Review Report from Reviewer 2

Comments and Suggestions for Authors

The study investigated the performance of thermo-activated cement pastes, showing that specific heat treatments affected the reactivity and mechanical strength of blended cements.

The paper is well written, the methods section is sufficiently detailed, the presentation of the results is brilliant, condensing a lot of information in the figures without making them difficult to follow. The paper could be improved in two aspects which, in my opinion, should not be considered as major corrections but as minor ones:

The authors would like to thank the reviewer for the constructive comments and reading the paper very carefully.

1) I think the authors should state a hypothesis (or an answer to their research question that is a priori consistent with their aim). This would make the paper easier to follow. The authors have set out the variables they are trying to use to demonstrate their aim, but I still think it is better to use a clear hypothesis.

It is updated according to the suggestion.

It is hypothesized that the thermal treatment of hydrated cement paste, CEM I and CEM III, could significantly enhance the performance of OPC as a supplementary cementitious material (SCM).

2) The conclusions listed are usually not to my liking, but by no means should this comment be taken negatively, it is my point of view. But what I would like is for the authors to say at the beginning of the conclusions whether or not their hypothesis has been fulfilled. Then they can conclude whether or not their variables have allowed them to prove that hypothesis.

The conclusions have been updated, including a comment on the hypothesis.

In this research, dehydrated cement (DC) powders (referred to as DCPs) were prepared via thermal treatment and comminution of CEM I and (synthetic) CEM III hydrated pastes to investigate their physico-chemical properties and cementitious performance as supplementary cementitious materials (SCMs). A full hydration study was conducted to explore the effects of partially replacing fresh cement (both CEM I and CEM III) with such SCMs. The hypothesis that DCPs can be used as SCMs was only confirmed in part and care should be taken because the DCPs change the workability, reactivity, and final strength. The main findings of this research are summarized as follows.

Increasing the dehydration temperature from 350 °C to 750 °C generally results in an increase in the grindability of these materials and their surface area, with an exception observed for DCIII dehydrated at 750 °C. Recrystallization of C-S-H at 550 °C and higher temperatures leads to the formation of γ-C2S and β-C2S. The content of potentially hydraulic phases increased with dehydration temperature at the expense of the amorphous or nanocrystalline phase (likely C-S-H and other cement hydrates).

According to the R3 reactivity results, DCPs prepared at higher dehydration temperatures and times generate significantly higher initial rates of reaction compared to those prepared at lower temperatures and also show higher reactivity in the long term (after 1–2 days). Positive correlations were found between the R3 heat results and the amount of potentially hydraulic phases formed in the DCPs (for both DCI and DCIII), although most correlations were not statistically significant. However, no direct correlation was found between the R3 heat release and the strength performance of blended cements comprising 30 wt% DCP.

Blended cement pastes incorporating DCP generated similar or lower bound water and portlandite content compared to the control paste (I-CTRL). Pastes incorporating DCPs prepared at higher temperatures generated higher bound water compared to other blended pastes, but this did not translate to better strength performance. Incorporation of DCP in the pastes results in high initial heat generation in the first 1–2 hours and expedites the acceleration period. However, pastes incorporating DCPs produced at higher temperatures (550°C/24h and 750°C/2h) show significant retardation of the main hydration of C3S, in line with the lower C3S hydration rate (according to the Rietveld analysis results of in-situ XRD) of such pastes at that time.

DCP blended cements contain higher levels of crystalline ettringite and AFm hydration products throughout the cement hydration process. AFm phases form early, and gypsum is depleted before the main C3S hydration event, indicating undersulfation and disturbance of the regular hydration process, which could affect further hydrate nucleation, growth, and microstructure development.

The best strength performance for CEM I pastes was observed for DCI/350°C/2h, followed by DCI/550°C/2h, 550°C/24h, and 750°C/2h. Therefore, dehydration at temperatures exceeding 350 °C is not recommended for this DC type. For CEM III pastes, DCIII/550°C/24h showed the best early-age strength performance, while DCIII/350°C/2h displayed similar strength activity at later ages (after 28 days). Hence, increasing the dehydration temperature to 550 °C for extended periods is not justifiable unless high early-age strength is desired. All DCIII materials and their respective blended mixtures had a slower rate of strength development compared to the corresponding DCI materials and pastes. However, they exhibited higher strength results at later ages compared to DCI.

Other minor formatting and typographical corrections are detailed in the attached PDF file, which I hope the authors will find useful.

They are corrected. Thanks a lot!

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