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Increasing the Pozzolanic Reactivity of Recovered CDW Cement Stone by Mechanical Activation †

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Abstract: The study focuses on enhancing the reactivity of the fine size fraction of construction and demolition waste (CDW) by mechanical activation in a stirred media mill. Systematic measurements were carried out to monitor the change in cement stone reactivity. The fine size fraction of CDW (<200 µm) was milled in a stirred media mill for 1, 3, 5, and 10 min. The dispersion characteristics (particle size distribution, specific surface area (SSA)) of the mechanically activated CDW powder were determined using a laser particle size analyzer. Changes in the structure of the mechanically activated CDW powder particles were determined by Fourier transform infrared spectroscopy (FTIR) measurements. The effect of the mechanical activation on the pozzolanic reactivity of CDW powder was measured by lime sorption test and compressive strength measurements. The results clearly show that Portland cement can be replaced with mechanically activated CDW powder; however, increasing its amount decreases the strength. Furthermore, the grinding fineness significantly influenced the pozzolanic reactivity of the mechanically activated CDW powder, and thus the strength of the specimens. The CDW powder milled for 10 min had 51% more lime uptake than the initial CDW sample, and the specimen strength at the age of 7 days was 23% higher using ground CDW powder than using initial CDW at a 20% cement replacement ratio.

Keywords: construction and demolition waste; cement substitution; mechanical activation; stirred media mill; reactivity

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1. Introduction

The construction sector can be considered the largest resource user and waste producer in the world. About 3 billion tons of construction and demolition waste are generated annually in the world [1], while in the European Union (EU), 37.1% of total waste (approximately 800 million tons) was generated in the construction industry in 2020 [2], and this trend is constantly increasing. For this reason, the reuse and recycling of construction and demolition waste (CDW) is a key point not only for increasing resource efficiency, but also for reducing the use of large amounts of primary materials, energy consumption, and waste generation.

Despite the fact that many ways of utilizing CDW are known [3–8], there are many problems that prevent their large-scale use as a substitute for primary raw materials [9]. One of the largest components of CDW waste is concrete waste (other than excavated soil), approximately 70–80% of whose volume consists of fine and coarse aggregates, causing continuous depletion of natural resources [7]. Fine and coarse aggregates can be extracted from concrete waste using different processes (crushing, magnetic separation and sieving process), which can then be used as recycled concrete aggregates in different structures [7,10–14]. At the same time, the fine powder fraction (<150 µm) mainly from the cement stone part of concrete can also be used in recycled concrete or geopolymer [15–17].

However, in order to increase the reactivity of the CDW particles, milling may also be necessary, which can be performed in various mills (planetary mill [18], ball mill [17–19], vibratory mill [18], and stirred media mill [20]). The advantages and disadvantages of CDW powder fineness were summarized in a study by Tang et al. [21].

The aim of the research work is to increase the pozzolanic reactivity of the CDW powder fraction using a high energy-density mill (stirred media mill) and to study the relationships between its reactivity, structural characteristics, and the properties of the produced cement stone.

2. Materials and Methods

The base materials for the experiments were CDW with particle size <63 mm from Mento Ltd. (Bodrogkeresztúr, Hungary) and Portland cement (CEM II/A-S 42.5 R). As a first step, the CDW sample was crushed with a jaw crusher with a gap size of 20 mm. The fine fraction (<200 μm) was removed with sieving, and the 20-8 mm fraction was used in a Deval apparatus for autogenous milling for 60 min. This way, due to the abrasion of the CDW, a higher amount of cement stone could be obtained. The fine products of the previous steps were used as feed for the mechanical activation. The mineralogical composition of CDW was determined with a Bruker D8 Advance X-ray powder diffractometer (XRD) (Cu-Kα radiation, 40 kV, 40 mA) in parallel beam geometry (Göbel-mirror), as can be seen in Table 1. The high amount of quartz and carbonates come from the sand and hydrated cement. Mechanical activation was carried out in a stirred media mill with 530 cm^3 volume, using ZS type, Ø 1–1.2 mm ceramic beads and 5 m/s circumferential velocity. Both the material and grinding media filling ratio was 0.7. The applied milling times were 1, 3, 5 and 10 min. The 0 min milling refers to the pre-processed sample that was not mechanically activated in the stirred media mill.

Table 1. Mineralogical composition of CDW (wt. %).

Phase Name	Raw CDW
Quartz	54.4
Muscovite 2M1	4.0
Calcite	9.7
Vaterite	6.3
Albite	6.1
Orthoclase	3.0
Chlorite IIb	0.6
Ettringite	0.3
Portlandite	0.2
Actinolite	0.4
Amorphous	15.0

The particle size distributions and SSA of the mechanically activated samples were analyzed with a HORIBA LA950-V2 laser particle size analyzer. The pozzolanic reactivity was assessed based on the lime sorption test carried out according to the MSZ 4706-2:1998 standard. For the structural analysis, a Jasco FTIR-4200 analyzer equipped with a diamond ATR was used, with 4 cm^{-1} spectral resolution. The spectra were recorded between 400–4000 cm^{-1} and baseline-corrected.

To further test the reactivity of the mechanically activated CDW cement, samples were prepared to examine the possibility of Portland cement replacement using the mechanically activated CDW cement stones. In the specimens, the Portland cement was replaced with 0%, 10%, 20% and 30% mechanically activated cement stone, and a 0.33 water-cement ratio was applied to all mixtures. The uniaxial compressive strength of the 20 × 20 mm cubical specimens was measured with an SZF-1 type hydraulic press.

3. Results and Discussion

3.1. Mechanical Activation of CDW

3.1.1. Particle Size, SSA

The effect of mechanical activation on the particle size and geometric SSA of CDW is shown in Table 2. Milling resulted in a decrease in the particle size of CDW, while the specific surface area increased. After 10 min of milling, the specific surface area of CDW increased by 33%, but at the same time, no significant decrease in particle size was observed. This can presumably be attributed to the high quartz content (Figure 1), which hindered the efficiency of the milling.

Table 2. The characteristic particle size and SSA values of the ground cement stones.

Milling Time, min	x_{10}, μm	x_{50}, μm	x_{80}, μm	SSA, cm^2/cm^3
0	2.61	10.74	22.76	9500
1	2.31	8.27	23.65	10,961
3	2.25	8.23	24.97	11,096
5	2.24	7.38	20.1	11,715
10	2.02	6.99	19.62	12,601

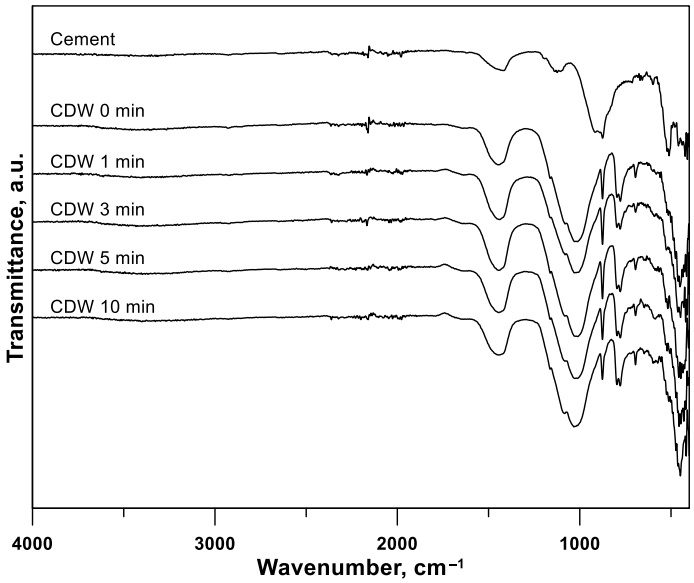

Figure 1. The FTIR spectra of the cement and ground CDW samples.

3.1.2. FTIR

The FTIR spectra of the cement and ground CDW can be seen in Figure 1. In the case of the raw cement spectrum, the transmittance bands at 1421, 874 and 713 cm^{-1} can be assigned to v3, v2, and v4 stretching modes of CO_3^{2-}, respectively. The weak band at 1124 cm^{-1} is generally the indication of Si–O–Si stretching vibrations and the band at 521 cm^{-1} corresponds to the Si–O deformation vibrations of the siliceous phases [22,23].

For the CDW samples, some new bands could also be observed compared to the cement sample. The broad band between ~3500–2800 cm^{-1} corresponds to the stretching O–H, and the weak band at 1614 cm^{-1} to the O–H bending mode, indicating the presence of a small amount of structural and weakly bound water in the samples. The bands centered between 1420–1450 cm^{-1} and at 875 cm^{-1} are associated with the calcite and other carbonate species. The broad band of 1010–1030 cm^{-1} originates from the asymmetric stretching vibrations of the C–S–H structure that was originally formed in the cementitious matrix. The bands at 795 and 695 cm^{-1} show the presence of quartz [20,24]. Comparing the spectra of the ground CDW samples, no significant changes occurred in the structure with

the different milling times. After 3 min or longer mechanical activation, the intensity of the C–S–H band slightly increased and broadened, indicating the possible amorphization of the material.

3.2. CDW Reactivity

3.2.1. Pozzolanic Reactivity

Lime adsorption is an indicative measure of hydraulic/pozzolanic reactivity. The results of the lime sorption tests are shown in Table 3. Based on the Table 3, it can be stated that the SSA and CaO uptake values followed a similar trend after the mechanical activation: the increased SSA significantly increased the amount of adsorbed CaO. The lime uptake of CDW increased from the initial 126.9 mg CaO/g solid material to 192.1 mg CaO/g solid material. Thus, as a result of a 33% increase in SSA, the CaO uptake increased by over 50%.

Table 3. Lime adsorption of CDWs.

SSA of CDW (cm^2/cm^3)	∑Adsorbed CaO (mg/g)
9500	126.9
10,961	131.4
11,096	126.7
11,715	169.0
12,601	192.1

Figure 2 illustrates the volume change (swelling) during the reaction between CDW and lime. It can be clearly seen that the ground CDWs had a larger volume than the original at the end of the lime sorption test (after 30 days), indicating a greater degree of reactivity.

Figure 2. CDW samples after the lime sorption test (from left to right: 0 min, 1 min, 3 min, 5 min and 10 min).

3.2.2. Compressive Strength

Figure 3 shows the effect of CDW dosage and powder fineness on the compressive strength of the cement-based specimens. Based on the results, it can be concluded that the replacement of cement with CDW reduced the strength of the specimens. This correlates well with the results of other studies [16,20,25,26]. However, it is also seen that the ground CDWs showed better results compared to the unground (0 min) sample, at 20 and 30% cement replacement. Furthermore, specimens prepared using CDW ground for 5 and 10 min gave similar results at 20 and 30% cement replacement, which can be explained by their similar particle size distribution (Table 2). Generally, no clear correlation can be observed between the grinding time and replacement ratio. This can be explained by the heterogeneity and the high quartz content of the recycled cement stone [16,27].

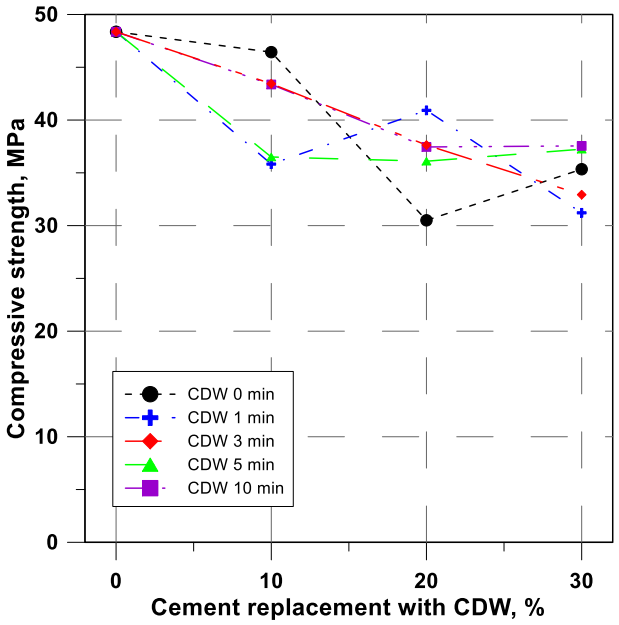

Figure 3. Results of compressive strength measurements.

3.2.3. FTIR

Figure 4 shows the FTIR spectra of the samples in the case of 0 min CDW dosing at different ratios. Based on Figure 4, it can be stated that the intensity of some bands decreased as a result of CDW dosing. The intensity of the bands around 1420 cm^{-1} and 874 cm^{-1}, which can be attributed to calcite and other carbonates, decreased to the greatest extent. In addition, the intensity of the band at 3640 cm^{-1} (which indicates the presence of portlandite ($Ca(OH)_2$) [28]) and the S–O stretching vibration of $[SO_4]^{2-}$ at 1150–1100 cm^{-1} also decreased with the addition of CDW.

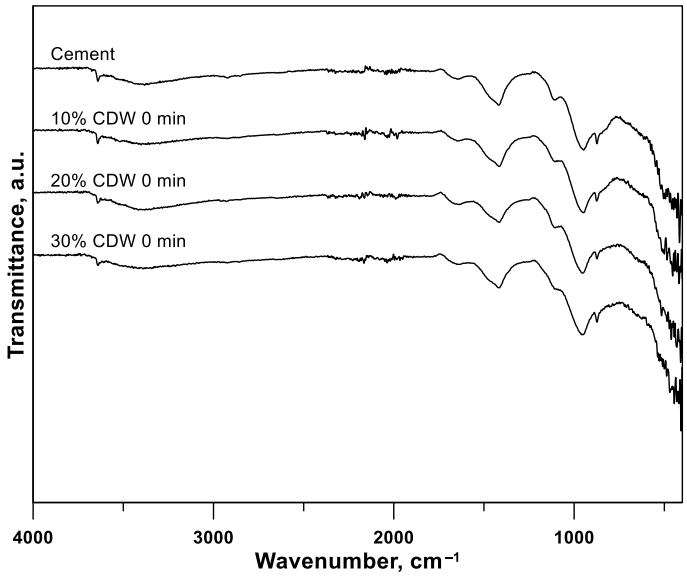

Figure 4. FTIR spectra of samples with different CDW content.

Figure 5 shows the FTIR spectra of the samples containing CDW with different powder fineness, produced with 20% cement replacement. Figure 5 shows that the same bands appeared for each sample. The intensity of the bands at 1420 cm^{-1} and 874 cm^{-1} belonging to calcite and other carbonates increased with the fineness of the CDW. The broad band at around 3400 cm^{-1} is attributed to the symmetric stretching vibration of the H_2O molecule,

while the sharp, narrow band at 3640 cm^{-1} can be assigned to O–H stretching vibration (portlandite) [26,28], which showed a slight decrease by 10 min milling. The band at around 960 cm^{-1} implies the Si–O stretching vibrations, indicating a wide range of C–S–H. The band at 960 cm^{-1} is assigned to Si–O stretching vibrations, which is the result of the C–S–H phase with Ca/Si ≈ 2. When it reaches 1080 cm^{-1}, it indicates the formation of silica gel [29]. Higher intensity C–S–H related bands were observed with longer CDW cement stone milling times, indicating the effectiveness of mechanical activation.

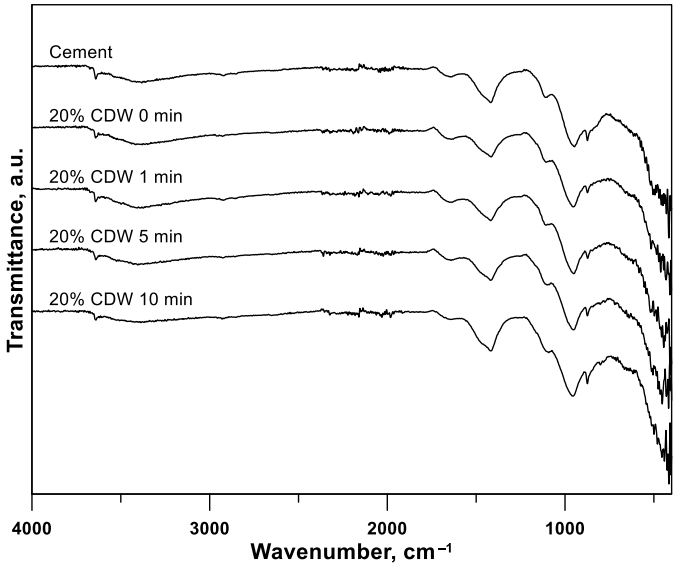

Figure 5. The FT-IR spectra of specimens (cement replacement with 20% CDW).

4. Conclusions

Based on the results, the following conclusions can be drawn:

- The increased specific surface area due to mechanical activation improved the pozzolanic reactivity of CDW dust (50% better CaO uptake after 10 min of milling).
- The use of CDW dust as a cement substitute reduced the strength of cement-based materials.
- The use of mechanically activated CDW led to a lower strength reduction than the unactivated sample.
- As a result, cement can be replaced with mechanically activated CDW in remarkable quantities (20 or 30%); thus it can be used primarily in areas where high structural performance is not required.
- No significant changes occurred in the structure due to MA. However, the increase in intensity of the C–S–H band indicates the possible surface amorphization of the particles.

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