Dear Editor Icy Liu and Reviewers:

Thank you for your letter and for the reviewers’ comments concerning our manuscript entitled “Study on the Preparation and Performance of Alkali-activated Coal Gangue-Slag Cementitious Materials” (Manuscript number: materials-533306). Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. We have studied comments carefully and have made correction which we hope meet with approval. Revised portion are marked in red in the manuscript. The main corrections in the manuscript and the responds to the reviewer’s comments are as following:

Responds to the reviewer’s comments:

Reviewer #2:

Special thanks to you for your good comments. We revised it according to your suggestions and replied to your questions. In the introduction section, we highlight the research background and significance, and supplement the references related to the research content of this paper. We have supplemented the experimental process and curing conditions, and we have also proofread and polished the conclusion, making the study of this manuscript more meaningful. Other modifications details are marked in red in the manuscript. Thank you for your time and recognition. I look forward to the opportunity to discuss academic topics with you. thank you!

1. Response to comment: Place the question addressed in a broad context and highlight
the purpose of the study;

Response: Thank you for your suggestion. According to the reviewer's comments, the
background of the study was supplemented in the introduction section and marked in
red in the text manuscript.

Research background: Coal gangue is the solid waste produced in coal mining. In
countries with developed coal resources, such as China, India, Australia, Germany, the
United States and South Africa, the discharge of coal gangue waste remains high. A
large amount of coal gangue waste not only occupies land, but also causes great damage
to ecological environment.

Coal gangue contains some approximate composition similar to clay and coal
gangue calcined after can can obtain higher activity of volcanic ash. With the deepening
of research on alkali-activated cementitious materials, coal gangue is used as raw
material for alkali-activated cementitious materials, and slag is used as calcium source
to prepare AACGS cementitious materials. Seeking to use coal gangue as a building
cementing material and increase the utilization rate of coal gangue. The background
and significance of the research are added in the introduction, and the specific
modifications are marked in red in the introduction.

2. Response to comment: Highlight the importance of the study and briefly place the
study in a broad context;

Response: Thank you for your suggestion. According to the opinions of reviewers, the
research background is elaborated in the introduction and the conclusion were polished.
The significance and bright spots of the research. The corresponding modifications are
3. **Response to comment:** Present the contribution of this work to the current state of knowledge on the subject;

**Response:** Alkali-activated cementitious materials became the most promising alternative materials of Portland cement due to their merits including early strength, high environmental adaptability and low environmental impact. Among many industrial solid wastes, coal gangue is one of the industrial solid wastes with the largest emission (about 10%~25% of the total amount of coal mining). Currently, coal gangue is mainly used to pave roads, produce cements and prepare sintered bricks, indicating the low comprehensive utilization ratio, and there is an urgent need to seek new ways of utilization. With the research of alkali activated cementitious materials in recent years, coal gangue as alkali activated cementitious material becomes a new utilization direction.

At present, the research of alkali-activated cementitious materials mainly focuses on slag, fly ash and metakaolin, while the research on coal gangue, red mud and rice husk ash only appears in recent years. The research on coal gangue is not systematic. The research group's previous research shows that it is not economical to use pure coal gangue as the raw material of alkali activated cementitious materials to increase the alkali concentration. In this paper, alkali activated cementitious materials from coal gangue and slag were prepared with slag as calcium source, which provided experimental and theoretical basis for the development of alkali activated cementitious materials and comprehensive utilization of coal gangue.
4. **Response to comment**: Considering the differences in SiO$_2$ and Al$_2$O$_3$ content % between the gang coal and the slag what are the expectations in terms of final results by replacing each other?

**Response**: Coal gangue is a silicon-aluminum material, and slag is a silicon-aluminum-calcium material. Our research group has studied the alkali-activated coal gangue cementitious materials before studying the alkali-excited coal gangue-slag binary cementitious materials. As shown in Fig.3, when the NH molar concentration exceeds 12M, the compressive strength value increases little, but the compressive strength value of 28d is lower than that of cement slurry, therefore, slag is used as the calcium source for reinforcement.

The content of SiO$_2$ and Al$_2$O$_3$ in the slag is lower than that in the coal gangue. The mixture of coal gangue and slag reduces the content of SiO$_2$ and Al$_2$O$_3$, but increases the content of CaO. Calcium aluminum silicates (C-(A)-S-H gels) achieve greater strength than sodium aluminum silicates (N-A-S-H gels). As the slag content increases, the compressive strength increases. From the chemical perspective, the reduction of SiO$_2$ and Al$_2$O$_3$ content in the mixed system does not affect the occurrence of chemical reaction, and the SiO$_2$ and Al$_2$O$_3$ content in the system is the highest.
Fig. 3 The compressive strength of alkali-activated coal gangue specimens with different NH molar concentration

5. **Response to comment:** A brief description of the manufacturing and mixing process of the samples studied could be presented, only the bibliographic reference seems to me little given the importance of the process.

**Response:** Thank you for your suggestion. and your suggestion is also very important. According to the suggestion of the reviewer, the specific experimental operation and test method are added in the manuscript in section 2, which makes the expression of the manuscript more abundant. Specific modifications are marked in red in the manuscript.

6. **Response to comment:** What was the time and temperature of the sample curing process?

**Response:** All samples in this study were cured by standard curing(95±1%, T=20±2℃). Poured the fresh paste into steel dies (40 mm×40 mm×40 mm) and vibrated them for 60 s on an electric vibration table to remove residual air. The dies were covered with thin polyethylene films and cured for 1 day at standard curing, they
were then demoulded, transferred to standard curing rooms and cured for 3d, 7d and 28 day respectively.

7. Response to comment: What is the reason for using mostly a NaOH molar concentration of 12?

Response: Thank you for your inquiry. This paper discusses the impacts of slag content, NaOH molar concentration, alkali lye amount and liquid-solid ratio on AACGS cementitious materials. The NH molar concentration was selected for 12M for two reasons:

(1) It is shown that the optimal modulus of the alkali activator is 1.3 by reading a large number of references. According to the sodium silicate and NH adopted in the experiment in this manuscript, when the NH molar concentration is 12M, the modulus of the alkali activator is calculated to be 1.3.

(2) Fig.3 shows the compressive strength of alkali-activated coal gangue under different NH molar concentrations (slag=0%). An increase in the NaOH concentration from 8 M to 12 M clearly increased the strengths of the coal gangue geopolymer. Increasing NaOH concentration from 12 M to 16 M also increased the strength of coal gangue geopolymer but to a lesser extent. In order to reduce NH consumption and save costs, the NH molar concentration was selected as 12M.

8. Response to comment: What is the justification for the remaining NaOH solution molarity (M) concentrations, namely 8, 10, 14 and 16?

Response: Thank you for your inquiry. The important influencing factor studied in this
manuscript is NH molar concentration. According to the previous adaptation of the research group, when NH molar concentration is less than 8M, the compressive strength value of alkali activated gangue is lower (As shown in Fig. 3, when NH molar concentration is 8M, the 28d compressive strength of alkali-activated gangue is only 29.82% of that of cement, and the compressive strength increases rapidly with the increase of NH molar concentration.). Late in the AACGS cementitious material, continued the previous ideas, found that the higher the molar concentration of NH after adaptation, the compressive strength value increases. However, when the NH molar concentration increased to 18M, the setting time of AACGS cementitious material is shorter, so the manuscript in NH molar concentration level was set to gradient larger 8M, 10M, 12M, 14M and 16M.

9. **Response to comment**: Would not it be equally important to evaluate the shrinkage of the samples studied?

**Response**: Thank you for your suggestion. It has been found that the micro-cracks caused by shrinkage are the most important reasons that limit the widespread use of this most promising alternative gelling material. Our research group has carried out research on the drying shrinkage characteristics and mechanism of AACGS mortar, and also found effective measures to inhibit drying shrinkage. Relevant academic papers will be published within this year. I hope to have the opportunity to have related academic discussion and study with reviewers.

10. **Response to comment**: In line 101, authors refer to table 2, but should probably be table 3!
Response: Our mistakes have brought difficulties to reviewers, we are very sorry for our negligence of writing. We have modified the errors in the manuscript, and marked them in red.

11. Response to comment: Would not it be possible to cross-reference the results obtained with XRD and FT-IR?

Response: Thank you for your suggestion. XRD analyzes the composition of the material and analyzes the crystalline or amorphous substance. FTIR analyzes the characteristic functional groups of compounds, and measures information based on the relative vibration of atoms within molecules and molecular rotation. The manuscript analyzes the stretching and bending vibration of the $-\text{OH}$, $\text{H-O-H}$, $\text{Si-O}$ and $\text{Si-O-Al}$ bands of the hydration products. XRD and FT-IR are both independent methods to determine the hydration of AACGS cementitious materials. However, through the analysis of the manuscript, it was found that XRD can be cross-referenced with SEM-EDS analysis results. We revised the section 4 and marked it in red in the manuscript.

12. Response to comment: The analysis of SEM-EDS results is somewhat reductive!

Response: Thank you for your suggestion. As shown in Fig. 2 of the manuscript, the compressive strength value increases with the increase of slag content. However, when the slag content is 20%, 30% and 40%, the increase range of compressive strength is not much different. Therefore, three groups of samples with large intensity gradient were selected for testing, so as to make analysis more clearly.
Moreover, by observing the SEM-EDS diagrams in the manuscript, it can also be seen that the difference of the SEM diagrams of the three diagrams lies in the amount of gelatinous and floccular products, and the comparison of the three groups with a large gradient will be more obvious.

Thank you very much for your letter and advice on our manuscript. We quite appreciate your favorite consideration and the reviewer’s insightful comments. We tried our best to improve the manuscript and made a little changes in the manuscript. These changes will not influence the content and framework of the manuscript. We did list the changes and marked in red in revised manuscript (except grammatical and English language). We appreciate for Editor Icy Liu and Reviewers warm work earnestly, and hope that the correction will meet with approval.

Once again, thank you very much for your comments and suggestions. We hope that the revision is acceptable and look forward to hearing from you soon.

With best wishes,

Ma Hongqiang, Zhu Hongguang*, Yi Cheng, Fan Jingchong, Chen Hongyu, Xu Xiaonan and Wang Tao

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