fine particles play an important role in filling the macropores within the mix leading to compactness and greater performance in transferring structural loading. Biochar's porous structure and high surface area promote the hydration reaction within cementitious composites which can also improve the compressive strength of concrete (Liu *et al.* 2022).

The recommended optimal biochar dosage differs amongst researchers due to the variations as outlined above. Most papers suggest that the compressive strength values are expected to peak at 2% biochar addition and remain above the control strength until 4% where they will then decrease with increasing biochar dosage (Aneja *et al.* 2022).

A study by S. Gupta *et al.* (2018) revealed that a 1% addition of biochar as a filler increased the compressive strength of mortar by 16.2% in comparison to the control mix. Additionally, without changing other parameters, increasing the percentage of biochar to 2% and 5% decreased the compressive strength by 5.23% and 13.21% respectively when compared to the control mix. On the contrary, a study by Sirico *et al.* (2021) showed that the addition of biochar made from woodchip has a positive effect on the compressive strength of concrete up to 5% cement replacement. This study also highlighted how the positive effects of biochar addition in concrete are more evident when dry curing takes place rather than wet curing. This is due to the water restrained in the biochar pores being gradually released over time promoting the development of hydration reactions in the concrete. 2.5% biochar dosage resulted in a 28-day compressive strength increase of just 5% under wet curing conditions and 25% for dry curing.

Sirico *et al.* (2021) also discovered the development of concrete compressive strength after 28 days was greater in the 5% biochar-added specimens than in the conventional concrete. For example, when samples were dry cured, the improvement in compressive strength from 28-365 days was equal to 12% in comparison to just a 2% increase in conventional concrete.

2.2.2. Flexural Strength

The addition of BC does not dramatically influence the flexural strength of the concrete unlike the compressive strength (Tan *et al.* 2021). This is also in contrast to conventional SCMs such as silica fume which can have significant effects on the flexural strength of concrete. This is due to the low pozzolanic reactivity of the BC material (Danish *et al.* 2021). A study by Gupta *et al.* (2020) supported this by finding the maximum flexural strength gain can be achieved at just 0.5% biochar addition with the flexural strength decreasing with increasing biochar content thereafter. This reduction in flexural strength is believed to be caused by the BC pores introduced in the tensile plane resulting in more weak interfacial zones, leading to pores in these zones becoming more vulnerable to cracking (Sirico *et al.* 2021).

In contrast, studies from Souradeep Gupta et al. (2018a) and Tan et al. (2021) suggest that a 1%-3% addition of BC was advantageous for improving flexural strength similar to the dosage used for compressive strength. Pore sizes are reduced due to the filling effect that additional biochar provides. This reduced no. of macropores helps avoid the risk of early fracture. (Danish et al. 2021). This observation corresponds to Cosentino et al. (2019) who discovered that BC can alter the crack path in a concrete composite. This is attributed to the strong interaction between the biochar

and the cement which promotes the crack resistance and toughness of the concrete.

2.2.3. Split Tensile Strength

Studies have shown the addition of BC has been found to enhance the split tensile strength of the concrete with a max increase of 0.5% (Akinyemi and Adesina 2020). The split tensile strength of concrete reduces with higher BC content from most sources, however, wood waste maintains flexural strength in dosages up to 2% cement replacement. Sirico *et al.* (2021) studied the effect of biochar from wood waste on structural concrete rather than just cement mortar like many of the other studies. They found that the maximum beneficial dosage of BC could reach 5%.

An excessive concentration of woodchip biochar resulted in a significant decrease in tensile strength. This was attributed to the fact that the addition of biochar introduces pores, leading to additional weaker areas in the tensile plane due to inhomogeneity (Akhtar and Sarmah 2018; Gupta et al. 2020). This is consistent with a study by Qin *et al.* (2021) which found that an excessive biochar dose (greater than 6.5%) would lead to an aggregation effect and form local weak zones resulting in overall concrete strength losses.

2.3. Durability Properties

A review of the literature suggests that the research carried out on the durability properties of biochar concrete is limited in comparison to the mechanical properties. These durability properties which include chloride and sulphate ion migration, water penetration and electrical resistivity are all dependent on the permeability of the concrete and are all important to better assess the long-term performance of biochar concrete composites (Yang and Wang 2021).

Studies have shown that the addition of biochar leads to an increase in the diffusion of chloride ions in cement mortars. This is accredited to the increase in porosity due to the BC (Yang and Wang 2021). This is in line with a study from Akhtar and Sarmah (2018) who found that the compactness of mortar decreases with increasing incorporation of biochar. In contrast, Yang and Wang (2021) found that biochar-blended mortar can create more stable carbonation products which form a dense structure to prevent the diffusion of chloride ions and thus enhance the resistance to chloride ion attack. Studies have shown the introduction of BC in mortar also reduces water permeability mainly through the filling effect and by promoting hydration. Gupta *et al.* (2020) found that the replacement of 3% cement with biochar could reduce water penetration by 33%.

3. Materials and Methods

3.1. Cement & Aggregates

Portland Limestone Cement CEM II/A-L 32,5N manufactured by Irish Cement in Castlemungret, Co. Limerick was used for all concrete mixes as part of this research. This cement is produced by grinding a combination of cement clinker, selected limestone and grinding aids as well as a small quantity of gypsum. Locally sourced coarse grain well-graded sand with a max particle size of 4.75mm (82.72% passing the 2.36mm sieve) was used for all laboratory testing. A particle size distribution analysis was carried out on the sand to ensure it met the requirements for sand used in an applicable concrete mix. The coarse aggregate consisted of 10mm high polished stone value (PSV) limestone chips quarried and crushed in O'Connell Quarries Ardncarusha, Co. Clare.

The moisture content of both the fine and coarse aggregates were taken at the beginning of every lab trial. The coarse aggregate was very dry and was found to have a moisture content of just 0.6%. The moisture content of the sand was monitored closely as it increased significantly towards the bottom of the container it was stored in. A moisture content of 5.2% was calculated for the first mix. This increased to 6.8% over the testing period.

3.2. Biochar

3.2.1. Production & Collection

Two sources of local Irish biochar were used in this study. The first was derived from Sitka Spruce pellets harvested in Co. Roscommon. The BC was prepared in an industrial pyrolysis unit at 650°C-700°C with a residence time of 35 minutes. The other source of biochar was produced in Co. Clare and was derived from Juncus or more commonly known as Rush. This was prepared in a mobile pyrolysis unit at 380°C- 450°C with a residence time of 5 minutes.

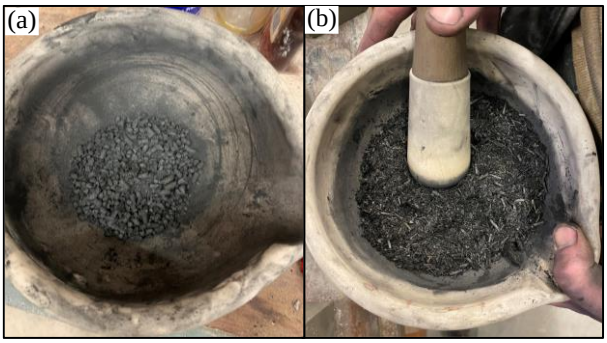

Figure 2: Raw (a) Timber (b) Juncus biochar in mortar and pestle

Several physical–chemical analyses were carried out on the biochar to understand its morphology, structure, and composition.

3.2.2. Scanning Electron Microscopy Analysis

The Hitachi SU-70 Scanning electron microscope was used to characterise the morphology of the biochar samples. The samples were first dried overnight at 50°C in a vacuum oven. The samples are placed on a conductive carbon tape, before being coated in a microscopic gold layer to enhance the imaging of the samples. The images were taken with an accelerating voltage of 10kV and magnifications ranging from x500 to x2500. Scanning electron microscopy (SEM) analysis was performed on both biochar and biochar-added concrete samples.

3.2.3. Elemental Analysis

The Hitachi SU-70 was also used to carry out an energy dispersive X-ray (EDX) analysis on the samples. The EDX analysis was carried out to determine the chemical compositions of the biochar.

3.2.4. Thermogravimetric Analysis

Thermogravimetric analysis of two biochar samples was performed using the TA Instruments SDT Q600. Nitrogen gas was flushed at a rate of 100ml/min. The platinum crucibles were ¾ filled giving a mass ranging from 10.3-18.5mg. The heating rate was set to 20°C/min and maintained until samples reached 1100°C. The weight loss of the samples corresponding to the rise in temperature was recorded and plotted to produce the TGA curves shown in Fig.3.

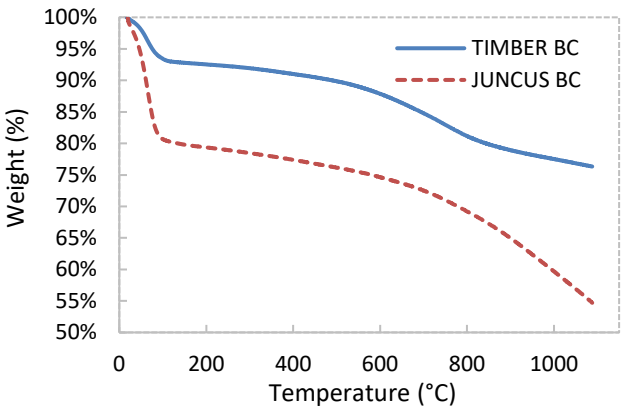

Figure 3: TGA of Juncus and Timber biochar

The Juncus TGA curve shows a steep initial drop as any water in the sample is evaporated. The 20% drop in weight over the first 200°C confirms the 20% moisture content calculated in lab tests. The rate of decomposition of volatiles in the sample slowly increases once the pyrolysis temperature of 380-450°C is reached. Full thermal degradation is not reached at 1000°C as the sample weight continues to drop steadily.

The Timber BC TGA follows a similar trend at the beginning of the test. Any moisture in the sample is removed at 200°C. The 8% weight loss confirms the moisture content of the sample. The decomposition then remains steady until the pyrolysis temperature of 650°C is reached. The remaining volatile organics are then removed until 800°C where the curve begins to plateau, marking the end of the pyrolysis process.

3.2.5. Absorption of the Aggregates

The saturated surface dry (SSD) specific gravity (SG) of the biochar and fine aggregates were found using the paper towel method as outlined in ASTM C1761 (ASTM 2017) and Barissov (2021). This method is designed for lightweight aggregates and is not optimum for the testing of sand, however, in the absence of other specific testing equipment it provides a value for the absorption of both the biochar and the sand. This was required to be certain of the free water available in the concrete mix. The absorption of the sand and biochar was found to be 3.27% and 96.62% respectively.

3.3. Biochar Added Concrete

3.3.1. Mix Design

The literature has identified that the addition of biochar reduces the workability of concrete. Several mix designs were trialled to ensure a mix of suitable consistency was developed for the control. The control needed to allow for the expected reduction in workability with the addition of BC, as the use of plasticizers is outside the scope of this paper.

A mix was established based on an initial standard C30/35 ready mix widely used across the Irish concrete industry. A mix ratio of 1: 2.36: 3: 0.55 (cement: fine: coarse: water) was used for the control. A percentage of cement was then substituted with BC for each replacement level (Table 1). The water-cement (w/c) ratio of 0.55 was kept constant for all mixes. The mix design was adjusted accordingly to accommodate the change in aggregate moisture content and absorption.

Table 1: Concrete Mix Designs

Mix	Cement (kg/m^3)	Biochar (kg/m^3)	Fine Aggregate (kg/m^3)	Coarse Aggregate (kg/m^3)	Water (kg/m^3)
Control	350	0	810	1065	192.5
2% BC Timber	343	7	810	1065	192.5
4% BC Timber	336	14	810	1065	192.5
6% BC Timber	329	21	810	1065	192.5
4% BC Juncus	336	14	810	1065	192.5

3.3.2. Concrete Mixing

A mixing sequence was established to achieve a homogenous mixture in accordance with BS 1881-125:2013 (BSI 2013). Materials were mixed in an ELE Concrete Paddle Mixer to ensure even and thorough mixing. The inside of the mixer was dampened using a moist cloth and any excess water was removed. Fine and coarse aggregates were first dry mixed for 30 seconds. Half of the water was then added and allowed to mix for 2

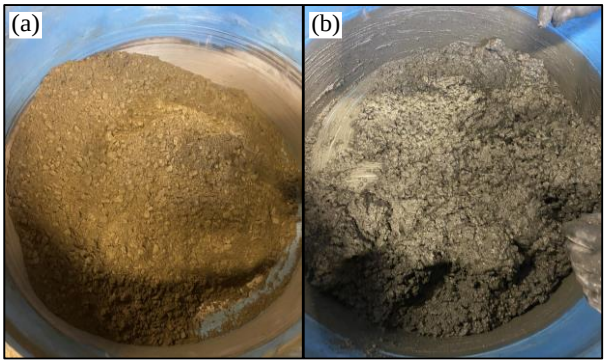

Figure 4: Paddle mixer with (a) dry aggregates (b) concrete after mixing

minutes. The mixer was then powered off and the contents were left covered for 5 minutes. The cement was then added and mixed for another 30 seconds. Any material adhering to the blades or sides of the pan was cleaned off before mixing recommenced. The remaining water was then added, and mixing was continued for 2 minutes. The consistency of the mix was monitored closely at each stage. For subsequent biochar-added concrete mixes, the biochar was pre-mixed with the cement and the same procedure was followed. Concrete sitting in the paddle mixer was always hand-mixed using a trowel to ensure uniformity before sampling.

3.3.3. Workability

The workability of the concrete was determined using the slump test. The slump testing procedure was performed in line with I.S. EN 12350-2:2019 (CEN 2019a). The procedure included filling the cone in 3 layers, each layer receiving 25 strokes of the compacting rod. The cone was raised in 2-5s in a steady upward lift. The slump *h* was measured immediately by measuring the difference between the height of the cone and the highest point of the slumped test specimen.

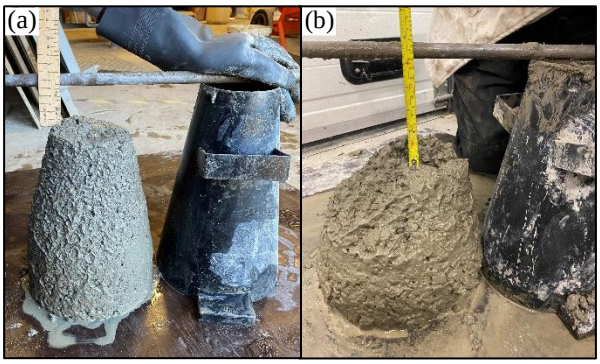

Figure 5: Slump test of (a) 4% Juncus BC Mix (b) Control Mix

3.3.4. Specimen Preparation & Curing Conditions

All moulds were coated with a non-reactive mould release agent prior to sampling. The specimens were prepared in accordance with I.S. EN 12390-2:2019 (CEN 2019c). The concrete was regularly remixed during the sampling stage. Specimens were compacted using a vibrating table in layers of no greater than 100mm and for a duration of 15 seconds on each layer. The cylinders were tapped with a rubber mallet after each layer to release any air bubbles adhered to the mould wall.

The samples were left in the moulds to cure for 1 day before being placed in a curing tank. It was not possible to cure the specimens at 20℃. The tank's temperature was monitored regularly and found to be in the range of 11℃-12℃. The specimens were cured for varying durations: 1 day, 7 days, 14 days, and 30 days.

3.3.5. Compressive Strength

Compressive tests on 100mm cube specimens were carried out in accordance with I.S. EN 12390-3:2019 (CEN 2019b). A Controls AUTOMAX PRO 3000kN

Figure 6: Concrete cubes from varying mixes

compression testing machine was used to apply the load to 1% accuracy at a constant rate until specimen failure. 10 cube specimens were tested for each mix design, 1 cube after 1 day, 2 at 7 and 14 days and 5 at 30 days.

3.3.6. Tensile Strength

Tensile splitting tests were performed to assess the concretes behaviour in tension in line with I.S. EN 12390-6:2009 (CEN 2009). A Controls Automax testing machine with Model 50-C9070/C splitting tensile device was used. 150mm x 300mm cylindrical test specimens were placed horizontally in the testing device between two parallel load beams. Packing strips are inserted between the load beams and the specimen before a constant compressive force of 0.05 MPa/s is applied longitudinally until failure. All tests were conducted after 30 days moist curing.

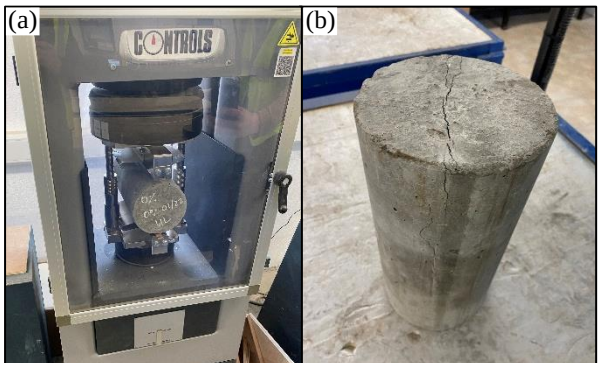

Figure 7: Tensile splitting test (a) test apparatus (b) cracked specimen after failure

3.3.7. Flexural Strength

The flexural strength of the concrete was tested using the 3-point beam flexural test. 100x100x500mm concrete beams were placed over a net span of 450mm and loaded in the centre with a test die. A Zwick Roell Z100 materials

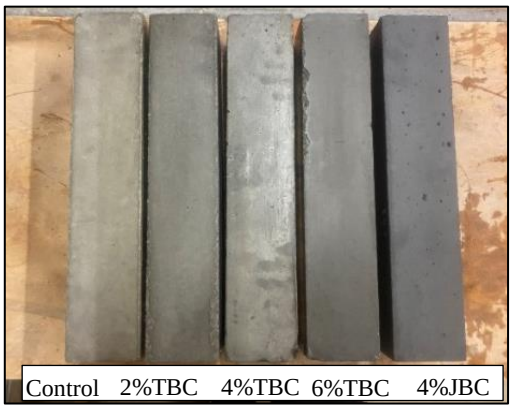

Figure 8: Concrete beams from varying mixes

testing machine was used to apply the force at a constant rate of 2 mm/min until failure. The flexural strength is calculated based on the maximum load applied to the specimen and its dimensions. All tests were conducted after 30 days moist curing.

3.3.8. Chloride Migration

The permeability of the concrete was determined by carrying out the chloride migration test in accordance with I.S. EN 12390-18:2021 (CEN 2021). The principle of this test involves placing a specimen of concrete between a chloride free and a chloride containing alkaline solution. An electric current is passed through the specimen which drives the flow of chloride ions into the concrete. The test is run for a given period often >24hrs. When complete the specimen is split and sprayed with a suitable colour indicator solution. The chloride migration coefficient is calculated based on the measured depth of penetration, the magnitude of the applied voltage and other parameters, see Equation 1.

$$M_{nss} = \frac{0.0239(T)h}{(U)t}\left(x_d - 0.0186\sqrt{\frac{(T)hx_d}{U}}\right) \quad [1]$$

Where:
M_{nss} = *Chloride Migration Coefficient* ($\times 10^{-12}\, m^2/s$)
T = *Temperature of the Anolyte* (°K)
U = *Applied Voltage* (V)
t = *Test Duration* (h)
h = *Height of Specimen* (mm)
x_d = *Average Depth of Penetration* (mm)

In this case, the test was run for 24 hours at 15 V, 0.3M NaOH was used for the anolyte and 5% NaCl solution was used for the catholyte. Silver nitrate was used as the indicator solution.

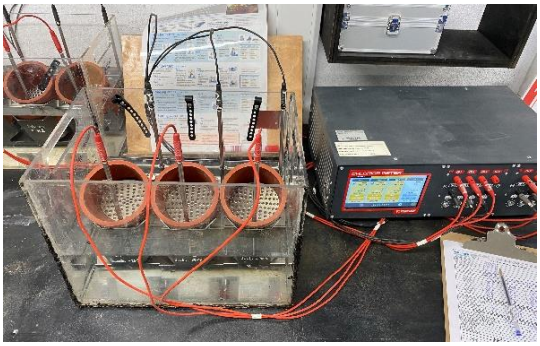

Figure 9: Rapid chloride migration test apparatus

4. Results and Discussion

4.1. Scanning Electron Microscopy Analysis

4.1.1. Biochar

The SEM images of the biochar show a wide dimensional distribution of particles in both sources. The heterogeneous pore sizes ranged from 2 µm – 20 µm across both samples. Biochar particles are elongated in shape with ridges on the surface. The Timber biochar particles appear to have a smoother surface and are more densely configurated than that of the Juncus (Fig.10). The increased number of ridges

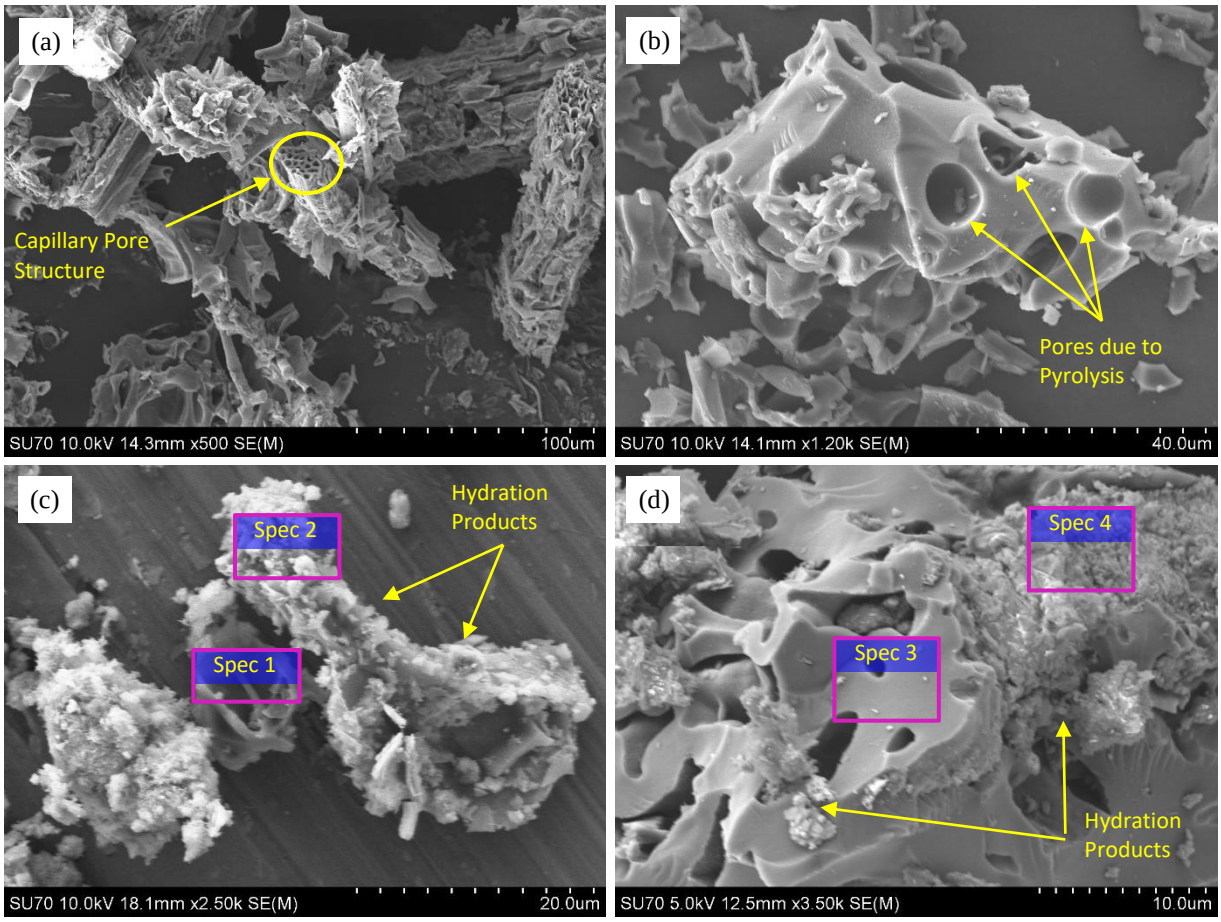

Figure 10: SEM images of (a) Juncus and (b) Timber biochar. Images (c) and (d) show biochar concrete composites with Juncus and Timber BC respectively, the hydration products in the biochar pores and EDX spectrums taken can be seen.

and pores observed in the Juncus biochar increases both the surface area and roughness.

Honeycomb macro-pores can be seen on the surface of both biochar samples. These pores are a product of pyrolysis from the release of volatiles, organics, and the remanence of the biological capillary structure of the feedstock (Fig.10(a)). These cellular pores are active in water absorption, thus modifying the effective w/c ratio. The water is later released facilitating internal curing during the hardening stage of the concrete (Gupta and Kua 2018).

4.1.2. Biochar Added Concrete.

SEM images of the biochar concrete composites show the high compatibility between the biochar particles and cement matrix. Hydration products can be seen to have formed in the BC pores of both sources with additional crystal growth in the Juncus sample due to its greater surface roughness and porous nature (Fig.10(c)). As well as improved bonding, the increased hydration products also lead to a higher level of carbon sequestration. This is due to the additional reactants available for carbonation (Liu *et al.* 2022).

4.2. Elemental Composition

4.2.1. Biochar

EDX elemental composition shows there is a greater range of elements present in the Juncus biochar in comparison to the Timber source. The additional metallic ions such as Sodium, Calcium, Potassium and Magnesium found in the Juncus BC (Table 2) may lead to the formation of ionic bonds in the BC-added concrete resulting in strength increases (Section 4.6). This is advantageous in comparison to the Timber BC where there is an absence of metallic ions. Timber BC is more carbon rich than Juncus BC. This may be due to the lower pyrolysis temperature experienced by the Juncus BC.

4.2.2. Biochar-added concrete

EDX analysis was undertaken on spectrums of the biochar concrete composites that displayed high interaction between the hydration products of the cement matrix and the hydrophilic functional groups found on the biochar's surface (Fig.10 (c), Fig.10(d)). The results were as expected with higher quantities of Oxygen, Aluminium, Silicon, and Calcium found in the cement matrix compared to the BC particles which were compromised of mainly carbon and oxygen.

4.3. Macro Imaging

Fig. 11 shows optical micrographs of the control and 4% Juncus BC samples. The Juncus sample is darker in colour with an even dispersion of the BC particles throughout. An increased number of air voids can also be seen in the Juncus sample, this may be due to the decreased workability as discussed in Section 4.4.

4.4. Concrete Workability

The workability of the concrete reduced by approximately 22% with every 2% addition of BC (Fig.12). This is due to the porous nature of the BC causing a localised reduction in w/c ratio. The Juncus BC appeared to have a greater effect on the w/c this is again due to the larger number of pores leading to a further reduction of free water available in the mix (Akinyemi and Adesina 2020).

Table 2: Elemental composition of biochar and hydration products within the cement matrix

Sample	Spectrum	C	O	Mg	Al	Si	K	Ca	Total
4% Juncus BC Concrete	**Spectrum 1 (Juncus BC)**	68.94	14.54	1.33	6.62	1.07	2.79	4.71	100.00
	Spectrum 2 (Hydration Products)	26.72	39.37	1.24	12.18	9.74	1.43	9.31	100.00
4% Timber BC Concrete	**Spectrum 3 (Timber BC)**	84.48	12.08	0.13	0.20	0.69	0.41	1.44	100.00
	Spectrum 4 (Hydration Products)	23.67	49.85	0.73	2.60	6.90	0.61	15.43	100.00

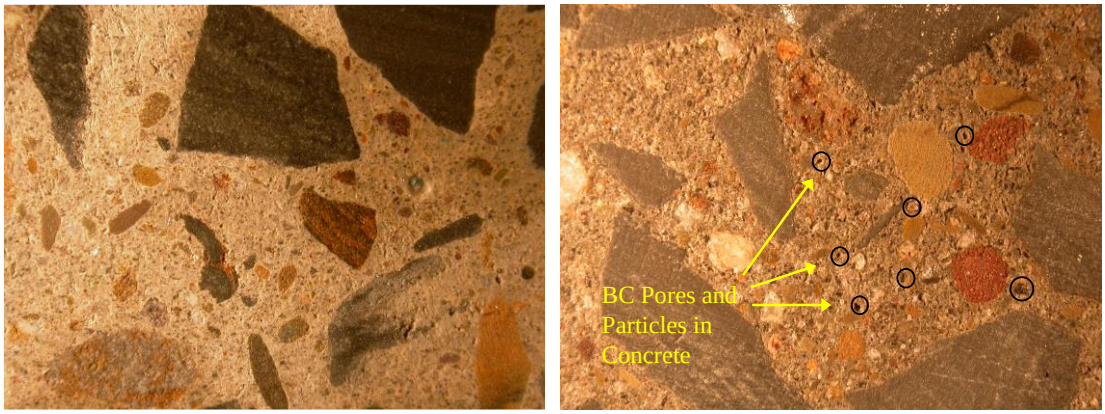

Figure 11: Macro images of (a) control mix (b) 4% Juncus BC mix concrete samples

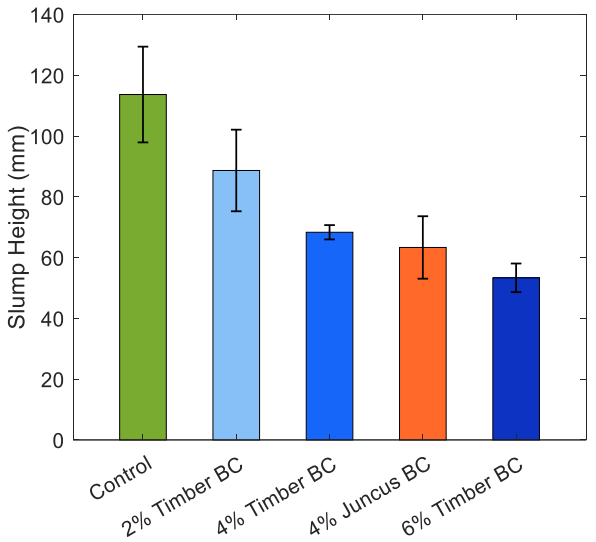

Figure 12: Average slump and corresponding standard deviation

4.5. Compressive Strength

The compressive strength results indicate that biochar has a negative effect on the compressibility of the concrete. With a 2% addition, there is an 18% reduction in strength after 30 days. 4% and 6% Timber BC dosages were found to have similar effects on the concrete's compressive strength with reductions of 11% and 10% respectively. The Juncus BC appears to perform slightly better than the Timber BC with a 10% reduction in compressive strength after 30 days (Fig.13).

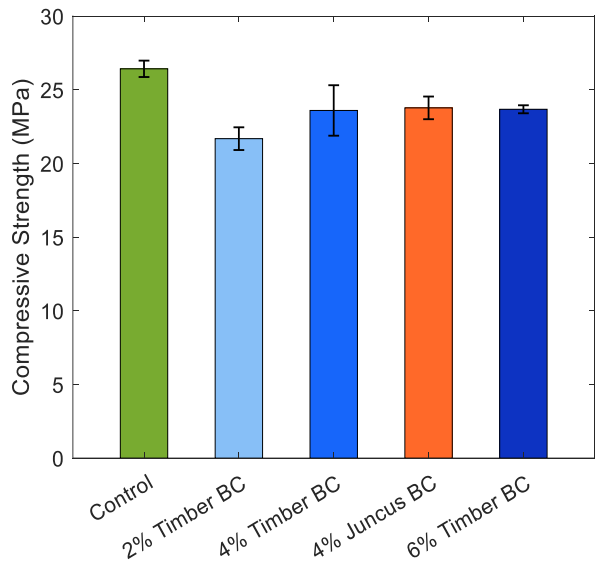

Figure 13: Average compressive strength and corresponding standard deviation

Fig. 14 shows the increase in compressive strength with time. The difference in strength between the 2% Timber BC mix and control is greater at 14 days than at 30 days. This suggests that BC-added concrete cures slower than traditional concrete. This is in line with findings from Sirico *et al.* (2021) who found that the improvement in compressive strength from 28-365 days was equal to 12% in BC-added concrete compared to just a 2% increase in conventional concrete.

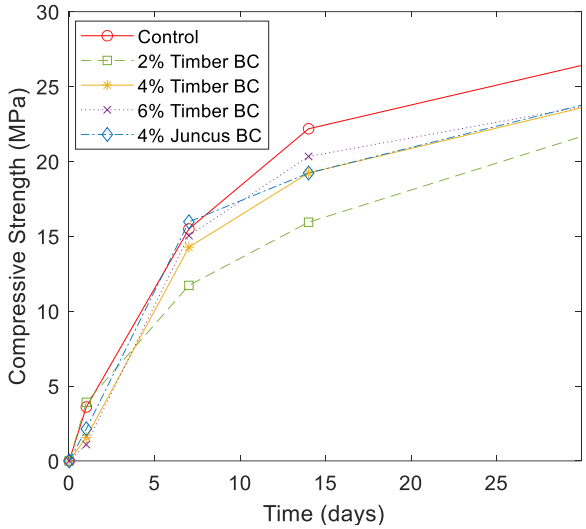

Figure 14: Average compressive strength vs time

Akhtar and Sarmah (2018) also observed an overall reduction in compressive strength with the addition of biochar. They suggest that substituting cement with biochar reduces the production of hydration products such as Calcium Silicate Hydrate (C-S-H Gel).

The increase in compressive strength with higher BC dosage can be attributed to the localised w/c ratio reduction caused by the hydrophilic nature of the BC. This happens during the mixing process when a portion of the water added to the mix gets absorbed by the BC. This temporarily reduces the amount of free water available and positively influences compressive strength as it reduces the proportion of capillary pores formed by the evaporation of water (Souradeep Gupta *et al.* 2018a).

4.6. Split Tensile and Flexural Strength

The addition of biochar has shown to have a mixed effect on the tensile behaviour of concrete, depending on BC type and replacement level. Tensile splitting and flexural strength results follow a similar pattern and will be discussed together.

Timber BC has an overall negative effect on the concrete's tensile strength. 2% addition of Timber BC gives a 14.23% and 6.47% decrease in split tensile (Fig.15) and flexural strength (Fig.16) respectively. This reduction in tensile strength is attributed to the inhomogeneity caused in the tensile plane due to the inclusion of the porous biochar particles (Gupta *et al.* 2020).

Like the compressive strength results, the loss in tensile splitting reduces at 4% cement replacement to 5.88%. This is also believed to be due to the reduced localised w/c ratio as excess free water is temporarily absorbed by BC pores. In contrast, the flexural strength loss at 4% BC increases to 12% compared to the control. 6% BC addition shows further reductions of 17.98% for split tensile and 12.9% for flexural strength. Here the hydrophilic benefits of BC are overcome by the increased pores and air voids in the tensile

plane which form local weak zones in the concrete (Gupta *et al.* 2020).

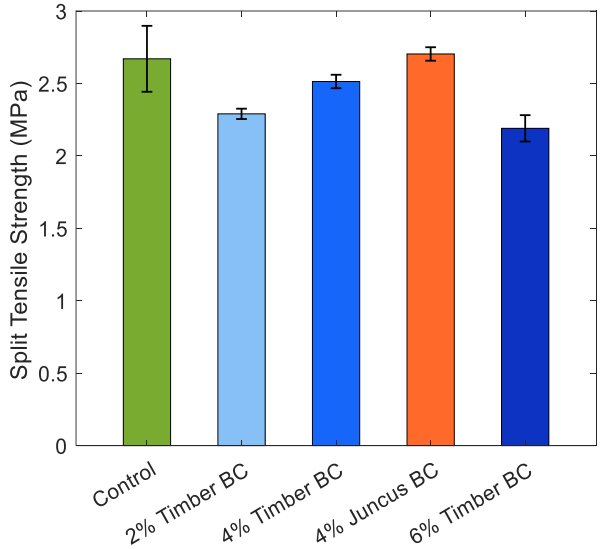

Figure 15: Average split tensile strength and corresponding standard deviation

4% Juncus BC addition showed the most positive results. An average increase of 9% in flexural and 1.25% in split tensile strength was recorded. This significant increase in tensile strength with the addition of Juncus BC in comparison to the Timber source is believed to be due to the following reasons.

- The additional metallic ions found in the Juncus BC during the EDX analysis led to additional ionically bonded crystals forming within the cement matrix improving the bond strength.
- The additional ridges and pores on the surface of the Juncus BC particles improve interaction with the cement matrix as well as increasing the amount of internal curing and formation of hydration products that take place.
- The increased volume (due to its lower density) of Juncus BC available for bonding and interacting with the cement matrix.

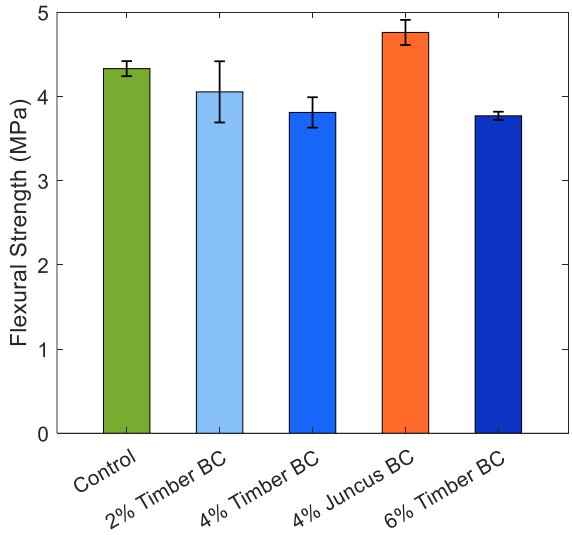

Figure 16: Average flexural strength and corresponding standard deviation

The bonds that form between the Juncus BC and cement particles prevent early cracks from occurring in the specimen by diverting the fracture (Aneja *et al.* 2022).

4.7. Chloride Migration

Biochar appears to negatively affect the concrete's permeability (Fig.17). The introduction of 2% Timber BC increases the chloride migration coefficient (CMC) by 45% when compared to the control. However, there are no further increases in permeability with higher dosages of BC with the 4% and 6% Timber BC mixes showing increases of 44% and 45% respectively.

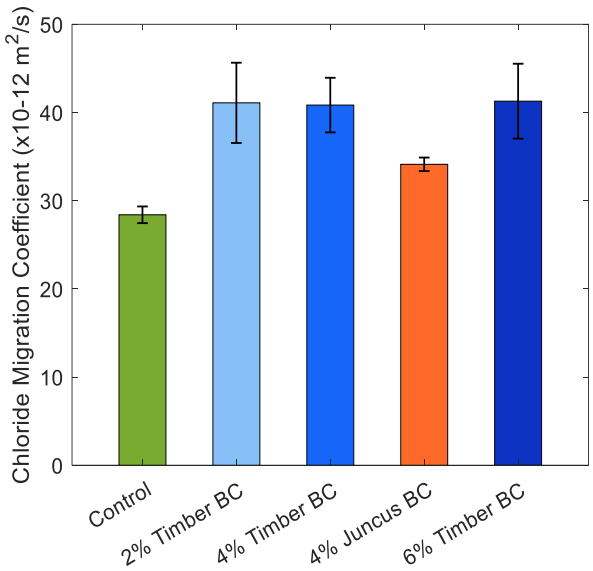

Figure 17: Average chloride migration coefficient and corresponding standard deviation

The addition of Juncus BC seems to have less of an impact on the permeability of the concrete. The CMC increased by 20% when compared to the control, 20% lower than that measured in the 4% Timber BC mix. The increase in permeability may be due to the dilution effect caused by the replacement of cement. The pores introduced into the concrete by the biochar may also be responsible as they allow chloride ions to move through the material resulting in a greater penetration depth. (Yang and Wang 2021)

It is possible that the improved performance of the Juncus BC, when compared to the Timber BC, is due to the

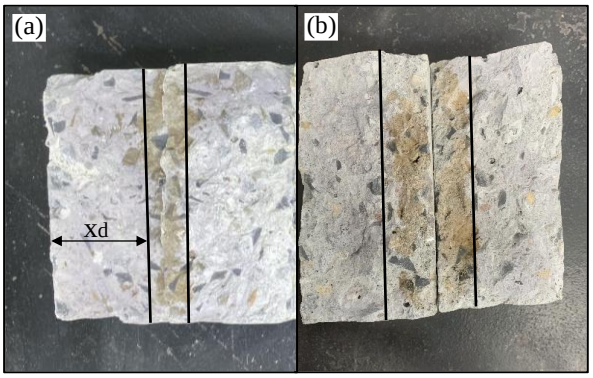

Figure 18: Depth of penetration xd in (a) 2% Timber BC mix and (b) 4% Juncus BC mix

presence of metallic ions in the biochar. These metallic ions form chemical bonds with the chloride ions, reducing their mobility and preventing them from penetrating the concrete. However, further research is required to confirm this hypothesis.

5. Limitations

The following were identified as possible limitations of this study.

- CEM II/A was used in this study, however, CEM I is more commonly used as the binder in studies throughout the literature. CEM I contains approximately 91% OPC (CMI 2022a) while CEM II/A contains approximately 83% OPC (CMI 2022b). Further addition of SCMs to the CEM II/A has led to an additional reduction of OPC in the cement. Therefore, the use of CEM II/A may be responsible for the overall reduction in strength in the biochar mixes when compared to the control.
- Standard practice dictates that concrete samples should be cured at 20℃. However, the samples in this study were cured at 11℃-12℃.
- It was not possible to carry out a statistical analysis to test the significance of the results due to the small sample size.

6. Conclusions

Two sources of locally produced biochar were assessed as SCMs in a structural concrete mix widely used across the Irish construction industry. Timber BC was chosen as it showed positive results across the literature and its cement replacement levels were varied from 2% - 6% as recommended. Juncus was identified as an alternative indigenous biomass source without a current waste stream. Juncus BC was investigated in concrete at 4% cement replacement. The results in this paper are based on a limited number of samples, however, good repeatability was observed across all tests carried out. The following conclusions can be drawn.

- An A1- A3 LCA found that the GWP of a blended cement can be reduced by 17% at 4% BC replacement. A carbon-neutral binder was found to be achievable at 20% BC substitution.
- Biochar reduces the workability of the concrete significantly. In the mix analysed, the workability reduced by approximately 22% with every 2% addition on BC.
- For the types and replacement levels of BC examined as part of this study, an overall reduction in compressive strength ranging from 10% -18% was observed in the BC-added concrete compared to the control.
- A reduction in tensile strength was observed generally, apart from the Juncus BC addition where increases of 9% and 1.25% were observed in flexural and split tensile strengths respectively.
- A consistent increase in permeability of the BC-added concrete was found across all ranges. Timber BC showed an increase in chloride migration of 45%, compared to 20% in Juncus BC.

7. Recommendations for Further Research

The following next steps are recommended to progress the research conducted in this paper on the use of biochar as an SCM in concrete.

- Further work on a greater number of samples is recommended to verify the findings of this paper.
- It is recommended to investigate the use of CEM I as the primary binder in a future study to validate results shown in the literature, which conflict with many of this study's findings. The use of CEM II A may be responsible for this discrepancy.
- The use of a lower w/c ratio and plasticizer is recommended as the effect of plasticiser on BC is relatively unknown and possible strength gains may be achievable.
- As the Juncus BC displayed the most promising results, further research on the effect of varying the conditions of pyrolysis and cement replacement level from 0% to 6% is recommended to optimise its performance as an SCM. The TGA analysis revealed that the Juncus BC was not fully pyrolyzed. Increasing the pyrolysis temperature would increase the carbon content and modify the internal pore structure.
- Pozzolanic activity is an important property to consider when investigating the use of SCMs. Pozzolans react with by-products of hydration to form additional C-S-H gel, leading to further strength increases in concrete. At present, no single index has been established to characterise the pozzolanic reactivity of biochar (Morales *et al.* 2021). The Chappelle Test is one method of quantifying such an index.
- Assessing the effects of dry curing the samples is recommended as there is strong evidence in the literature of improved performance of biochar-added concrete vs a control when dry cured, due to the magnified effects of internal curing that takes place as well as simulating actual concrete curing conditions. (Sirico *et al.* 2021)
- Determining the long-term performance of biochar concrete composites is also recommended. Research should focus on investigating possible long-term strength gains due to additional internal curing taking place, in addition to the long-term durability concerns highlighted in this study.

8. Acknowledgements

The authors would like to acknowledge Green Belt Ltd. and Biomass to Biochar Ltd. for providing biochar for this study. They would also like to express their gratitude to Banagher Precast Ltd. and the technical staff at the University of Limerick for their assistance throughout.

The authors would also like to thank Mr Terence Ryan and Dr Anne Beaucamp Mc Loughlin for their support and guidance throughout the preparation of this dissertation.

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Appendix A

Literature Review

Below is a summary of findings from selected studies.

Table A1 - Summary of mix designs from the literture

Reference	Cement (kg/m^3)	Fine Aggregate (kg/m^3)	Coarse Aggregate (kg/m^3)	W/C	Plasticizer (wt.%)	28 Day Strength (MPa)	Slump (mm)
(Aneja *et al.* 2022)	383.2	707.6	1066.75	0.5	-	33	90
(Barissov 2021)	374.95	967.63	673.96	0.41	-	32.75	120
(Sirico *et al.* 2021)	426.67	1173.33	586.67	0.5	0.95	39.5	200
(Zanotto *et al.* 2022)	408	1126	562	0.5	0.98	39.58	160-210
(Gupta *et al.* 2020)	390	890	890	0.4	0.3	59.51	95

Table A2 - Summary of tests completed

Reference	Slump/ Workability	Fresh Density/ Air Content	Compressive Strength	Flexural Strength	Tensile Splitting	Water Penetration	Chloride Migration
(Aneja *et al.* 2022)	x		x	x		x	
(Barissov 2021)	x		x		x		x
(Gupta *et al.* 2020)	x		x	x	x	x	
(Sirico *et al.* 2021)	x	x	x	x	x		
(Zanotto *et al.* 2022)	x		x				x

Table A3 - Descriptions of biochars and findings

Reference	Biomass Source	Pyrolysis Conditions	Findings
(S. Gupta *et al.* 2018)	Food Waste	Pyrolysis at 500 °C for 45 –60 min	A dosage of 1 – 2% biochar as a SCM shows an increase in air content, a reduction in flowability, and an improvement in mechanical strength across the board.
(Akhtar and Sarmah 2018)	Papermill sludge Rice husk Poultry litter	Slow pyrolysis at 450 °C	Dosage of 0.2% pulp, sludge, and rice husk biochar enhanced the mechanical strength. The poultry litter improved water absorption.
(Qin *et al.* 2021)	Eucalyptus plywood	Slow pyrolysis at 500 °C for 2 h at 10 °C/min	Increased compressive and splitting tensile strength at dosages below 6.5%. Maintains permeability properties of pervious concrete.
(Gupta *et al.* 2020)	Wood Waste	Slow pyrolysis at 500 °C for 1 h at 10 °C/min	Dosages of 0.5% and 2% of biochar improved compressive strength by 16% and 9% respectively. A dosage of 2% resulted in a decrease in permeability of 40%.
(Sirico *et al.* 2021)	Wood Waste	Gasification Between 900 ◦C and 1100 ◦C	5% optimal dosage, shows an increase in flexural and compressive strength.

Table A4 - Barriers to biochar in concrete

Reference	Barrier	Explanation
(Kwapinski *et al.* 2010)	Availability of quality feedstock,	As mentioned previously not all biochar is created equally. Based on the literature, mixed wood waste produces the highest-performing biochar. However, this particular feedstock already has a well-established waste stream in the production of engineered timer, paper and wood pellets. It is also important to note that other industries such as agriculture are already adjusted to the application of biochar in soil and thus increasing the demand for an already limited commodity.
(Winters *et al.* 2022)	Material availability	Most biochar produced at the moment is tailored for soil applications.
	Cost	For biochar concrete to work it first has to make financial sense. The cost of biochar is highly variable depending on the location and feedstock, which makes it difficult to put a cost on its production.
	Revisiting design codes	The current design codes do not incorporate a method for the design of biochar in concrete, these would need to be revised for biochar concrete to make a real impact in the construction industry.
	Uncertainties cause resistance to change	*Legal Issues* – Which stakeholder takes ownership of risk? *Lack of incentive* – Legislation must be brought forward to incentivise design teams to use more sustainable building materials. *Unproven supply chain* – Large risk associated with the procurement of a product that is new to the market.

Appendix B
Below is the output from the aggregate grading, and absorption tests.

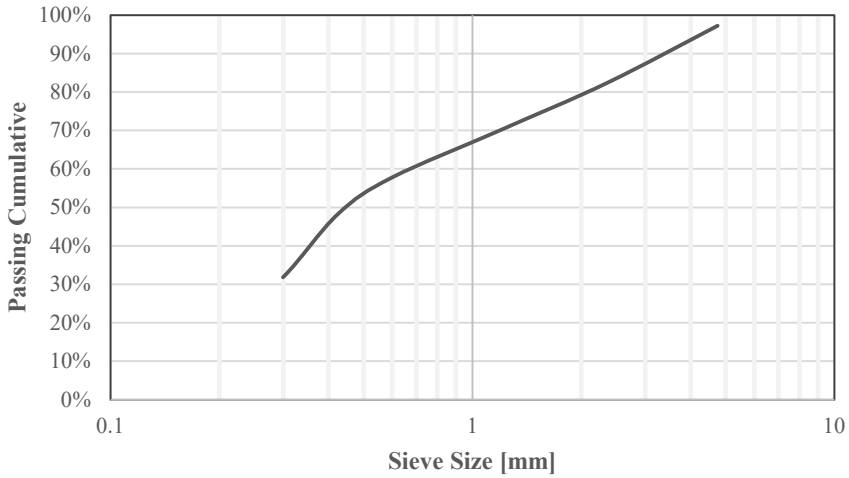

Figure B1: Particle Grading curve

Figure B2: Mechanical sieve shaker and sieves used in particle distribution analysis

Table B1 – Biochar and sand absorption calculations and results

	Test 1		Test 2		Test 3		
Sand	Beaker	188	Beaker	188	Beaker	188	Average
	Dry Sand	183	Dry Sand	182	Dry Sand	185	
	Wet Sand	189	Wet Sand	188	Wet Sand	191	
	Absorption	3.28%	Absorption	3.30%	Absorption	3.24%	3.27%
Biochar	Beaker	134	Beaker	134	Beaker	134	Average
	Dry biochar	53	Dry biochar	59	Dry biochar	67	
	Wet Biochar	104	Wet Biochar	116	Wet Biochar	132	
	Absorption	96.23%	Absorption	96.61%	Absorption	97.01%	96.62%

Figure B2: Paper towel method (a) biochar and sand soaking in water for 24h (b) Sand drying to SSD (c) Biochar drying to SSD

Appendix C

Results and apparatuses used in SEM, EDX and TGA analysis

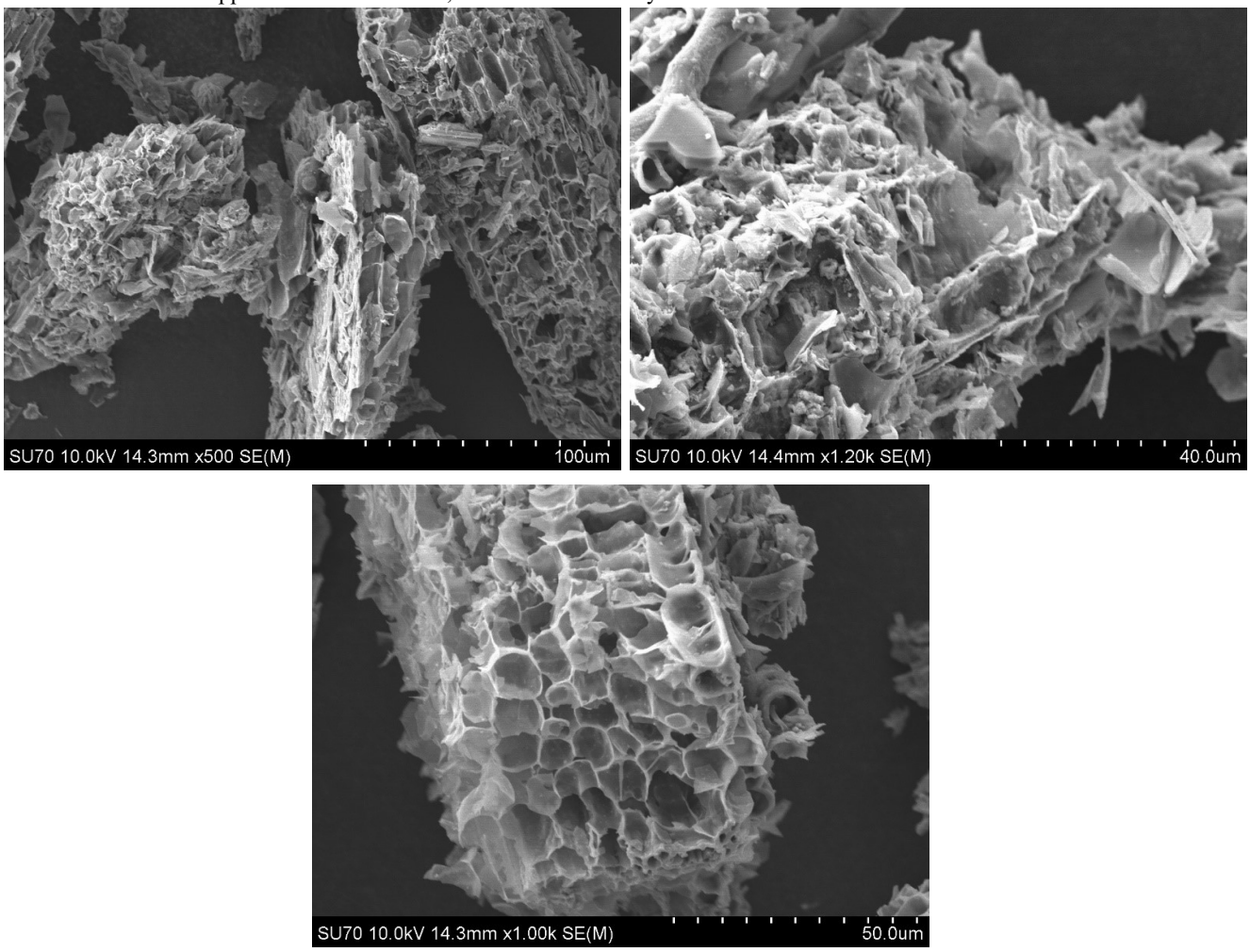

Figure C1: SEM images of Juncus Biochar

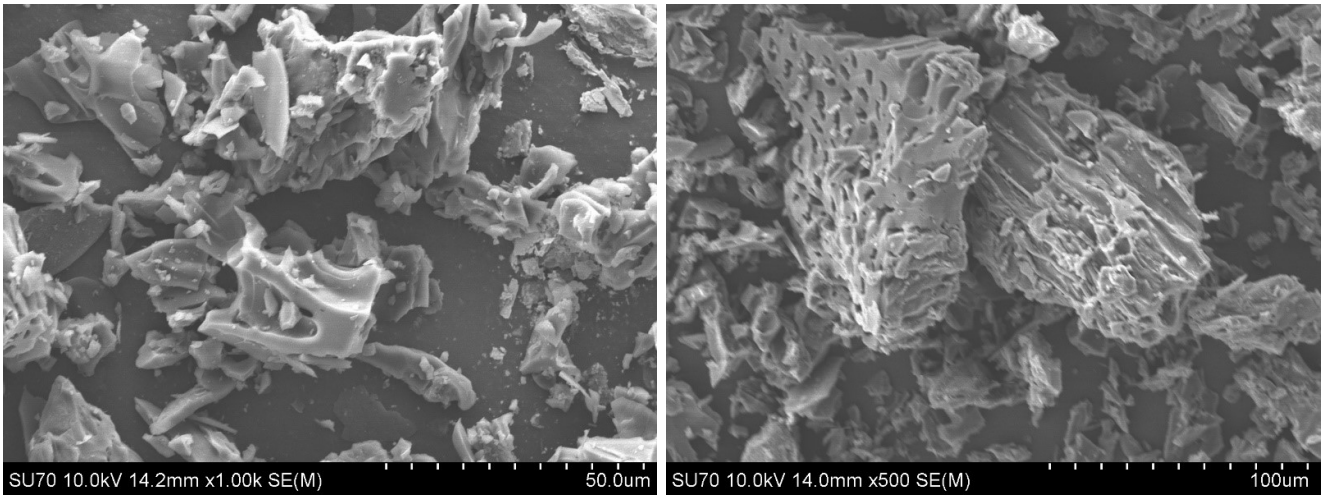

Figure C2: SEM Images of Timber Biochar

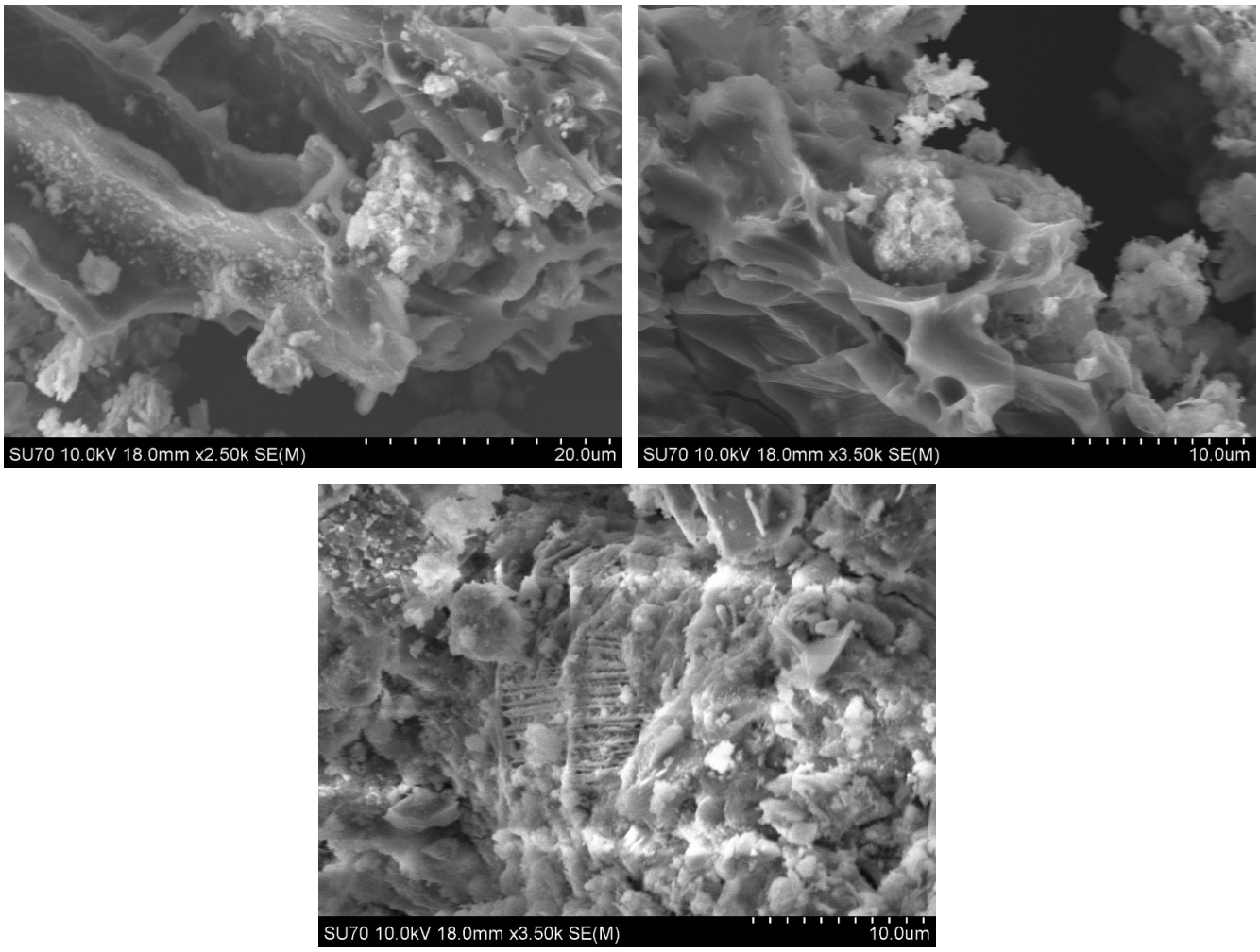

Figure C3: SEM Images of Juncus Biochar added composites

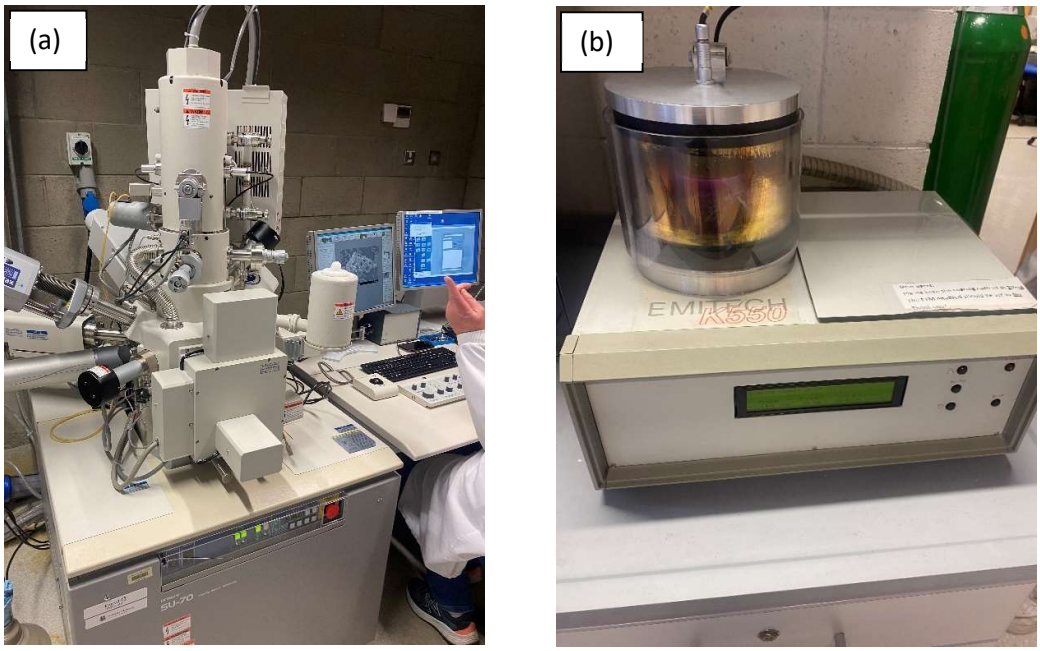

Figure C4: (a) Hitachi SU-70 SEM machine used (b) Emitek H550 sputter coater used to prepare samples for the SEM

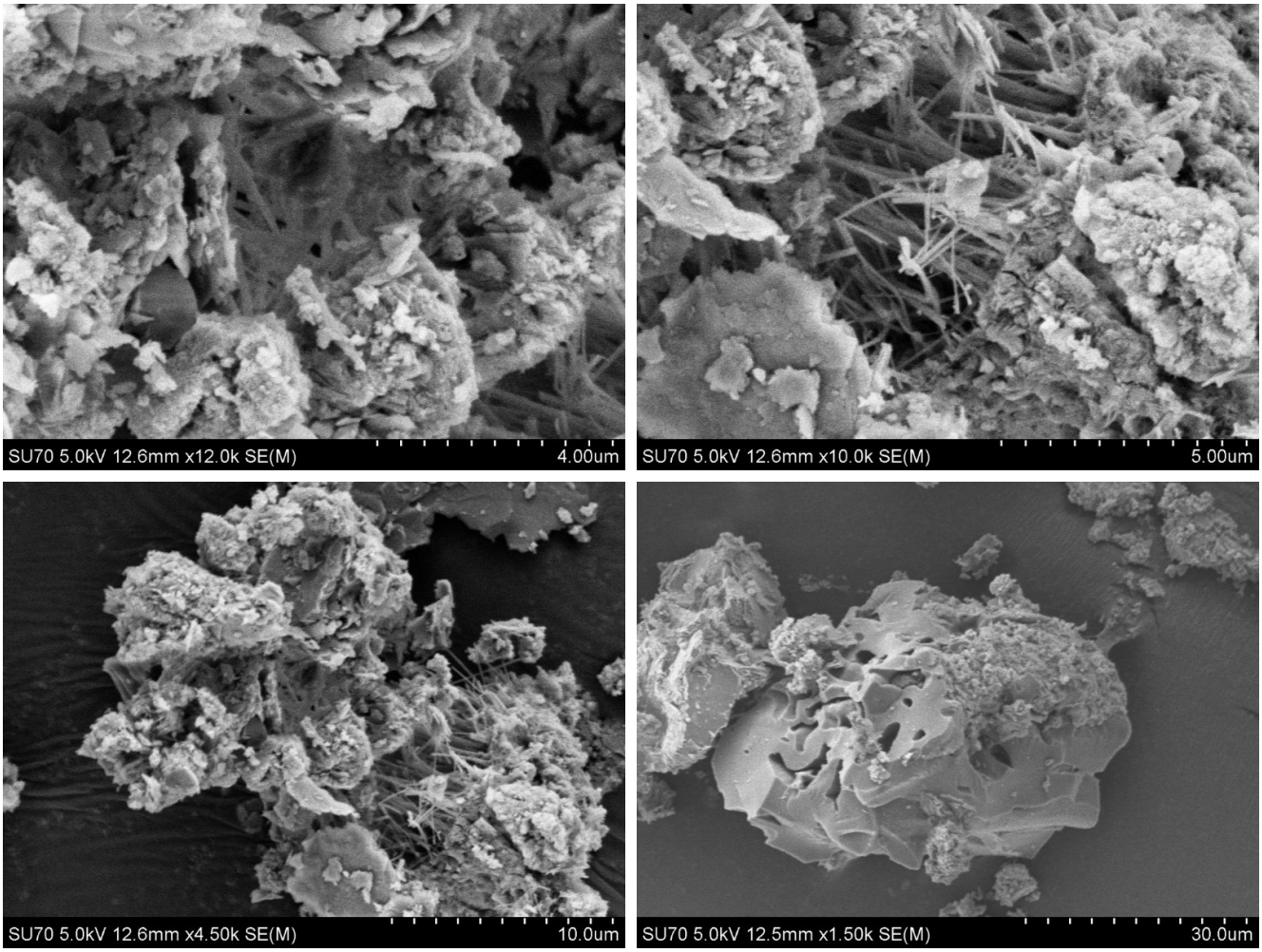

Figure C5: SEM Images of Timber Biochar concrete composites

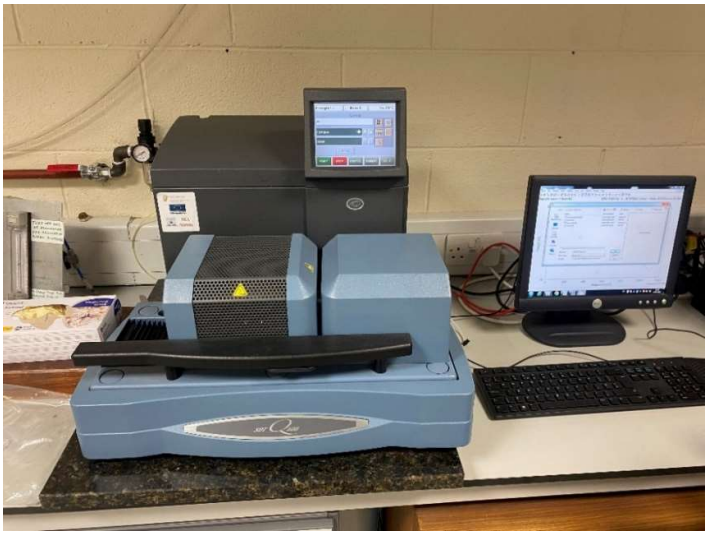

Figure C6: SDT Q600 TGA machine

Appendix D

Concrete Mixing laboratory images

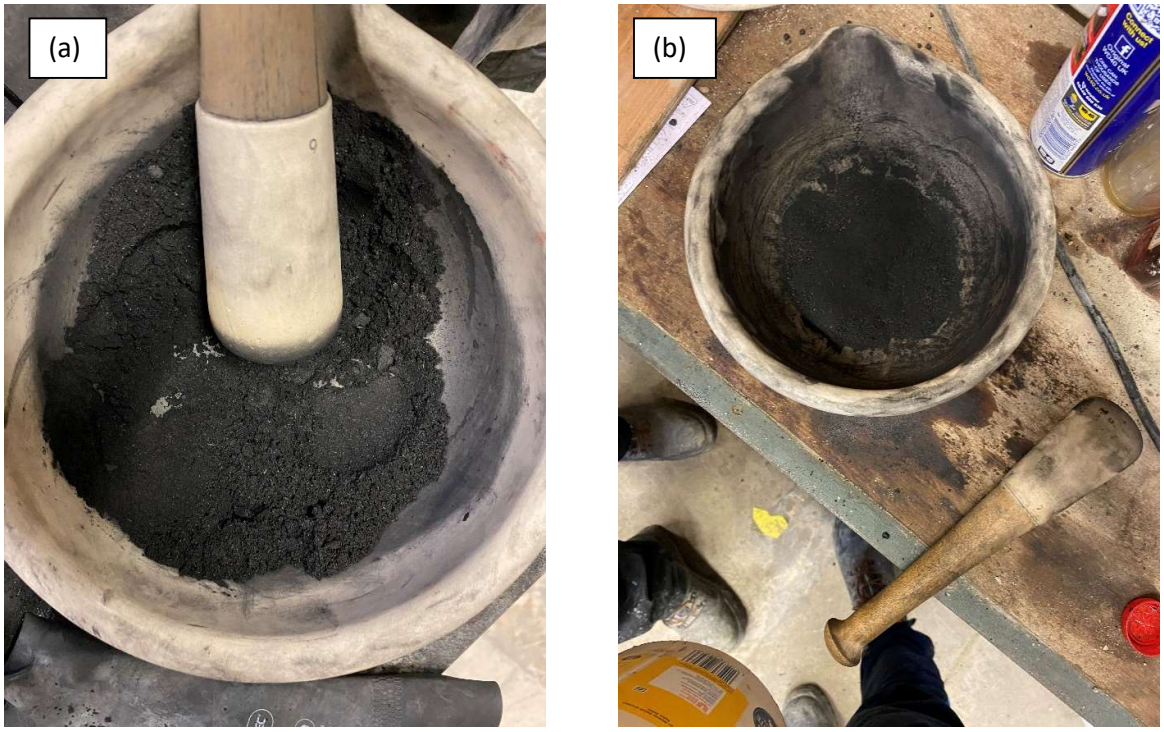

Figure D1: Ground (a) Juncus and (b) Timber biochar samples

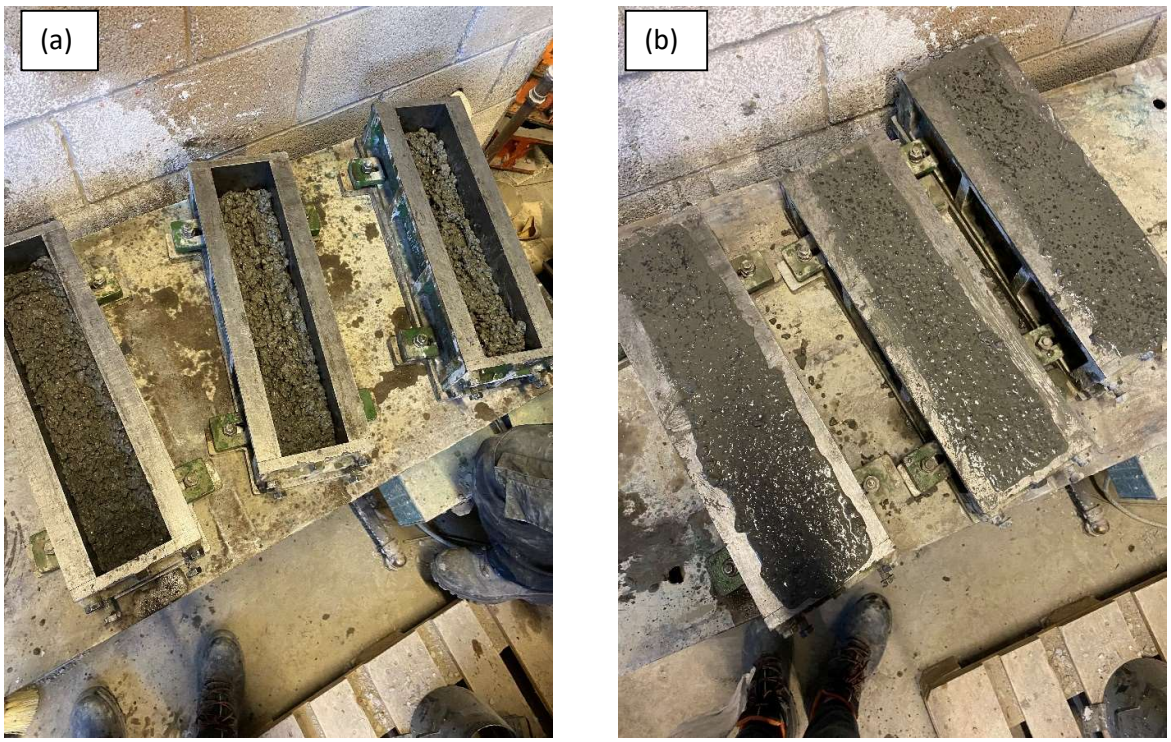

Figure D2: (a) Beam moulds being filled with 4% Juncus BC Concrete. (b) Moulds filled with concrete after vibrating.

Figure D3: (a) Full set of 6% BC concrete moulds after sampling (b) Full set of 6% BC specimens after demoulding.

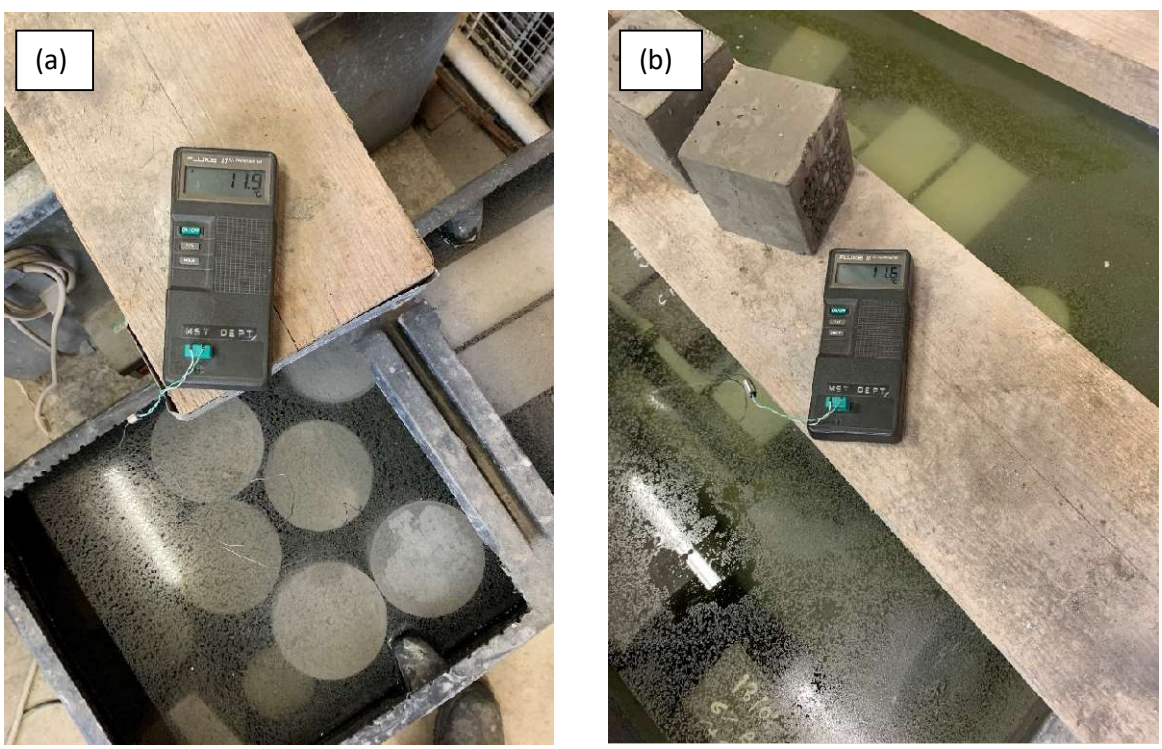

Figure D4: Concrete curing (a) 11.9°C (b) Full set of 11.6% BC°C specimens after demoulding.

Appendix E

Additional Concrete Testing Photos

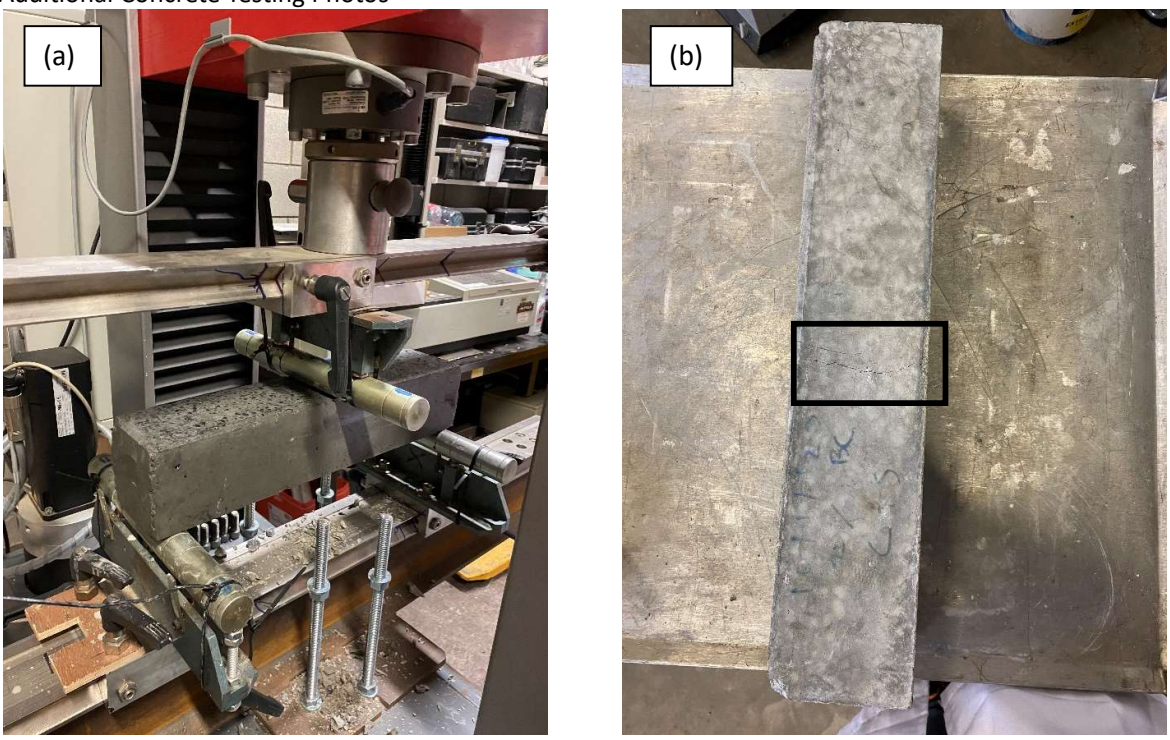

Figure D4: (a) Zwick Roell Z100 3-point beam flexural strength test apparatus used (b) Fractured beam specimen after test

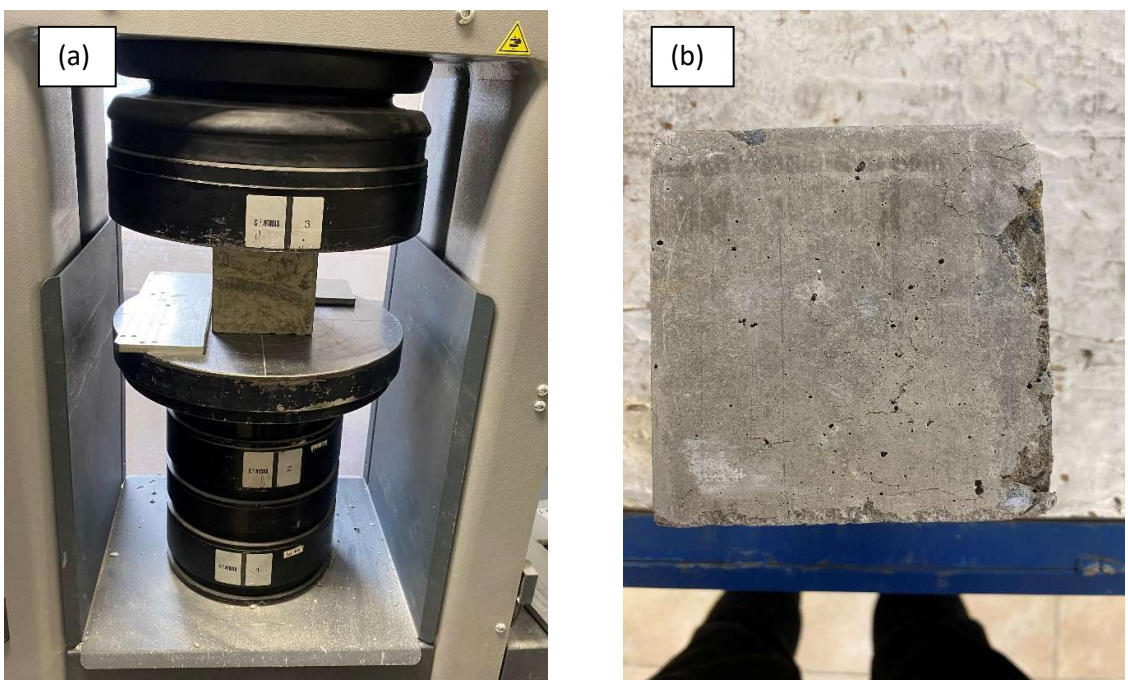

Figure D4: (a) A Controls AUTOMAX PRO compression testing machine used (b) Fractured cube specimen after test