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The Properties of Normal Concrete with Ground Manganese Slag as Binder Replacement

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ABSTRACT

Manganese slag is a by-product from the production of ferrosilicon and manganese alloys. It has been found to possess pozzolanic properties and is therefore suitable for use in concrete mixtures. This paper investigates the properties of normal concrete with ground manganese slag as a binder replacement in terms of workability, compressive strength, flexural strength, and leaching characteristics. In this study, it is found that using ground manganese slag as a binder replacement increases the slump of fresh concrete. It is also found that using ground manganese slag as a binder replacement reduces both the development of compressive and flexural strengths. However, at a 10% binder replacement rate, the deterioration is considered minimal. The results of leaching characteristics from 'The Tank Test' show that using ground manganese slag as a binder replacement increases the alkalinity of the solution surrounding the concrete, indicating that the metal ions in the ground manganese have diffused and dissolved in the solution.

1. Introduction

Concrete is utilized in construction in two times the amount of all other building materials combined and will undoubtedly continue to be used in building construction for a very long time [1]. Cement, water, and aggregate (sand and gravel or crushed stone) make up the three main ingredients of the commonly used building material known as concrete. This robust and versatile material may be used to build structures like buildings, roads, bridges, and other kinds of infrastructure. Concrete is a popular building material because of its durability, affordability, and strength.

The cement, which binds the elements of concrete together, is currently accountable for 8% of the world's emissions of carbon dioxide (CO_2) gas [2], the primary greenhouse gas causing climate change. Even though it appears clear that since its invention in the 19th century in its current form, concrete has been and will continue to be a fundamental material for the construction industry. After fossil fuels and land use change, cement production has dramatically expanded globally in recent

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years, making it the third-largest source of anthropogenic carbon dioxide emissions. With a production growth of 2.5% annually, it is anticipated to increase to 3.7-4.4 billion tons by 2050 [3]. The cement production in 2022 has been recorded as 4.1 billion tons by Garside [4].

In Sarawak, Malaysia, due to the development of the Samalaju Industrial Park, a number of metal smelting plants have been established recently and has been in full production since almost 5 years ago [5-7]. Manganese slag has been the by-products from these industries and they are categorised as scheduled wastes by the Department of Environment (DOE), Malaysia. As a result, storage of these wastes have become an issue with substantial amount of waste pilling up on the premises of the factories. According to Chen *et al.,* [8], manganese slag comprises of harmful heavy metals and will produce excessive sulphate and ammonia nitrogen that can potentially pollute the surrounding environment and harm the aquatic ecosystem and agriculture system. Therefore, a proper disposal method of the industrial waste residue is vital to ensure the safety of the environment in this region. One of the strategies to ensure the sustainability of the environment is to implement manganese slag as a binder replacement in concrete. To the best knowledge of the authors, the research work in this respect is still onging and therefore motivated the investigation reported herein. Thus, a literature review is reported in the impending discussion followed by the report on experimental work on using ground manganese slag as binder replacement in normal concrete. To the authors' best knowledge, ongoing research in this domain has served as the drive for the current investigation. Consequently, the forthcoming disussion includes a comprehensive literature review, followed by an exposition of experimental works focused on the utilization of ground manganese slag as a substitute binder in conventional concrete.

Generally, there are two types of manganese slag, namely ferromanganese slag and silicomanganese slag; that can be produced from the metal smelting process [9] as shown in Figure 1. The differentiation between these two types of manganese slag is through their manganese oxide (MnO) and silica (SiO_2) content [10]. The comparison of MnO, SiO_2, Calcium Oxide (CaO), magnesium oxide (MgO), Aluminum Oxide or Alumina (Al_2O_3) and Iron (III) Oxide (Fe_2O_3) content in ordinary Portland cement, ferromanganese slag and silicomanganese slag [11-13] is shown in Table 1.

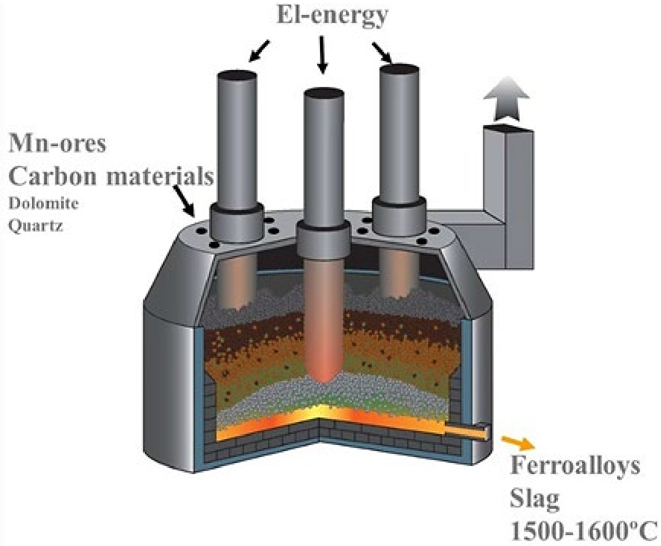


Fig. 1. Typical production of manganese slag in closed furnace (Tangstad *et al.,* [9])

Table 1
Comparison of oxide content in ordinary Portland cement, ferromanganese slag and silicomanganese slag

Oxide (%)	Ordinary Portland cement (Shahidan *et al.*, [11])	Ferromanganese slag (Groot *et al.*, [12])	Silicomanganese slag (Frias *et al.*, [13])
MnO	-	28-30	4.20
SiO_2	21.06	28-30	42.60
CaO	57.98	28-30	25.20
MgO	2.74	6-6.5	4.20
Al_2O_3	6.10	5-5.5	12.20
Fe_2O_3	3.08	1-1.3	1.0

It can be seen in Table 1 that both ferromanganese and silicomanganese slags exhibit a similar composition as ordinary Portland cement. Due to pozzolanic properties, manganese slag can be used as a substitution of cement. Nath and Kumar [14] performed a study on the suitability of ground granulated silicomanganese slag in Portland cement. According to the study, the major component in silicomanganese slag has a similar hydration property as typical cement, which can produce calcium silica hydrate (C-S-H) and calcium aluminate hydrate (C-A-H) when exposed to water. Meanwhile, the strength gain of the manganese slag blended cement has also shown an optimistic result. However, research on effect of manganese slag on other mechanical properties such as durability and workability are fairly limited. Thus, further research on the effect of manganese slag on concrete properties is deemed necessary.

Some preliminary studies on utilising silicomanganese slag as binder replacement material in concrete had been carried out by Frias *et al.,* [13], and Altun and Yılmaz [15] with 5% to 45% of replacement. It was found that the 7th day compressive and flexural strengths of the mortars had been compromised by 10-25% but the effect was less profound at the 28th and 90th day strengths. It is important to note that this study was carried out on mortars with limited variation on percentage of replacement. Therefore, there is still a need in research of actual concrete behaviour with more variation in replacement percentage in order to utilise this waste material efficiently. Hence, the need persists for further investigation into the concrete's actual behaviour, incorporating a more diverse range of replacement percentages, to optimize the efficient utilization of this waste material.

Liang *et al.,* [16], attempted the utilization of manganese slag as cement replacement material at a range of 0-60% of replacement. Flexural strength and compressive strength testes were carried out at 3rd and 28th day of curing. It was found that the flexural strength at 3rd day shows a decreasing trend when the manganese slag content is increased. The low flexural strength in the early stage may be due to slow reactivity of alumina in manganese slag. Meanwhile, the compressive strength shows a slight increase with the increase in manganese slag content. However, the trend prickled up when manganese slag content is more than 20%. On 28th day of curing, both compressive and flexural strengths development show an almost identical growing pattern with an increase in strength of concrete with manganese slag content up to 20%. The sign of the strength increase is less profound when manganese content is beyond 20%.

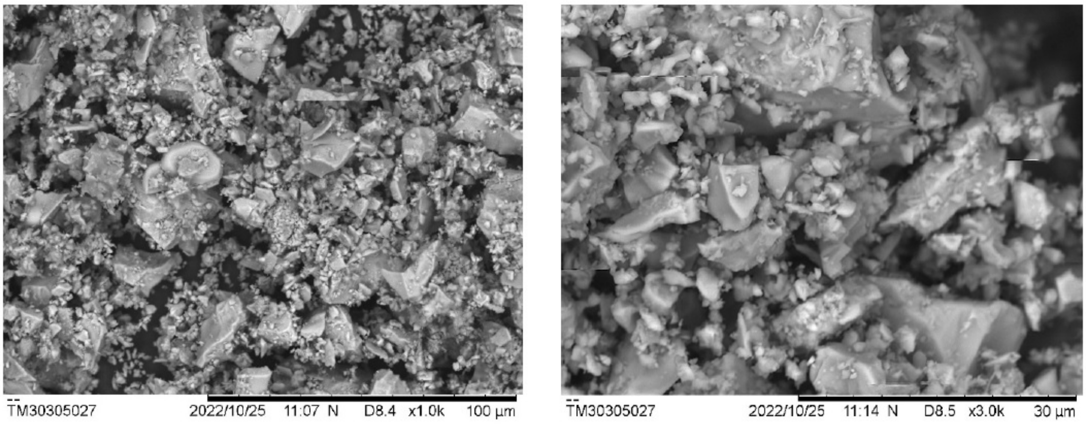
In a separate study conducted by Wang *et al.,* [17], it was found that manganese slag mixing with fly ash has a potential application in geopolymer concrete through alkali activation. However, this study only provides preliminary results indicating the potential use. Therefore, further investigation into utilising manganese slag in geopolymer concrete is still a necessity. A further review on alkali-activated binder was carried out by Marsh *et al.,* [18], in regards to the understanding of relationships between slag composition and hardened cement properties, as well as their long-term safety and durability. Beyond purely material aspects, this review puts forward the development of alkali-activated binder into context of the recycling industry and discusses the opportunities and hurdles for greater adoption.

An extensive review on characteristics of silicomanganese slag and its utilization into construction materials was conducted by Nath *et al.,* [19]. It was found that crystalline hard stone type slag was mainly used as aggregates whereas granulated glassy form slag was used as binder replacement or precursor for alkali-activated binder. It was also reported that the binding capability of the cement gel can be improved by the reactive component such as but not limited to CaO and SiO_2. As for the leaching of toxic and heavy metals from the silicomanganese slag reacted binder matrix, no report has been found. However, Drinčić *et al.,* [20], found that there is possible leaching of heavy metals when waste material was used in building composites even when immersed in solution like demineralized water. Liu *et al.,* [21], reported that mortars with cement replaced by 30-50% silicomanganese slag showed excessive manganese leaching concentration through the sulphuric acid-nitric acid leaching tests, indicating the high environmental risk of silicomanganese slag utilization under special service conditions.

It is noteworthy that besides manganese slag, there are some other sustainable materials that can be used in concrete and geopolymer concrete such as metakaolin [22] and alkali-activated cement [23-25] as well as multiwall carbon nanotubes [26].

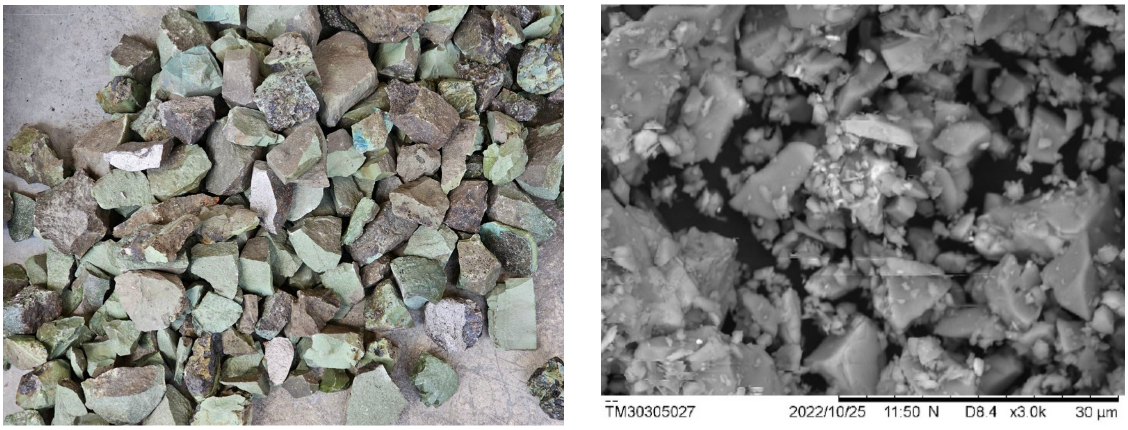
2. Materials and Methods

The concrete mix used in this study is grade C25 with cement-to-fine aggregate-to-coarse aggregate ratio of 1:2:4 and water-to-binder ratio of 0.56. The targeted cube strength is 25 MPa at 28 days of curing with medium workability for the fresh concrete. For compressive strength and flexural strength tests, cubes of 100 mm dimension and prisms with 100 mm square cross section and length of 500 mm were cast in accordance with guidelines in BS EN 12390-1:2021 [32] and BS EN 12390-2:2019 [27,28]. For leaching characteristic test, cylinders of 50 mm diameter and 100 mm length were cast. Crushed granite of 20 mm nominal diameter was used as coarse aggregates whereas river sand was used as the find aggregates for casting of the specimens. For the binder, ordinary Portland cement with a median particle size (D50) of 20 µm was used as shown in the scanning electron microscopy (SEM) images in Figure 2. Crystalline hard stone type manganese slag was crushed to gravel size as shown in Figure 3(a) and then ground into powder form to achieve similar D50 of cement as shown in Figure 3(b), which was recorded as 15 µm. The oxide content of the manganese slag used is similar to that used in Groot *et al.,* [12].



(a) 100 µm scale (b) 30 µm scale

Fig. 2. Scanning electron microscopy (SEM) images of ordinary Portland cement



(a) Crushed manganese slag (b) SEM at 30 µm scale

Fig. 3. Crushed manganese slag and scanning electron microscopy (SEM) of ground manganese slag

Four batches of specimens were cast with ground manganese slag serving as binder replacement by weight of cement, namely 0% replacement (control), 10% replacement (R10), 20% replacement (R20) and 30% replacement (R30), respectively. For each ~~batch of~~ casting, the coarse aggregates were immersed in potable water for 24 hours before casting and the surfaces were wiped dry to achieve saturated-surface-dry condition during casting. Cube, prism and cylinder moulds were wiped with a thin layer of recycled engine oil before casting. During casting, slump test was carried out on the fresh concrete in accordance with BS EN 12350-2:2019 [29]. The cast concrete specimens were unmoulded 1 day after casting and labeled. The cubes and prisms were put in curing tank filled with potable water at ambient temperature until all the specimens were fully submerged. The cylinders were put in a container filled with demineralized water as shown in Figure 4(a), and the specimens were submerged in the container as shown in Figure 4(b), with sufficient space in between specimens in accordance with NEN 7375:2004 [30] leaching characteristic test procedure.

(a) Demineralized water filling (b) Submerging specimens in container

Fig. 4. Preparing specimens for leaching characteristic test in accordance with NEN 7375:2004 [30]

The compressive strength and flexural strength tests were carried out in accordance with BS EN 12390-3:2019 [31] and BS EN 12390-5:2019 [32] respectively. Compressive strength test was carried out for 1, 3, 7, 14 and 28 days of curing whereas flexural strength test was carried out for 7 and 28 days of curing. On the day of testing, the specimens were removed from the curing tank and let dry for at least 2 hours before testing. Each cube was tested using compression machine, and the recorded compressive strength is the average of 3 test cubes. Each prism was tested under two-point loading, and the maximum load was converted to flexural strength, with the recorded flexural strength is the average of 3 prisms data.

The solution for the leaching characteristic test is demineralized water, as described earlier. The solution was replenished at specific intervals in accordance with NEN 7375:2004 [30]. The monitoring of the solution pH was carried out at 1, 7, 28, 56, 64, 84 and 90 days of concrete age. For comparison purposes, another set of modified test was conducted on non-replenished solution.

3. Results and Discussion

3.1 Workability

Slump test is one of the most common and effective methods that is used to measure the consistency and the workability of the fresh concrete before it sets. The slump value of the fresh concrete indicates the deformation magnitude of the mixture under the influence of gravity. This value helps to ensure that the concrete mixture possesses the desired flowing properties for the specific application. On the other hand, the workability of fresh concrete can also have a direct impact on the strength development of the concrete final products.

Figure 5 shows the results of the slump is replaced with manganese slag up to 20%, with a maximum increase in workability of 157% in R20 concrete batch compared with control concrete batch. However, the increase of manganese slag content in concrete mix will result in a higher surface area of pozzolans and creates a higher water demand. Since the amount of water is constant for all design mixes, further increase of manganese slag beyond 20% of replacement will result in inadequate water to act as a lubricant between particles. This is shown by the decrease in workability when manganese slag replacement is increased to 30% as depicted in Figure 5. In general, the utilization of ground manganese slag as cement replacement material increases the workability of fresh concrete, as shown by the lowest slump value of the control concrete batch.

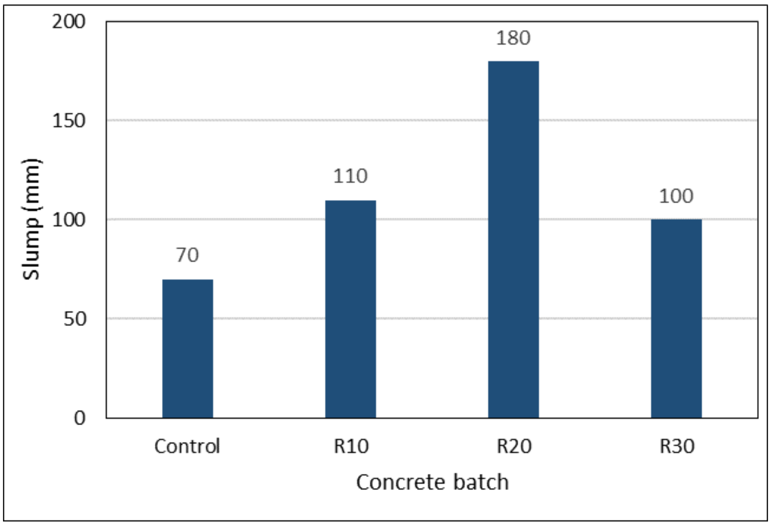
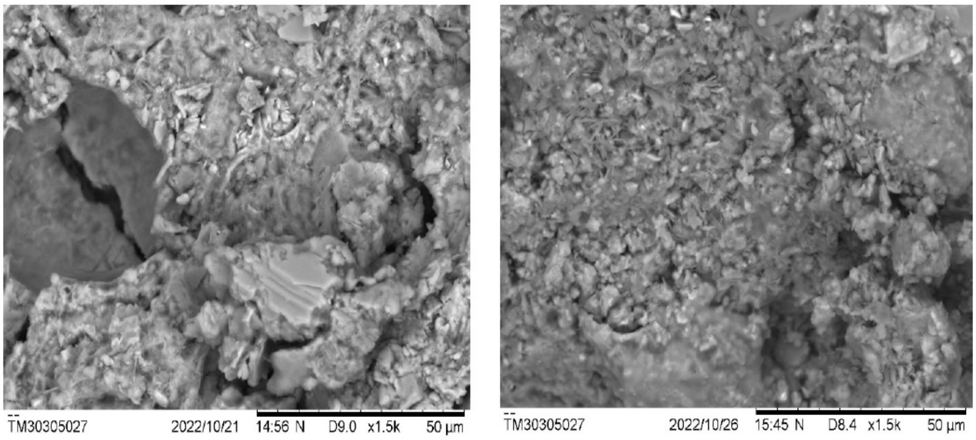


Fig. 5. Workability of concrete measured with slump test

The sign of increase in workability was attributed to the fact that the ground manganese slag used has finer particles (D50 = 15 µm) and a smoother surface then the cement used (D50 = 20 µm), which makes it to act as a filler on the voids between the cement particles. The packing density of concrete was improved with cement replaced by ground manganese slag, resulting in a more homogeneous and workable mixture as evident in the comparison of microscopic structure of hardened control specimen and R20 specimen as shown in the SEM images in Figure 6. R20 specimen with 20% ground manganese slag replacement [Figure 6(b)] has a more homogeneous microstructure compared with control specimen [Figure 6(a)] that has no cement being replaced with ground manganese slag. Due to a finer and smoother surface of slag particles than cement particles, it creates a better interconnectivity between the cement particles, and results in a more homogeneous arrangement of the particles. Ultimately, it reflects in a higher workability of sample in fresh state.

In addition, calcium hydroxide that was produced by cement hydration reacted with ground manganese slag leading to the formation of extra cementitious materials, which subsequently enhances the workability of the mixture. However, the incorporation of more ground manganese slag can potentially increase the water demand due to higher surface area of finer particles as can be seen from comparison of concrete batches R20 and R30 in Figure 5. Thus, it is essential to determine the optimum amount of ground manganese slag as cement replacement to achieve the desired properties.



(a) Control specimen (b) R20 specimen

Fig. 6. SEM images of control and R20 hardened concrete specimens

3.2 Compressive Strength

Concrete compressive strength is an indicator of the maximum compressive stress that a concrete can withstand before failing in compression. This indicator is important as it determines the quality of the concrete and guarantees a safe and reliable concrete structure throughout the service life. In normal circumstances, the compressive strength of concrete is influenced by the degree of hydration and the reactivity of the pozzolanic reaction. Therefore, an investigation of the relationship between the concrete content and the associated pozzolanic activity is deemed necessary.

The compressive strength development of the concrete samples from the first day of curing until 28 days of curing are shown in Figure 7. According to the compressive strength test results as shown in Figure 7, one noticeable observation is that the strength for control specimens at 28 days is lower than the anticipated designed strength of 25 MPa. This may due to the poor-quality cement in preparing the specimens affecting the quality of the concrete. However, since all the specimens were prepared using the same cement, this does not affect the trend of the results of specimens with cement replaced by ground manganese slag.

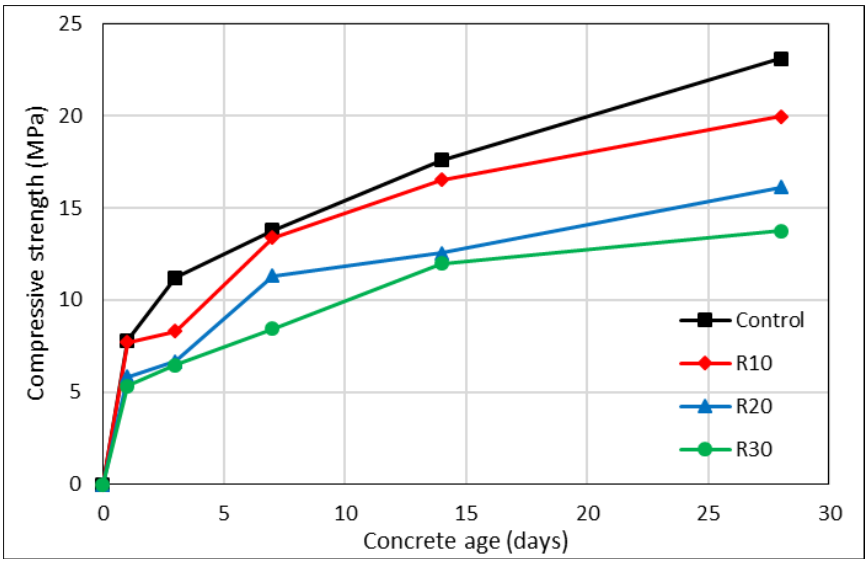


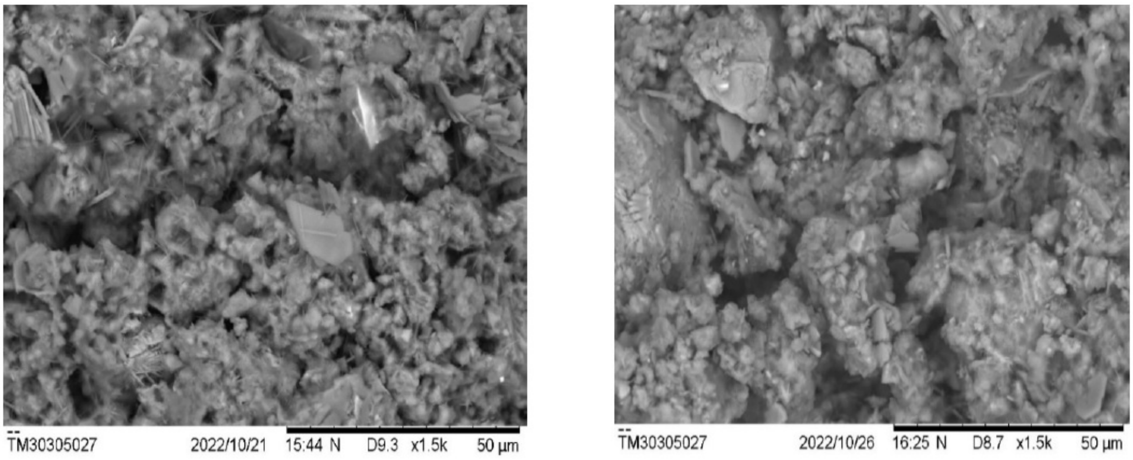
Fig. 7. Compressive strength development

From Figure 7, the compressive strengths at the ages of 1 to 28 days of curing show a steady decrease when the amount of cement replaced by ground manganese slag increases. The main reason of this may be attributed to two main aspects, namely low reactivity of manganese slag and the fineness of ground manganese slag. Owing to the elevated content of silica and alumina, recognized as inherently inert materials in ground manganese slag, there is a retardation in pozzolanic activity. This delay is evident in lower compressive strength during the early age, especially with an increased amount of ground manganese slag, aligning with findings from prior research [16].

Meanwhile, ground manganese slag that was utilized in this research is finer compared to cement. Due to the higher surface area of finer particles, the demand of water during the pozzolanic reaction is high. Considering that the amount of water provided in all designed mix is kept constant, the concrete with more ground manganese slag will have a weaker cement matrix due to the incomplete hydration, which is shown by the lower strength development for concrete with higher ground manganese slag content.

Throughout the curing age of 28 days, control specimens exhibit the highest average compressive strength at 23.1 MPa, followed by R10, R20 and R30 specimens with the average compressive strengths of 20.0 MPa, 16.1 MPa and 13.7 MPa, respectively. These results show that the compressive strength decrease is proportional to the increase in ground manganese slag replacing cement, corresponding to 13%, 30% and 41% reduction in compressive strength when cement was replaced by ground manganese slag at 10%, 20% and 30%, respectively. As mentioned earlier, the ground manganese slag contains silica and alumina which are both inherently inert and results in a slower strength development. However, both of these constituents can undergo reactions with calcium hydroxide, yielding additional cementitious materials such as ettringite—a calcium aluminum sulfate compound characterized by needle-like crystals. This material forms in the presence of calcium, demonstrating the capacity to fill voids in the concrete, enhance density, and contribute to concrete strength development at a later stage. The outcomes suggest that a 10% replacement of cement with ground manganese slag may serve as a threshold to avoid compromising the strength development of concrete. These findings align with the observations in some prior researches [14, 15].

Figure 8 shows the comparison of SEM images between R10 specimen and R30 specimen, which has the lowest compressive strength. The formation of crystal-needle-like ettringite in the R10 specimen is more conspicuous compared to the R30 specimen. This observation may be attributed to the higher content of ground manganese slag in the R30 specimen, contributing to a slower pozzolanic reaction. Additionally, the diminished ettringite formation in the R30 specimen may result from incomplete hydration, driven by the elevated water demand. Moreover, increased replacement of cement with ground manganese slag leads to a reduced presence of cement in the mix, potentially resulting in fewer hydration products.



(a) (b)

Fig. 8. Comparison of ettringite formation in (a) R10 specimen and (b) R30 specimen

3.3 Flexural Strength

Concrete flexural strength refers to the maximum bending stress in tension that a concrete can withstand before failing. This strength is typically measured by applying two-point loading on a concrete prism until the concrete fails in cracking. Flexural strength of concrete with curing age of 7 and 28 days is reported in this study.

Figure 9 shows the results of the flexural strength test, displaying a similar growth pattern across all the concrete specimens. In general, control specimens with no ground manganese slag exhibit the highest average flexural strength among all the concrete specimens. The flexural strength development decreases with the increase in ground manganese slag replacement percentage up to 30%. The relationship between the flexural strength development with the increase in ground manganese slag in the concrete specimen showcases an identical pattern as the results from compressive strength test. Thus, a similar explanation, as discussed earlier can be expected behind these findings. The decrease in flexural strength is not as substantial compared to the effect on compressive strength, with the reduction in flexural strengths of 6.1%, 27%, and 26% when cement was replaced by ground manganese slag at 10%, 20%, and 30%, respectively, at 28 days of curing. The results of R20 and R30 are almost identical at curing age of 28 days, probably due to the fact that the ground manganese slag is assuming the role of filler instead of acting as pozzolanic material at these replacement percentages.

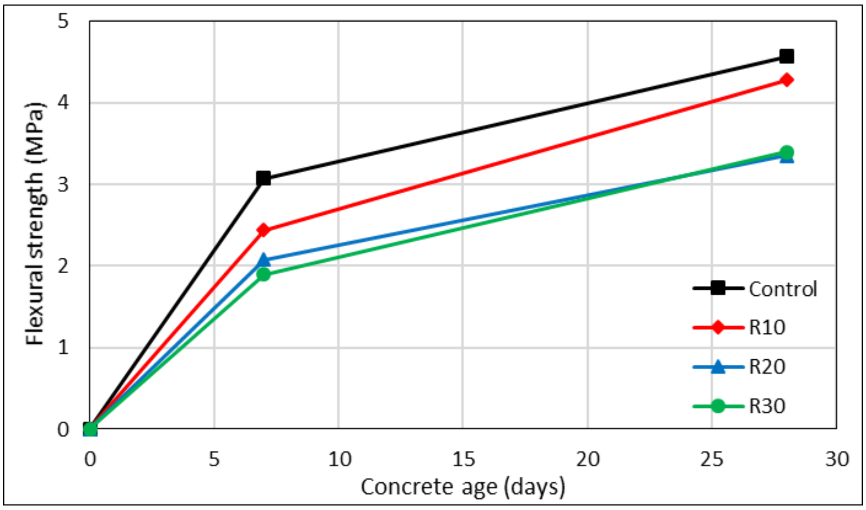


Fig. 9. Flexural strength development

3.4 Leaching Characteristic

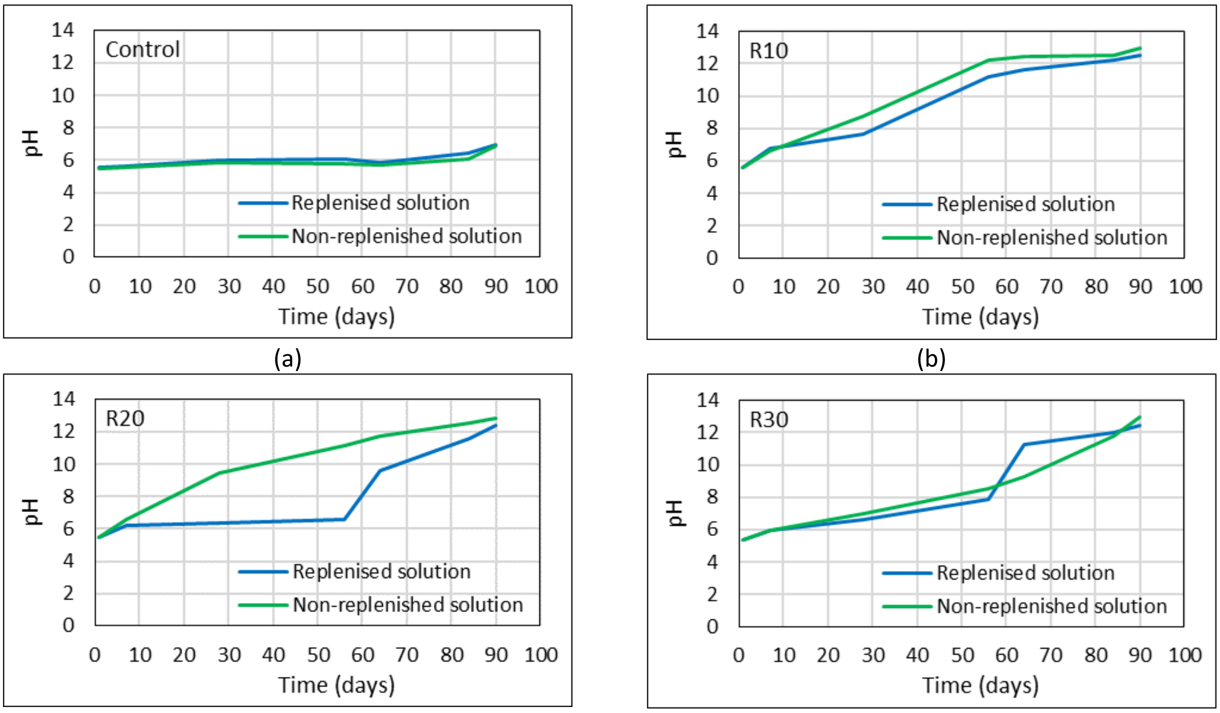
Leaching characteristic tests are conduct to gain information regarding the potential risk of waste to release organic and inorganic contaminants into the environment. Since ground manganese slag used to replace cement in this study is a waste material from the metal smelting industry which contains high metal oxides, the potential risk is assessed concerning the pH of the leachate, which indirectly indicating the concentration of metal ions in the leaching solution. Metal ions can convert into hydroxides, thus causing the leachate to have a higher pH value.

Figure 10 shows the results of the leaching characteristic test using demineralized water as the leaching solution. It can be observed that the pH value of the control specimens in Figure 10(a) changes from slightly acidic to almost neutral on the course of 1 to 90 days. The test with replenished solution (NEN 7375:2004) shows insignificant differences from the test with non-replenished solution (modified NEN 7375:2004), indicating that diffusion was the key process governing the leaching of elements from the control specimens, even when the leaching solution was not-replenished.

When cement was replaced with ground manganese slag by 10% (R10) and 20% (R20), the results of concrete specimens in Figures 10(b) and (c) show that the pH value increases faster over the course of 1 to 90 days in the non-replenished solution than the replenished solution. While diffusion is the main leaching mechanism in the replenished solution, it is observed that the additional dissolution process in the non-replenished solution accelerates the leaching of the specimens.

Figure 10 (d) demonstrates that when cement was replaced with ground manganese slag by 30% (R30), the rate of diffusion and dissolution are reduced at the earlier age of concrete up to the 56th day, possibly due to slower hydration and pozzolanic reactions. After 56 days, the rate of diffusion and dissolution increase, wherein the diffusion process alone in the replenished solution can be as rapid as the diffusion and dissolution processes in the non-replenished solution from the 56th to the 90th day. This may be due to the higher metal oxide content with more ground manganese slag present in the concrete, where higher leaching of elements can take place mainly by diffusion at the later age of concrete.

These results indicate that using ground manganese slag as a binder replacement in concrete poses the potential risk of releasing inorganic contaminants into the environment through leaching, even when exposed to demineralized water. If the concrete containing ground manganese slag is exposed to water containing aggressive substances, a higher risk of metal leaching may be expected.



(a) (b) (c) (d)

Fig. 10. pH of leachates from the tested concrete specimens

4. Conclusions

The use of ground manganese slag as a cement replacement up to 20% can lead to drastic increase in workability of fresh concrete by up to 157%. When the ground manganese slag replacement is further increased to 30%, a decrease in workability was observed.

The effect of using ground manganese slag to replace cement in concrete is the decrease in compressive strength. Higher amount of ground manganese slag used to replace cement results in higher compressive strength reduction. A 30% ground manganese slag replacement may lead to concrete compressive strength reduction of 41% at 28 days of curing.

The relationship between the flexural strength with the increase in ground manganese slag in the concrete specimen showcases an identical pattern as the results from compressive strength test, but the reduction in flexural strength is not as substantial. A 30% ground manganese slag replacement may lead to concrete flexural strength reduction of 2.6% at 28 days of curing.

In the concrete specimens containing ground manganese slag, there is a potential risk of releasing inorganic contaminants into the environment through leaching even when exposed to demineralized water. Up to 20% of ground manganese slag replacing cement in concrete, the leaching process through diffusion is slower compared with the process of combined diffusion and dissolution. When 30% of ground manganese slag was used to replace cement, the leaching process through diffusion can be as rapid as combined diffusion and dissolution at the later age of concrete.

Based on the results obtained, 10% of cement may be replaced by ground manganese slag without compromising too much on both compressive and flexural strengths. However, it is not advisable to use ground manganese slag in concrete which is exposed to leachate. On top of these, there is still opportunity for future work of using ground manganese slag as binder replacement – for instance, the influence of particle fineness on mechanical properties of concrete. As for leaching characteristic, future work may focus on the chemical content of the leachate in order to study on heavy metal leaching and how to prevent it. In addition, there exists a prospect for future research involving the utilization of ground manganese slag as a binder replacement, such as investigating the impact of particle fineness on the mechanical properties of concrete. Regarding leaching characteristics, forthcoming studies could delve into the chemical composition of the leachate, specifically examining heavy metal leaching and strategies for its prevention.

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