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Effect of Water to Geopolymer Solids Ratio on Properties of Fly Ash and Slag-Based One-Part Geopolymer Binders

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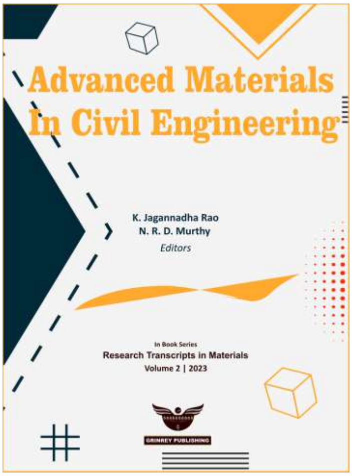

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Abstract

Geopolymer binders are claimed to be a satisfactory substitute for ordinary Portland cement (OPC) nowadays. These binders showcased their impracticalities in the execution part concerning transport, storage, and handling of strong alkaline solutions. This study describes a new "one-part" or "just add water" geopolymer binder which can be made in the same way that OPC is made by dry-mixing solid alumina-silica-rich materials, solid alkali activators, and free water. The flowability and compressive strength of one-part geopolymer binders produced from fly ash and slag were investigated as a function of the water to geopolymer solids (W/GS) ratio in this study. The W/GS ratio was considered as 0.25, 0.30, and 0.35. At each level of W/GS ratio, 25%, 50%, and 75% of the fly ash was substituted with ground granulated blast furnace slag by weight, where the solid activator content was maintained at 12% for all the mixes. The experimental and microstructural observation shows that the 50/50 fly ash/GGBS mixture at 0.30 W/GS had the best flowability and compressive strength, and their microstructure exhibited signs of being dense and compact compared to the other mixtures.

1. Introduction

The greenhouse gas emissions, rising manufacturing costs, and overall unsustainable nature of Ordinary Portland Cement (OPC), required development in new, more ecologically friendly cementitious materials. To produce OPC, a significant extent of carbon dioxide (CO_2) is emitted straight into the air; this accounts for about 7% to 8% of worldwide CO_2 emissions [1]. Cement-less binders like Alkali Activated Materials (AAMs) have been developed by researchers worldwide to solve the problems of cement production's impact on the environment and its use of natural resources, these are also known as Geopolymers (GPs). Compared to the production of Portland cement, geopolymer uses roughly 60% less energy and results in 60-80% fewer CO_2 emissions [2]. Geopolymers are resistant to the intrusion of numerous harmful substances, including chloride and sulphate, and possess exceptional mechanical properties [3]. These benefits have elevated geopolymers to a possible substitute for OPC in the sustainable construction industry.

While geopolymers include several benefits, geopolymer blends can be difficult to create because of their drawbacks and restrictions. Being that geopolymers are two-part systems, their technological utility is limited by the need for liquid alkali activators, which are viscous, caustic, and corrosive. As a result, research into geopolymers that only require water to be added is expanding. Geopolymer precursor is combined with powdered activators (consisting of sodium silicate and sodium hydroxide), followed by the addition of water to produce OPG. The reaction in these one-part blends occurs upon the addition of water to the dry mix, as opposed to the traditional geopolymer systems where heating is required to initiate the reaction [4].

Recently, a new approach, called OPG technology, for creating geopolymer materials has been developed to overcome difficulties with silicate solution activated geopolymers. Askarian et.al [8] examined introduction of slag in the OPG binder and found reduced workability but no compromise in the strength of the OPG binder. However, numerous research has been done to find out how the amount of slag in OPG systems changes the way the mixture works. Regardless, the effect of GGBS addition and water-to-geopolymer solids ratio (W/GS) on the OPG mix properties is rarely reported. As a result, experimental and microstructural characterization investigations have been performed to determine how increasing the slag and water content of OPG mixtures affects their flowability and compressive strength properties. Intent on investigating the impact of slag on the OPG system, fly ash was substituted with GGBS at 25%, 50%, and 75% replacement by weight. The W/GS was, on the other hand, altered in three levels, namely 0.25, 0.30, and 0.35, to investigate the effects of water concentration on OPG mixture flowability and compressive strength. Microstructural characterization methods were also used to investigate the reaction process of OPG hydration products.

2. Materials and methods

2.1 Raw materials

OPG binders were developed in the laboratory by combining aluminosilicate precursors such as fly ash (FA) and ground granulated blast-furnace slag (GGBS). According to ASTM C618 standards, low calcium FA (class-F type) meets the criteria of $SiO_2+Al_2O_3+Fe_2O_3 > 70\%$. The GGBS utilized in this investigation to make OPG mixtures conforms to ASTM C989. The X-ray fluorescence spectroscopic measurements of FA and GGBS chemical compositions are reported in Table 1. The observations of an XRD study of the crystallinity and phase of FA and GGBS are depicted in Figure 1. Crystallinity peaks in the fly ash include quartz, hematite, calcite, and mullite. The amorphous nature of the slag indicates its high level of reactivity. Figure 2 depicts the microstructure of the FA and GGBS acquired using a scanning electron microscope (SEM). The micrographs showed that GGBS has irregular angular grain particles while fly ash has a spherical shape. To produce OPG mixes, anhydrous sodium metasilicate was utilized as a solid activator in powder form. Solid activator appears to be white in color consists less than 5% of chemically bounded water with modulus (SiO_2/Na_2O) of 0.97. Solid activators are utilized to counteract the disadvantages of alkaline activator solutions.

Table 1. Chemical oxide compositions of FA and GGBS

Oxides (% by weight)	**Al_2O_3**	**SiO_2**	**CaO**	**Fe_2O_3**	**MgO**	**SO_3**	**Na_2O**	**K_2O**	**LOI[a]**
FA	28.62	60.65	1.70	3.19	1.84	1.26	1.11	0.11	4.52
GGBS	16.72	32.45	33.75	1.22	9.61	0.85	0.15	0.06	0.40

[a] Loss on ignition

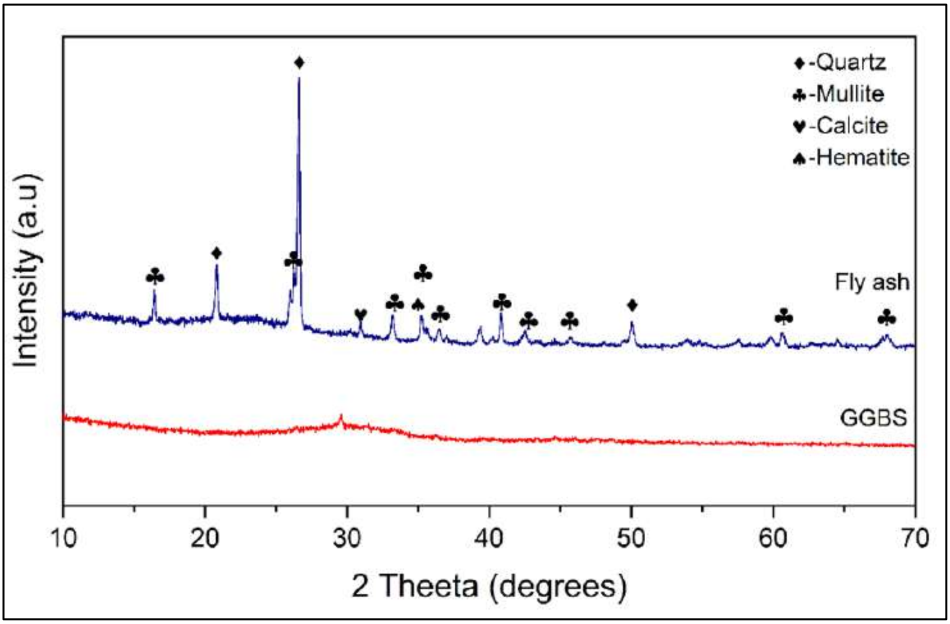

Fig. 1. X-ray diffraction RD analysis of FA and GGBS

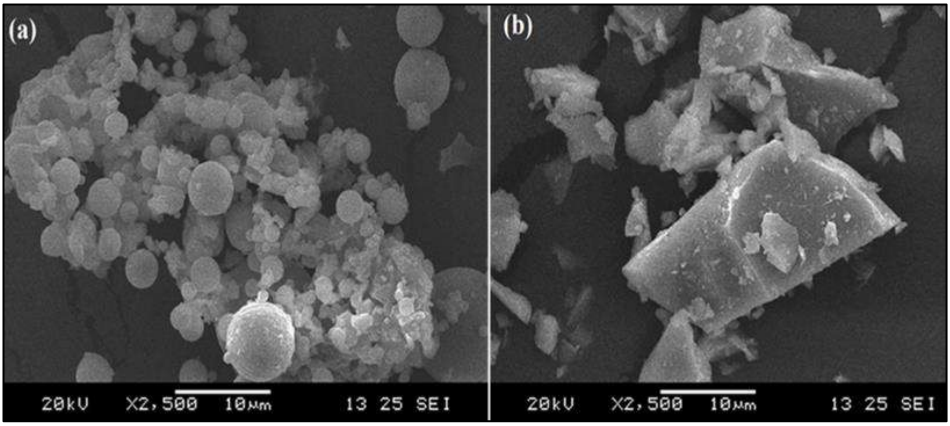

Fig. 2. SEM micrographs: (a) FA and (b) GGBS

2.2 Methodology

OPG binders were made using anhydrous sodium metasilicate powder, GGBS, and low calcium FA in order to conduct laboratory experiments. Consistent OPG blends were formed by manually dry mixing FA/GGBS with a solid sodium metasilicate powder for 2 to 3 minutes. Subsequently, water is introduced into the blends and meticulously mixed for a duration of 3 to 4 minutes at a rotation speed of 65 rpm using a Hobart mixer. Mixing continued until uniformity and consistency were achieved without any remaining discrepancies. The OPG mixtures' flowability was then evaluated using a flow table test calibrated to ASTM C1437 specifications. OPG pastes were poured into 50 mm cubes while still wet in order to examine their compressive strength. The cubes were de-molded and until the testing phases, the samples were left out in the open to cure in the air. The compressive strength of OPG pastes was evaluated using the testing protocols outlined in ASTM C109, both at 7 and 28-day time intervals. The fragments resulting from breakage were gathered to conduct microstructural analyses on OPG paste systems. Microstructure and mineralogy of the OPG paste system were characterized by SEM and X-ray powder diffraction analyses. In this work, the impact of slag and water on the properties of OPG pastes were studied. In order to investigate the impact of slag content, the FA was progressively substituted with GGBS in increments of 25% by weight. This substitution involved replacing

25%, 50%, and 75% of the original FA content with GGBS. The W/GS was also considered in three levels in increment of 0.05 ratio, i.e., 0.25, 0.30, and 0.35. The dosage of activator is taken as 12% for all produced mixes. The mix proportions considered in this work is shown in Table 2. For this investigation, the mixtures were designated as F75G25W0.25, where F75 represents fly ash 75%, G25 stands for GGBS 25%, W0.25 for W/GS ratio as 0.25 for easier identification of the mixes.

Table 2. OPG mix proportions.

Mix ID	FA	GGBS	W/GS	Activator
F75G25W0.25	0.75	0.25	0.25	0.12
F50G50W0.25	0.50	0.50	0.25	0.12
F25G75W0.25	0.25	0.75	0.25	0.12
F75G25W0.30	0.75	0.25	0.30	0.12
F50G50W0.30	0.50	0.50	0.30	0.12
F25G75W0.30	0.25	0.75	0.30	0.12
F75G25W0.35	0.75	0.25	0.35	0.12
F50G50W0.35	0.50	0.50	0.35	0.12
F25G75W0.35	0.25	0.75	0.35	0.12

3. Results and discussions

3.1 Flowability

OPG mix flow values are illustrated in Figure 3. The flow value clearly decreases with increasing slag content and increases with increasing water concentration in the combination. The F75G25W0.35 combination achieved the greatest flow value of 213 mm. Similarly, the mix F25G75W0.25 had the lowest flow value, at 117 mm. It can be examined that, as the GGBS content increases by 25% and 50% in the mix consists of W/GS of 0.25, the flow values decreases by 8% to 10% for every increment of GGBS content. The similar trend in decrement of flow values observed for the mixes consists of water to geopolymers solids ratios of 0.30, and 0.35. This is because, the slag particle's larger explicit surface area and irregular form, and their quick reaction process of slag particles which slows down the flow value of the OPG mixes [5]. As the W/GS content increases by 0.05 and 0.1 in the mix, with irrespective of percentage of GGBS content, the flow value increases by 40% to 55% for every increment of W/GS ratio. This occurs because higher water content results in lower viscosity, leading to improved flowability [6]. The mix F50G50W0.30 has shown the flow value of 172 mm where this slump value is neither less nor more. The results show that the W/GS ratio has the greatest influence on paste flow values, followed by GGBS substitution.

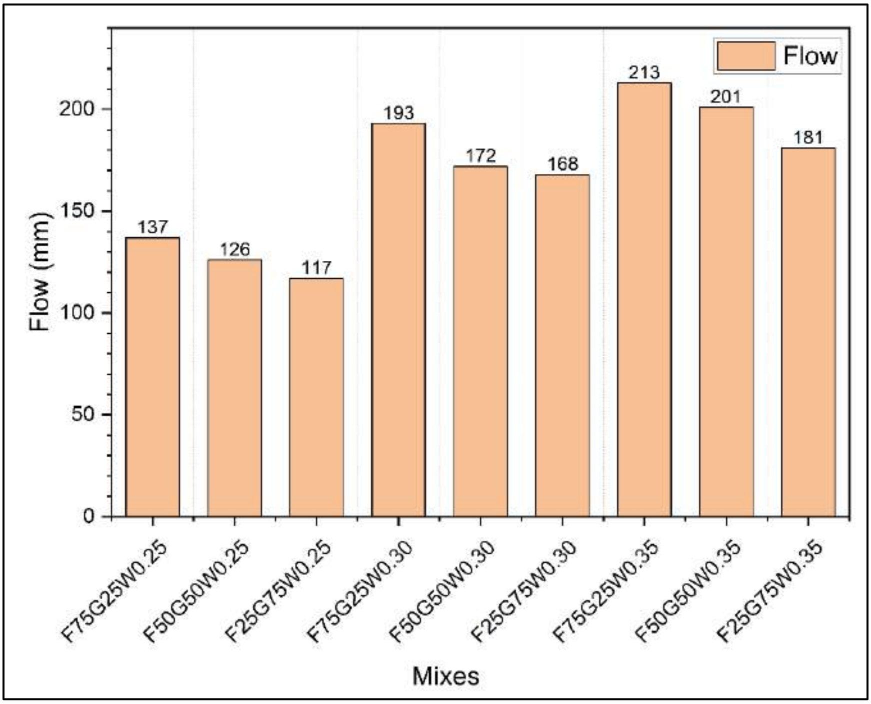

Fig. 3. Flow value of OPG mixes

Figure 4 illustrates the 7 and 28 days compressive strength (CSr) of OPG mixes. As the percentage of slag in OPG mixes is increased, the 7 and 28 days CSrs are found to increase. After 7 days, the CSr of all mixes was roughly 65% that of OPC after 28 days. At 28 days, the OPG CSr of the mixes ranges from 31.75 MPa to 79.48 MPa. Mix F75G25W0.35 had the weakest CSr after 7 days (23.35 MPa) and 28 days (31.75 MPa). It can be examined that, as the GGBS content increases by 25% and 50% in the mix the 28 days CSr increases by 10% to 80% for every increment of GGBS content. This suggests that the GGBS replacement has a considerable impact on the CSr of the OPG paste. This is due to the synthesis of C-A-S-H gel influenced by an alkali activator occurs in tandem with N-A-S-H gel, and the calcium content of the mixture increases when GGBS replacement is increased. Moreover, greater calcium speeds up the polycondensation process, resulting in tetrahedral silica and aluminum structures that eventually form a three-dimensional network with higher CSr [7].

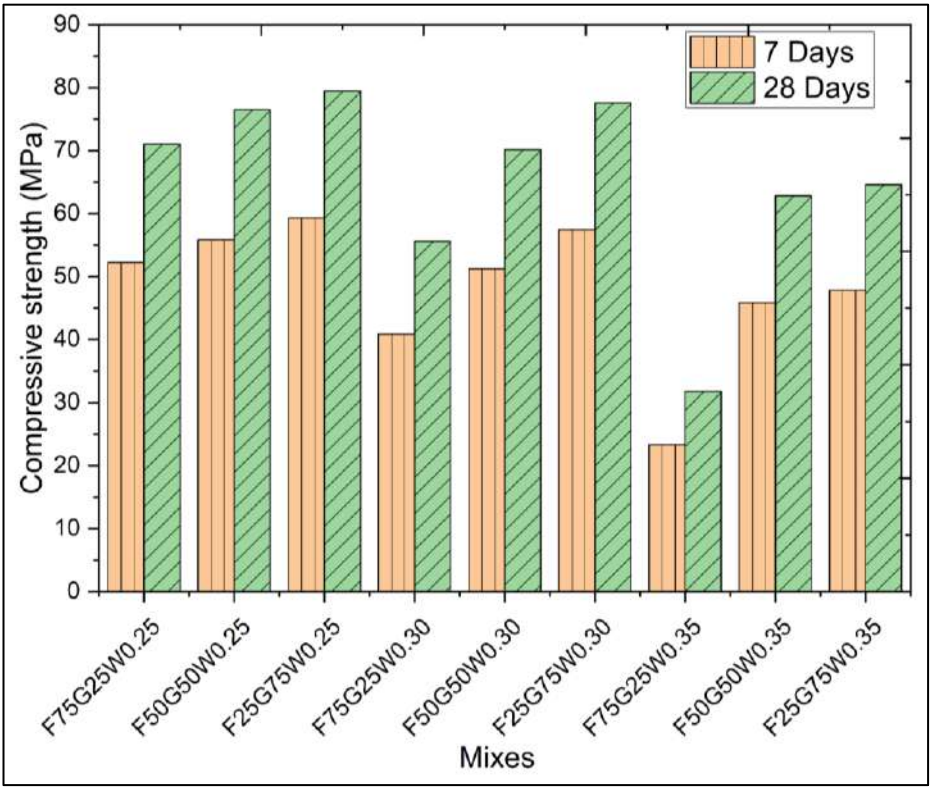

Fig. 4. Compressive strength of OPG mixes

3.2 Compressive strength

To the contrary, as W/GS content increases by 0.05 and 0.1 in the mix, with irrespective of percentage of GGBS content, the 7 and 28 days CSr decreases by about 22% to 56% for every increment of W/GS ratio. This is because the polymerization procedure is affected by the amount of water present in the mixture, which increases as the W/GS ratio rises. The amount of water in one part geopolymer is determined by the quantity necessary to achieve complete activator breakdown; any excess water over this limit results in an increased porosity in strength degradation [8]. The mix F50G50W0.30 has performed with the better 7 and 28 days CSr results along with which had the good flow value of 172 mm.

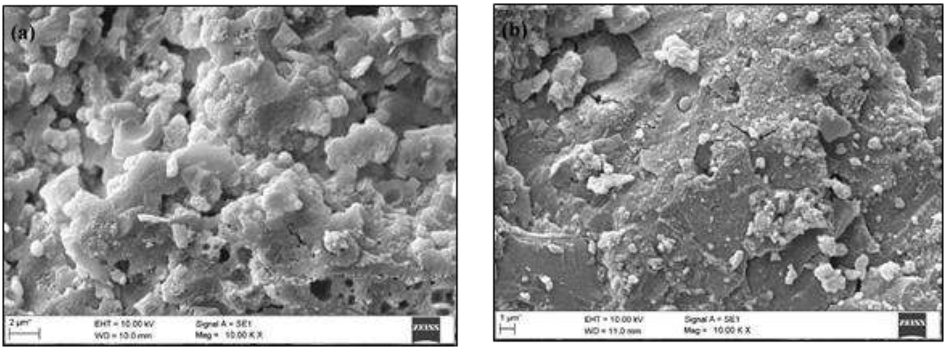

Fig. 5. SEM micrographs of OPG mixes a) F50G50W0.30, and b) F75G25W0.35

3.3 Microstructural studies

Figure 5 displays SEM micrographs for the mixtures taken to examine the impact of the slag and water contents. SEM micrographs were taken from the small chunk of 28 days compressive strength tested specimen. The Figure 5a and 5b shows the micrographs of mixes F50G50W0.30 and F75G25W0.35, respectively. These mixes for the SEM analysis were selected to study the microstructure of best performed mix and to observe the water effect and slag content effect on OPG systems. Increasing the slag content in the combination gives rise to the production of a C-A-S-H gel, as shown in Figure 5a, which has a highly homogenous, dense, and compacted microstructure and is similar to the greater strength growth in the mix [5]. The micrographs show that, along with the geopolymer products, each mixture also contains unreacted FA and GGBS particles in varying concentrations. Due to the irregular dispersion of sodium metasilicate powdered particles, the amount of silica available in the mix varies, preventing a complete formation of C-A-S-H gel. As can be seen in Figure 5b, the compressive strength of the OPG mixture diminishes as the water content increases due to the uneven synthesis of C-A-S-H gel. This is due to uneven mobility of the particles in higher dose water content which results increased porosity leads to strength decrement [9].

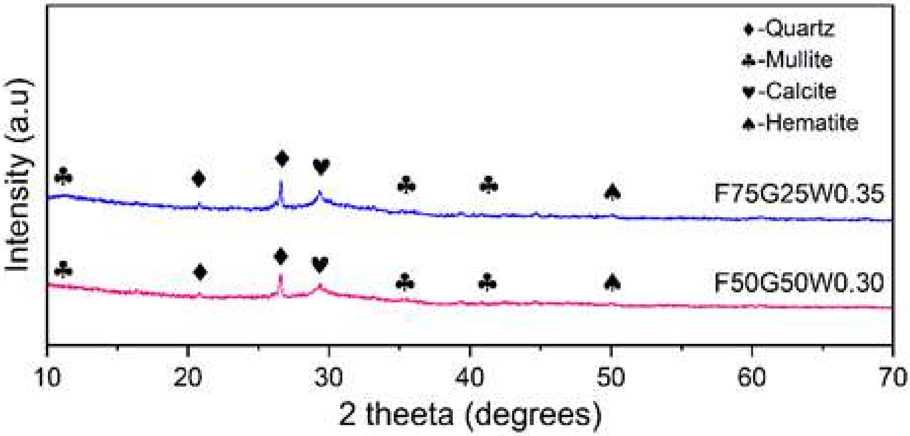

Fig. 6. XRD peaks of OPG mixes F50G50W0.30, and F75G25W0.35

To study mineralogical changes in the system the advanced XRD analysis was carried out. Figure 6 depicts the XRD peaks of OPG mixtures F50G50W0.30, and F75G25W0.35. These mixes were chosen for the XRD examination in order to examine the mineralogy changes, the development of calcium silicate hydrate gel in the best-performing mix, and the impact of water and slag concentration on OPG systems. The peaks were measured between 2θ =10° and 70°. By comparing the crystal phase results from the one-part geopolymer paste to those from the raw ingredients, no new peaks are seen; however, the diffraction pattern magnitude is different. The fact that FA based OPG paste still contains nonreactive minerals like quartz, mullite, and hematite demonstrates that not all aluminosilicates are used as reactants. There is only a statistically significant variation in peak intensity at roughly 29° between the mixtures F50G50W0.30 and F75G25W0.35, both of which are indicative of the existence of C-S-H gel and calcium carbonate phase. The emergence of peaks between $2\theta = 20°$ and 40° indicates that the C-(N)-A-S-H presence which indicate the strength of the paste is increasing [10]. With its smoother curve suggesting less unreacted particles in the solution, the F50G50W0.30 mixture not only has the highest compressive strength but also the densest microstructures compared to the mix F75G25W0.35.

4. Conclusions

This study aimed to investigate the impact of water to geopolymer solid ratio on the flowability and compressive strength characteristics of mixtures containing fly ash and GGBS-based OPG. The following conclusions were drawn based on the experimental and microstructural evidence.

- The higher slag content, along with the specific characteristics of slag particles and the increased CaO concentration within the slag, results in a decrease in flowability.
- As the GGBS content increases, the compressive strength shows improvement because of the higher CaO content in the slag, which speeds up the formation of polymerization reactions.
- When the water content in the mixture increases, the mixture can be worked more easily since it is less viscous.
- Compressive strength drops off rapidly as the amount of water added to the combination increases.
- The OPG mix comprised of 50/50 FA/GGBS for 0.30 w/s ratio and 12% activator performed better than all other mixes with respect to flowability and compressive strength. It exhibits dense and compacted microstructure properties.

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