STRENGTHENING OF FE-RICH INORGANIC POLYMERS IN A GAMMA RADIATION FIELD

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Introduction

Mortars and concretes based on the ordinary Portland cement (OPC) are currently used for shielding and radioactive waste encapsulation worldwide. However, multiple studies[1-3] have already proven the radiation-induced degradation of OPC-based matrices. Changes in compressive strength, changes in average pore diameter, radiation induced carbonation and the formation of micro cracks were observed. Therefore, alternatives to the calcium(alumino)silicate cements are increasingly being studied.

Inorganic Polymers (IP) is one of the alternatives currently being studied intensively.[4] These materials are proven to be highly chemical resistant and highly temperature resistant.[4] Only a few studies investigating the effect of gamma radiation on IP are published.[5-7] Changes in compressive strength, microstructure and pore size distribution of IP were observed and described because of the radiolysis of water, the reorganisation of pore walls and a decrease in the average Si-O-Al bonding angle in IP matrices.[7]

In this study, the effect of gamma radiation is investigated on synthetic plasma slag (PS) based IP as a function of dose rate [0.0016 kGy/h – 2 kGy/h] and total absorbed dose [0.032 kGy – 624 kGy]. The changes in macromechanical behaviour are studied.

Material and methods

A synthetic slag was produced based on urban solid waste incinerated bottom ash, iron ore, limestone and sand. The melt was quenched with water jets and a water tank to obtain a highly amorphous (> 98%) vitrified material.[8] The glass was milled using a ball mill until a Blaine value of $(4.07 \pm 0.04) \cdot 10^3$ cm²/g according to EN 196-6. The milled slag has a density of (3.094 ± 0.009) g/cm³.

The IP pastes were produced by mixing the dry milled glass with an activation solution (SiO_2/Na_2O = 2.0; H_2O/Na_2O = 29.0) in a solid to liquid ratio of 2.6 g/ml. The IP paste was poured in open plastic moulds of (25 x 25 x 20) mm³ and cured at a temperature of (28.8 ± 0.5)°C and a relative humidity of (50 ± 10)%. A microstructure as shown in Figure 1 is obtained (Tabletop TM3000, Hitachi, Krefeld, Germany).

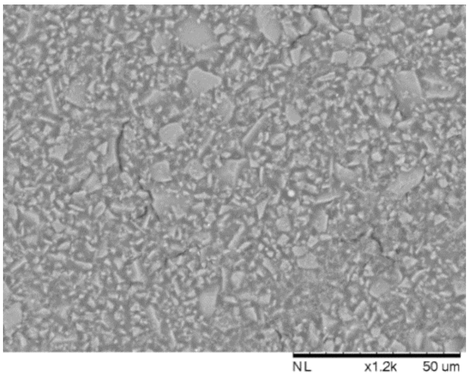

Figure 1: SEM-image of the inorganic polymer

Samples were hardened for 1 hour, 1 day or 28 days prior to irradiation. Table 1 shows the three types of gamma sources used in the irradiation experiments. In all set-ups, samples were irradiated in air.

Table 1: Three types of gamma sources used for irradiation

	Low dose rate	Intermediate dose rate	High dose rate
Source	^{60}Co	^{137}Cs	^{137}Cs
Energy	1.173 MeV, 1.332 MeV	0.662 MeV	0.662 MeV
Dose rate	7.1 ± 0.7 Gy/h 1.6 ± 0.2 Gy/h	152 ± 8 Gy/h	2 kGy/h
Absorbed dose	0.032 kGy – 16 kGy	0.152 kGy – 95 kGy	4.7 kGy – 624 kGy
Time before irradiation	24 h or 28 d	1 h	24 h

Once the irradiation was stopped, the maximum compressive strength at fracture load (NBN EN 12390-3) of the samples was tested using ZWICK Z050 with a compression speed of 1.0 mm/min. The relative change of the irradiated samples relative to the non-irradiated samples was calculated.

Results and discussion

A graphical overview of the results from the compressive strength tests is given in Figure 2. At low dose rates (1.6 Gy/h and 7.1 Gy/h), the irradiated samples show in general a lower compressive strength in comparison to the non-irradiated samples when irradiated up to 3 kGy. Above 3 kGy, an increase in compressive strength was observed. These changes are however not significant due to the large spreading in the results (t-test < 5%). At a dose rate of 152 Gy/h, irradiated 1 hour after casting, a significant decrease in compressive strength was found for the samples which received doses up to 3.6 kGy. For doses higher than 3.6 kGy, an increased strength was observed. The samples irradiated for 7 days at 152 Gy/h showed an increase with (+71 ± 24)%. The reference samples had a compressive strength of (34 ± 7) MPa and the irradiated samples had a strength of (58 ± 4) MPa. For these samples, the short curing period before irradiation could explain the higher gain in strength. Samples irradiated 24 hours after casting with a dose rate of 2 kGy/h show an overall increase in compressive strength.

In general, samples with an absorbed dose above 5 kGy show an increase in strength independent of the irradiation set-up. The observed dose seems to play a more important role than the applied dose rate. Lambertin *et al.*[7] studied metakaolin based IPs and also found an increase of compressive strength by 10% assuming a densification of the IP network. He used a ^{60}Co source with a dose rate of 600 Gy/h until 750 kGy under argon atmosphere.

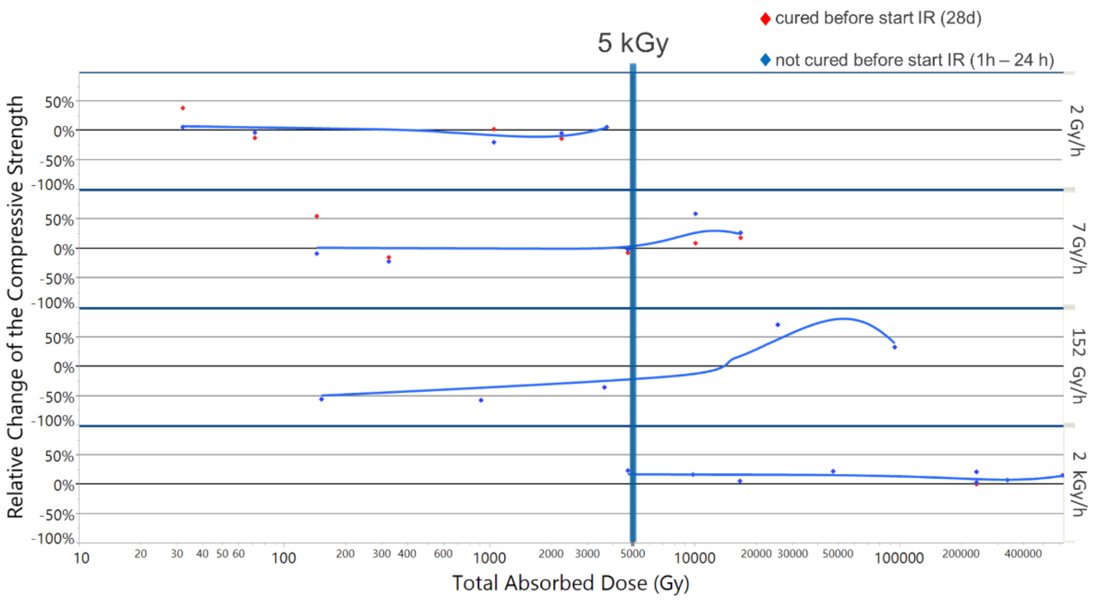

Figure 2: Relative change in compressive strength of the irradiated samples compared to the reference samples in function of total absorbed dose (log scale) sorted per ascending dose rate with 80% confidence interval

It is expected that during irradiation of IP different effects occur simultaneously. Radiolysis of water is expected to play a major role, *e.g.* causing micropores. The hydrogen gas can more easily leave the sample giving rise to a sample with a higher density and a higher strength. The hydroxides may interact with the matrix with the formation of *e.g.* $Ca(OH)_2$ and $Fe(OH)_2$. But radiolysis can also lead to dehydration and formation of microcracks and thus a higher porosity.

Radiation induced carbonation can occur with the formation of *e.g.* $CaCO_3$ and Na_2CO_3, leading to a lower average pore size diameter and a higher strength.[9] Another effect which might occur is radiation induced geopolymerisation causing the formation a better network and thus higher strength for the matrices.

Conclusion

Inorganic polymers based on a synthetic slag were irradiated using gamma sources (^{60}Co and ^{137}Cs) with dose rates varying from 1.6 Gy/h to $2 \cdot 10^3$ Gy/h with total absorbed doses from 32 Gy to 624 kGy. Above 5 kGy, an increased compressive strength was observed for all the irradiated samples compared to the non-irradiated samples. The total absorbed dose seems to be of higher importance than the applied dose rate.

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