Abstract. The concrete construction and demolition waste (CDW) is usually recycled as coarse aggregate. The fine fraction remains unused – if recovered at all, it is for downcycling in backfilling. Part 1 of this article is dedicated to a technical study on recycling of the waste concrete powder (WCP) as supplement in blended cements. Part 2 presents assessment of the environmental impacts of those binders. XRD and DTA/TG analyses of WCP are performed. WCP from two types of concrete CDW is used. The behaviour of fresh and hardened pastes with replacement of CEM I 52,5 by WCP (15%, 30% and 45%) is investigated. Standard methods of testing with some modifications are used. The influence of WCP content on the setting, consistency, strength and water absorption is evaluated. Four different curing regimes are applied. The results confirm the initial assumption that WCP can be consider as cementitious supplement. The replacement rate up to 15% practically does not change the behaviour of the binder, while 45% seems to be the maximum, because of binders performances degradation. The replacement up to 30% allows to keep satisfactory technical properties, but offers greater economic and environmental advantages.

1. Introduction
The Portland cement concrete is still the most used construction material. Cement production generates more than 30% of CO$_2$ [1] emissions, because of high fossil energy demand and chemical transformations of raw materials to get the clinker. Replacement of clinker by mineral additives is one of the ways to reduce the negative environmental impact of Portland cement, but also a path for cost optimisation. Moreover, some mineral additives, such as ground-granulated blast furnace slag (GGBFS), fly ash, silica fume, burnt shale, natural pozzolana are active, i.e. they modify the properties of cement and lead to increased durability in aggressive environment or exposure to high temperature. Other additives such as limestone filler, quartz mill, etc. are rather inert (at normal hardening conditions) and contribute mainly to the costs optimisation.

Blended cements become increasingly used because of their technical and economic advantages. However, it shall be considered that some mineral supplements coming from industrial waste are going to be in a shortage in Europe because many of the metallurgical factories have been closed and the coal thermal power plants are going to be decommissioned in the next 2-3 decades as per the Grean deal [2]. Natural pozzolana such as volcanic ashes and pumice are not always available at
reasonable distances from cement production plants. For example, in Bulgaria all 3 cement factories are situated in the Northern part, while the deposits of natural pozzolana are concentrated in a small region in the southern part, which means transport distances of more than 400 km. Thus, alternative binders such as geopolymers [3] or alternative supplementary cementitious materials are being explored [4] with increasing interest.

Notwithstanding the foregoing, the use of recycled waste concrete powder (WCP) from construction and demolition waste (CDW) possesses several advantages: a) environmental: it contributes to closing the loop of concrete by increasing the CDW recovery degree, the recycling process of concrete CDW is not sophisticated and is low-energy consuming, the WCP is available at a short distance and CDW generation is relatively constant; b) technical – depending on the original concrete and the recycling technology the old cement stone can have higher or lower hydraulic activity, due to non-hydrated clinker, while the hydrated Portland cement stone might serve as germs of crystallization, i.e. WCP is expected to be an active supplement. The main source of WCP is the fine fraction (crushed sand and powder) which results from the recycling of CDW into crushed stone and aggregate. At present in Bulgaria, this residue, if recovered at all, is downcycled as backfilling material.

There are a couple of studies on the utilization of WCP as a substitution material for cement. The results are not consistent and often contradictory, but all report a significant decrease in compressive strength when WCP is used. To overcome this problem, some researchers suggest a thermal activation at 800°C to be performed in order to compensate the strength reduction [8]. There are even more radical approaches where the WCP replaces the clinker after being treated at 400–1450°C and is further blended with supplementary cementitious materials to produce cement [9-11]. Although technically feasible and chemically justified, such approach does not resolve the major environmental drawbacks of cement production – CO2 emissions and huge energy consumption.

The purpose of the first part of the presented study is to assess the limits in clinker replacement by WCP from technical perspective – presence of hydraulic activity, influence on the structure of the cement stone, impact on strength and sourcing of WCP.

2. Materials and methods

2.1. Materials

Portland cement CEM I 52,5 R having fineness of 3420 cm2/g measured as per Blaine method (BDS EN 196-6 [12]) is used for the preparation of alternative binders with different amount of WCP. Two different sources of concrete CDW to produce WCP are explored:

- “young concrete” waste, at age of 2 months, originating from concrete cubes prepared in the laboratory compressive strength conformity assessment for a high-rise building in Sofia; the binder content in concrete is expected to be relatively high and cement class is most likely to be CEM I 52,5, so a great WCP hydraulic activity is theoretically probable; however, the real activity might be lower, because this is a common practice in Bulgaria to replace the Portland cement by fly ash and to achieve the higher classes of cement by a greater fineness, resulting in reduction of the amount of non-hydrated cement particles;
- old concrete” waste at age of more than 35 years, originating from CDW from large panel buildings; it is expected that the majority of concrete CDW of interest to be recycled (incl. as WCP) in Bulgaria, will be of that type; the used cements were coarser at that time, so the amount on non-hydrated part of cement stone is expected to be big; the amount of carbonates is also expected to be higher due to the carbonation process.

A laboratory recycling of CDW was performed. A preliminary crushing of concrete elements was done to reduce their maximum size to approximately 10 cm. The CDW pieces were oven dried at +105°C to a constant mass. After jaw crushing of concrete waste, the coarse fraction (above 4 mm) was removed and the WCP was obtained by grinding the sand fraction in a ball mill to a fineness of 3000-3500 cm2/g, measured as per the Blaine method. Particles bigger than 0.125 mm were removed.
by sieving. The WCP was stored in airtight PE bags for not longer than 2 days, before being incorporated in alternative binders.

A (DTA/TG) and (XRD) analyses of WCP (from the young concrete) were performed at Bulgarian Academy of Sciences, in the partner Institute of Physical Chemistry. The DTA results show two main endothermic effects related to the chemical dissociation of hydrated products and the release of chemically bound water: the peak at 450-480°C is associated with the calcium hydro-silicates and calcium hydro-aluminates (Figure 1). The second, more pronounced peak (i.e. representative for a bigger amount of the compound) at 700-800°C is related to the dissociation of carbonates resulting mainly from limestone/dolomite aggregate, present in the primary concrete [4].

![Figure 1. DTA and TG curves of WCP from the “young” concrete waste.](image)

The XRD results confirms the findings (Figure 2) on the dominant presence of hydration products and carbonates.

![Figure 2. XRD diffractograms of WCP from the “young” concrete waste.](image)
The amorphous phase, due to the non-hydrated cement and calcium hydro-silicates (looking amorphous because of their nano-size structure) is around 30% of total mass. The crystalline phase consists of calcium magnesium carbonate (ca. 25% of total mass), portlandite (ca.26% of total mass), calcium aluminum silicate hydroxide (ca. 12% of total mass), hillebrandite (ca. 3% of total mass) and silicon dioxide (ca. 4% of total mass). The last two components and the carbonates are due to the presence of aggregate (dolomitic coarse aggregate and river sand) in the primary concrete, which are unavoidable in the WCP.

No chemical admixtures or other mineral supplements are used, in order to avoid some possible side effects on the properties of the blended binders.

2.2. Blended binders composition and mix design

Three types of so-called alternative binders are investigated, designated as WCP15, WCP30, WCP45. They represent dry mixes of Portland cement CEM I 52,5 and three varieties of amounts of WCP blend- 15%, 30% and 45% of the total mass of the dry mix, respectively. Smaller amounts might have insignificant technical effects. Bigger amounts would have a greater environmental effect, but the technical properties are not likely to be satisfactory.

When mixed with water, the same water/binder ratio is applied (0,27), because the standard consistency does not change much (Table 1). At that water/binder ratio the workability of WCP15 and WCP30 was practically identical to that of the control paste, while the workability of WCP45 was slightly lower but convenient for the samples preparation. No sand is added to produce mortars, in order to clearly distinguish the effect of WCP on the binder properties. Also, a control mix (designated as C100) is prepared based only on CEM I 52,5.

| Binder designation | Substitution by WCP, % | Standard consistence, % | Initial setting time, min | Final setting time, min |
|---------------------|-------------------------|-------------------------|--------------------------|------------------------|
| C100                | 0%                      | 27                      | 105                      | 200                    |
| WCP15               | 15%                     | 28                      | 132                      | 205                    |
| WCP30               | 30%                     | 28                      | 135                      | 230                    |
| WCP45               | 45%                     | 29                      | 140                      | 255                    |

2.3. Samples preparation and curing

Samples of size 40x40x160 mm were produced from each binder. For each testing 3 specimens were tested and the average value is reported. The specimens were kept in molds for 24 hours (at 20 ± 3oC and R.H.>95%), wrapped in PE foil.

After demolding of samples, 3 storage regimes were applied for mixes based on C100 and WCP30: in water at 20 ± 3°C, moist curing (wrapped in PE foil, stored at 20 ± 3°C and R.H.>95%), air conditions (20 ± 3°C, R.H. =60±5%). One series of samples was autoclaved at the age of 2 days (100±5°C and 1,3 MPa for 8 hours) and was stored at air conditions until testing. WCP15 and WCP45 samples were stored only in moist conditions.

2.4. Testing methods

Standard methods and modified standard testing methods were applied:
- Standard consistence and setting time as per BDS EN 196-3 [13];
- Compressive and flexural strength as per BDS EN 196-1 [14] on binder pastes (not mortars);
- Water absorption as per BDS EN 12808-5 [15] with a longer time of immersion in water, because of the fine porous structure;
- Water saturation after 5 hours of treatment in boiling water; it is representative for the open porosity, accessible to water in the hardened paste;
Coefficient of saturation is calculated as ratio of water absorption and water saturation and is representative for the volume of empty voids, accessible to water, then the hardened paste is immersed in water in normal conditions.

3. Results and discussion

3.1. Properties of fresh pastes
The properties of fresh mixes do not differ much in terms of workability, assessed by the Vicat plunger penetration 5±1 mm. Both initial and final setting time are influenced by the substitution of cement with WCP – the bigger the amount, the longer the time required for setting, up to 30% in average, measured for WCP45 (Table 1). Such behaviour is similar to other ordinary blended cements of type CEM II.

3.2. Hydro-physical properties of hardened pastes
The water absorption and the water saturation increase when WCP is more than 15% (Table 2) which can be attributed to the fine internal capillary porosity of WCP particles due to the presence of old cement stone. The incorporation of WCP at 15% does not cause any changes in the absorption process (even the absorption is marginally lower), because the WCP particles remain “wrapped” by the new cement paste and the blended binder properties are governed by the new cement paste. When the amount of WCP is increased, its porous particles can absorb water more easily and this explains the very high value of the coefficient of saturation of WCP45 (Table 2) – 93% of voids, accessible to water are filled during 120h of immersion in water, while for C100 and WCP15 this percentage does not exceed 80%. The materials are considered vulnerable to freeze-thaw cycles when the coefficient of saturation is more than 80% but this negative result for WCP 30 and WCP45 can be attributed also to the presence of more structural defects due to the relatively lower workability of this mix.

Table 2. Hydro-physical properties of hardened blended binders (WCP from “young” concrete).

| Binder designation | Water absorption, % | Water saturation, % | Coefficient of saturation |
|--------------------|---------------------|---------------------|--------------------------|
|                    | After 24h | After 36 h | After 120 h | After 24h | After 36 h | After 120 h | After 24h | After 36 h | After 120 h | After 24h | After 36 h | After 120 h |
| C100               | 2,18     | 2,47     | 2,52    | 3,21     | 0,79     | 0,79     |
| WCP15              | 2,12     | 2,42     | 2,48    | 3,27     | 0,76     | 0,76     |
| WCP30              | 3,00     | 3,67     | 3,73    | 4,59     | 0,81     | 0,81     |
| WCP45              | 5,11     | 5,96     | 6,08    | 6,50     | 0,93     | 0,93     |

The coarser porosity of WCP45 is best illustrated by the capillary absorption (Figure 3). The initial capillary absorption (usually measured after 1 hour of partial immersion) is more than 3 times higher than that of the control mix, but the coarse porosity continues to be filled-in up to the 4th hour and the absolute value is 4,6 times higher. The sorptivity of WCP45, representative for the fine porosity, is also significantly higher, while that of WCP15 and WCP30 is practically the same as that of C100. The initial absorption of WCP15 and WCP30 is also more significant than that of the control paste.
3.3. Strength of hardened blended binders.
The replacement of cement by WCP originating from old concrete waste leads to a certain strength decrease. At 28 days the decrease for WCP15 and WCP30 is marginal (only 2 and 3% respectively), within the limits of test precision and can be therefore neglected. The replacement by 45% has also non-proportional reduction of only 10%.

It shall be noted that at early age (7 days) WCP30 and WCP45 have lower strength by 10 and 17% respectively, but the hardening process seems to be more intensive and the difference for WCP30 with the control paste is practically eliminated at 28 days and significantly reduced for WCP45 (Figure 4a). There is practically no difference in compressive strength of blended binders when WCP from the given “young” and “old” concrete waste were used in the same proportion for cement replacement – Figure 4 (a) and (b). This finding needs to be further investigated.

The curing seems to play more important role on the strength development of blended binders. The strength of C100 follows the usual trends (autoclaving accelerates the hardening, but exhausts the strength gain capacity; the air conditions are less favourable; at 28 days practically there is no difference in strength between the moist curing and curing in water), while the curing impact on WCP looks somehow surprising – the lowest strength is determined after the autoclaving and the moist curing is more favourable than the curing in water for 28 days strength (Figure 5).
Figure 5. Impact of curing regime on control hardened binder and WCP30 (“young” concrete waste).

It seems the autoclaving has induced self-destroying processes, while a mobilization of quartz mill in pozzolanic type reactions was expected. This result requires further research. Because of the more porous structure of WCP30 it is normal that the air conditions lead to more intensive evaporation and overdrying of concrete surface, which results in lack of water for hydration processes.

4. Conclusion
This first technical part of the study confirmed the possibility to use recycled waste concrete powder as a cementitious supplement in blended binders, without the need of additional thermal activation of the recycled material. When the replacement of cement by WCP is not more than 15%, the main properties of the blended binder – workability of fresh paste, compressive strength, values of water absorption and water saturation, remain practically the same as those of cement without supplements.

The replacement degree of 30% has no significant effect on strength at 28 days but leads to an increased coarse porosity of hardened paste and, thus, to higher water absorption. The early age strength is about 13% lower. The initial setting time is extended by almost 30%. The use of this binder would be then limited by certain ambient conditions and a longer moist curing will be required.

WCP of 45% in the blended binder can be considered as a maximum, because the water absorption and water saturation are up to 3 times higher and the 28-days compressive strength is reduced by 10%. The setting of the binder is even more postponed than in WCP-30. The use of WCP in such big proportion can be motivated by higher positive environmental effect but is limited to a few applications.

It can be therefore concluded that WCP is rather an active supplement able to compensate the reduction of clinker in the blended binder than just quasi-inert filler. Further investigations are to be made on the dependency of this effect on the amount of non-hydrated primary cement, or on a complex of factors (such as alcalis, products from primary clinker minerals hydration, etc., carbonates and quartz dust) in order to control the performance of blended binders. Another significant challenge to be resolved is the unavoidable presence of impurities in WCP that result from CDW.

Acknowledgments
The research is performed within the Project BG05M2OP001-1.002-0019: “Clean technologies for sustainable environment – water, waste, energy for circular economy” (Clean&Circle), for development of a Centre of Competence, financed by the Operational programme “Science and Education for Smart Growth” 2014-2020, co-funded by the EU through the European structural and investment funds.
References

[1] Andrew R M, Global CO₂ emissions from cement production, 1928–2018, Open Access Earth System Science Data, https://doi.org/10.5194/essd-2019-152 (last accessed July 5th, 2020)

[2] A new circular economy action plan for a cleaner and more competitive Europe, Communication from the Commission to the European parliament, the Council, the European economic and social committee and the Committee of the regions, COM(2020) 98 final, 2020

[3] Hassan A, Arif M, Shariq M, Use of geopolymer concrete for a cleaner and sustainable environment – A review of mechanical properties and microstructure Journal of Cleaner Production Volume 223, 20 June 2019, pp 704-728 https://doi.org/10.1016/j.jclepro.2019.03.051

[4] Paris J M, Roessler J G, Ferraro C C, DeFord H D, Townsend T G, A review of waste products utilized as supplements to Portland cement in concrete, Journal of Cleaner Production, Elsevier, Volume 121, 10 May 2016, pp 1-18, https://doi.org/10.1016/j.jclepro.2016.02.013

[5] Kim Y J, Choi Y W, Utilization of waste concrete powder as a substitution material for cement, Construction and Building Materials, Volume 30, May 2012, pp 500-504

[6] Berndt M L, Properties of sustainable concrete containing fly ash, slag and recycled concrete aggregate. Construction and Building Materials, Volume 23, Issue 7, July 2009, pp 2606-2613.

[7] Jaroslav T, Prošek Z, Plachý T, Influence of increasing amount of recycled concrete powder on mechanical properties of cement paste. IOP Conference Series: Materials Science and Engineering 236 012094, 2017.

[8] Sui Y, Ou C, Liu S, Zhang J, Tian Q, Study on properties of waste concrete powder by thermal treatment and application in mortar, Applied Sciences 10(3):998, doi:10.3390/app10030998.

[9] Wang J, Mu M, Liu Y, Recycled cement. Construction and Building Materials Volume 190, 30 November 2018, pp 1124-1132

[10] Xuan D X, Shui Z H, Rehydration activity of hydrated cement paste exposed to high temperature, Fire and Materials 35(7), pp 481–490, 2011

[11] Shui Z, Xuan D, Chen W, Yu R, Zhang R, Cementitious characteristics of hydrated cement paste subjected to various dehydration temperatures. Construction and Building Materials Volume 23, Issue 1, January 2009, pp 531-537

[12] BDS EN 196-6: Methods of testing cement - Part 6: Determination of fineness

[13] BDS EN 196-3: Methods of testing cement - Part 3: Determination of setting times and soundness

[14] BDS EN 196-1: Methods of testing cement - Part 1: Determination of strength.

[15] BDS EN 12808-5: Grouts for tiles - Part 5: Determination of water absorption

IOP Publishing
IOP Conf. Series: Materials Science and Engineering 951 (2020) 012008 doi:10.1088/1757-899X/951/1/012008