

The Eurasia Proceedings of Science, Technology, Engineering & Mathematics (EPSTEM), 2022

Volume 21, Pages 388-395

**IConTES 2022: International Conference on Technology, Engineering and Science**

## **Investigation of High Range Water-Reducing Admixture Requirement in Cementitious Systems Containing Fly Ash with Different Utilization Ratio and Fineness**

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**Abstract:** Mineral additives are widely used in cementitious systems to reduce CO<sub>2</sub> emissions. In this context, it was understood that fly ash, an industrial by-product, is the most widely used mineral additive. It is necessary to examine the binder-water reducing admixture compatibility in cementitious systems containing mineral additives. The fineness and utilization ratio of mineral additives seriously affect the compatibility in question. In this study, the effect of the use of fly ash in different utilization ratios and fineness on the flow value and consistency retention performance of mortar mixtures containing water-reducing admixture in different ratios was investigated. For this purpose, 4 different usage dosages of water-reducing admixtures (0%, 1%, 1.5% and 2%), 2 different utilization ratios (0, 15% and 30%) and fine fly ash (4000 and 6000 cm<sup>2</sup>/g) were used. A total of 20 mortar mixtures were prepared. The flow performance of the prepared mixtures for 60 minutes was examined. As expected, the initial and 60th-minute flow values of all mixtures increased with the increase in the use of water-reducing admixture. It was understood that the change of fly ash fineness did not have a significant effect on the initial and 60th-minute flow values of the mixtures. Similarly, it was observed that the change in the utilization ratio of fly ash did not have a great effect on the initial and 60th-minute flow performance in the mixtures with no admixtures, 1% and 2% admixture usage dosages. However, it was determined that the increase in fly ash utilization ratio in mixtures with an admixture usage dosage of 1.5% increased the initial and 60th-minute flow values of the mortar mixtures. Also, it was observed that mortar mixtures containing 2% water-reducing admixture generally have the highest consistency retention capacity (95-100%), regardless of fly ash fineness and utilization ratio.

**Keywords:** Admixture usage dosage, Fly ash fineness and utilization ratio, Flow value, Consistency retention capacity

### **Introduction**

During cement production, 8% of the total CO<sub>2</sub> emitted in the world is formed. In addition to reducing the carbon footprint, the use of mineral additives in cementitious systems has become inevitable in order to expand sustainability (Mardani-Aghabaglou, 2016; Al-Kutti et al., 2018; Ozen et al., 2022a; Sahin & Mardani, 2022a). In this context, mineral additives such as blast furnace slag, silica fume, metakaolin, rice husk ash, trass and fly ash are used in cementitious systems (Biricik et al., 2022a; Mardani-Aghabaglou et al., 2019; Sezer et al., 2016; Yuksel et al., 2016; Bayqra et al., 2022). It was understood that the fineness values of mineral additives resulting from industrial production can vary in a wide range (Win et al., 2022). In this context, it was reported

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by various researchers that the effect of mineral admixture fineness on the fresh and hardened properties of cementitious systems should be investigated (Aytekin et al., 2022; Biricik et al., 2022b). Some studies on the subject are summarized. It was reported that the compressive strength and carbonation resistance of the concrete mixture increase with the increase in the fineness of the blast furnace slag (Rivera et al., 2021). However, it was reported that with the increase of the fineness of the fly ash, the heat of hydration increased (Han et al., 2019), interfacial transition zone (ITZ) (Fanghui et al., 2015), compressive strength (Arel and Shaikh, 2018), porosity (Hsu et al., 2019) and sulfate resistance (Chindaprasirt et al., 2004) improved in concrete mixtures. It was stated that this is due to the increase in the physical filling effect and the pozzolanic reaction with the increase in fineness (Han et al., 2019). However, in another study by Hsu et al (2018), it was stated that the pozzolanic reactivity is limited due to the increase in the risk of flocculation with the increase of fly ash fineness. It was stated that the heat of hydration decreases with the increase of fly ash content, and its fluidity increases due to the ball-bearing effect and smooth surface texture (Sahin & Mardani, 2022b; Moghaddam et al., 2019; Biricik and Mardani, 2022c). However, it was determined that fly ash substitution negatively affects the fluidity of the mixtures (Nguyen et al., 2019).

In addition, the presence of these fines in the system directly affects the adsorption properties of the water-reducing admixture (Mardani et al., 2017). It was declared by various researchers that the need for water-reducing admixtures will increase depending on the increase in the total surface area with the increase in the amount of fine material in the system (Faltin, 2022). However, Mardani et al. (2017) and Karakuzu et al. (2021) reported that the water-reducing admixture can adsorb more strongly to fine particles. In addition, Burgos et al. (2012) stated that the substitution of mineral additive with a higher specific surface area compared to cement causes extra surface formation leading to competition with cement for adsorption of PCE. Palacios et al. (2009) stated that the need for admixtures decreased with the substitution of blast furnace slag in the paste mixture. Yingliang et al. (2020), on the other hand, emphasized that ultrafine blast furnace slag negatively affected the fluidity of mortar mixes, but this negative effect decreased with the use of PCE. It was declared that the amount of water-reducing admixture adsorbed on cement paste containing fly ash depends on the utilization ratio of fly ash, its chemical properties, fineness and density. It was emphasized that the PCE requirement decreased with the use of fly ash (Ng & Justnes, 2016). Similar results were found by Altun et al. (2021) for mixtures containing up to 30% fly ash.

It was understood from the literature that various studies were conducted on the effect of fly ash usage rate and fineness on adsorption properties, fluidity and time-dependent behavior. However, contradictory results were found due to the large number of active parameters and the increase in fineness and the existence of 2 different mechanisms. In this study, the effect of the use of water-reducing admixture on the time-dependent flowability of mortar mixtures containing fly ash at different utilization ratios and fineness was investigated.

## Material and Method

### Material

Within the scope of the study, CEM I 42.5R type portland cement (PC) produced by OYAK Cement and fly ash supplied from Orhaneli Thermal Power Plant were used as binders. The chemical component, physical and mechanical properties of cement and fly ash supplied by the manufacturer are given in Tables 1 and 2, respectively.

Table 1. Chemical composition, physical and mechanical properties of cement

Chemical Properties		Physical Properties		
Oxide	(%)	Specific gravity		3.15
SiO <sub>2</sub>	18.74	Blaine specific surface area	cm <sup>2</sup> /g	3600
Al <sub>2</sub> O <sub>3</sub>	5.37	Fineness	45 μ %	7.4
Fe <sub>2</sub> O <sub>3</sub>	3.04		90 μ %	0.4
CaO	64.11	Setting time	min	180
MgO	1.21	Volume expansion	mm	1
SO <sub>3</sub>	2.68	Mechanical Properties		
Na <sub>2</sub> O	0.34	Compressive strength	1-Day (MPa)	28.8
K <sub>2</sub> O	0.62		28-Day (MPa)	56
Cl <sup>-</sup>	0.038			
F.CaO	2.12			
LOI	3.6			

Table 2. Chemical and physical composition of fly ash

Chemical properties		Physical properties	
Oxide	%		
SiO <sub>2</sub>	59.22	Specific gravity	2.31
Al <sub>2</sub> O <sub>3</sub>	22.86	Residual on 0.045 mm sieve (%)	10
Fe <sub>2</sub> O <sub>3</sub>	6.31	Blaine Fineness (cm <sup>2</sup> /g)	4000
CaO	3.09	Mechanical Properties	
MgO	1.31	Pozzolanic Activity Index (%)	7-Day 85.9
SO <sub>3</sub>	0.17		28-Day 100.7
Na <sub>2</sub> O+0.658 K <sub>2</sub> O	1.4		90-Day 110.2
Cl <sup>-</sup>	0.001		
IR	0.32		
LOI	3.2		
Free CaO	0.00		
Class	F		

In order to examine the effect of fly ash fineness on the performance of mortar mixtures, fly ash with a Blaine fineness of 4000 cm<sup>2</sup>/g was ground to 6000 cm<sup>2</sup>/g fineness by means of a laboratory ball mill. Crushed limestone sand with 0-2 mm grain size, water absorption capacity and specific gravity of 0.40 and 2.6%, respectively, was used in the production of mortar mixes. Some properties of the polycarboxylate ether based high water reducing admixture (HRWRA) used in the study are given in Table 3.

Table 3. Some properties of HRWRA

Type	Density (g/cm <sup>3</sup> )	Solid Content (%)	pH	Chloride content (%)	Alkaline content, Na <sub>2</sub> O (%)
Polycarboxylate ether-based	1.060	32	2-5	<0.1	<10

### Preparation of Mixtures and Experiments

The mortar mixtures prepared within the scope of the study were produced according to ASTM C109 (2013) Standard. The ratio of water/binder and sand/binder in all mixtures was kept constant as 0.485 and 2.75, respectively.

Table 4. Material proportions of mixtures

Mixture	Cement (g)	Fly ash (g)	Water (g)	Water/bin der	Sand (g)	HRWRA (weight of binder %)
FA0_0%						0
FA0_1%						1
FA0_1.5%	500	0	242.5	0.485	1375	1.5
FA0_2%						2
FA4K_15_0%						0
FA4K_15_1%						1
FA4K_15_1.5%	425	75	242.5	0.485	1375	1.5
FA4K_15_2%						2
FA4K_30_0%						0
FA4K_30_1%						1
FA4K_30_1.5%	350	150	242.5	0.485	1375	1.5
FA4K_30_2%						2
FA6K_15_0%						0
FA6K_15_1%						1
FA6K_15_1.5%	425	75	242.5	0.485	1375	1.5
FA6K_15_2%						2
FA6K_30_0%						0
FA6K_30_1%						1
FA6K_30_1.5%	350	150	242.5	0.485	1375	1.5
FA6K_30_2%						2

HRWRA: High range water-reducing admixture

In addition to the control mixture that does not contain fly ash, a total of 5 series of mortar mixtures were prepared by replacing the fly ash with two different Blaine fineness values, 4000 and 6000 cm<sup>2</sup>/g, with cement at the rates of 15% and 30% by weight. A total of 20 mixtures were obtained by adding HRWRA at 3 different usage dosages rates, 1%, 1.5% and 2%. The amount of material used in the production of mortar mixtures is shown in Table 4. The denotation of the mixtures was made according to fly ash fineness, utilization ratio and HRWRA dosage. For example, the mixture containing 15% fly ash with 4000 cm<sup>2</sup>/g Blaine fineness and adding 1% HRWRA admixture was named FA4K\_15\_1%. Flow values of mortar mixtures were determined in accordance with ASTM C1437 Standard. In addition, the flow test was repeated in the mixture, which was kept for 60 minutes, in order to examine the time-dependent behavior of the mixtures.

## Results and Discussion

The time-dependent flow values of the mortar mixtures are shown in Table 5. Not surprisingly, the flow values of the mixtures increased as the HRWRA usage dosage increased, regardless of the fly ash fineness and utilization ratio. This situation was caused by the electrostatic effect and the steric hindrance mechanism of the HRWRA. It was reported by Altun et al. (2020) that the structure of HRWRA consists of carboxylate functional groups and polyethylene side chains. The electrostatic effect occurs when the carboxylate groups of the HRWRA adhere to the cement surface. (Karakuzu et al., 2021; Ozen et al., 2020a; Yigit et al., 2020; Sahin et al., 2022c; Kalipcilar et al., 2016; Latifi et al., 2022; Sahin et al., 2020). At the same time, polyethylene oxide side chains provide steric hindrance (Altun et al., 2021; Ozen et al., 2021; Sahin & Mardani., 2022d; Mardani-Aghabaglou, 2021a; Ozen et al., 2020b; Ozen et al., 2022b; Mardani-Aghabaglou, 2021b).

It was understood that the change in fly ash fineness did not have a significant effect on the flow value and consistency retention capacity of the mortar mixtures. However, according to the literature, it is understood that the workability of the mixtures generally increases with the increase of fly ash fineness (Snellings et al., 2019). It was stated by Chindapasirt et al. (2004) that this may be due to the fact that fine fly ash has a smoother surface compared to coarse fly ash. However, it was emphasized by Hsu et al. (2018) that the use of fly ash with high fineness (6300 cm<sup>2</sup>/g) in the cement mortar mixture negatively affects the workability of the mixture. However, in this study, it was determined that the mixture with the highest flow value was FA6K\_15\_2% containing fly ash with higher fineness value. It was thought that this situation is due to the decrease in the water requirement of the mixture as a result of the increase in sphericity due to the increase in the fineness of the fly ash (De Maeijer et al., 2020).

Table 5. Time-dependent flow values of mortar mixtures

Mixtures	Time-dependent flow values (mm)	
	0 min.	60 min.
FA0_0%	80	75
FA0_1%	175	150
FA0_1.5%	240	185
FA0_2%	300	300
FA4K_15_0%	70	70
FA4K_15_1%	175	160
FA4K_15_1.5%	280	185
FA4K_15_2%	280	280
FA4K_30_0%	75	75
FA4K_30_1%	175	160
FA4K_30_1.5%	280	197.5
FA4K_30_2%	300	290
FA6K_15_0%	80	70
FA6K_15_1%	170	155
FA6K_15_1.5%	250	187.5
FA6K_15_2%	305	290
FA6K_30_0%	70	70
FA6K_30_1%	180	167.5
FA6K_30_1.5%	270	195
FA6K_30_2%	300	300

Regardless of the fly ash fineness, it was determined that the flow value and the consistency retention capacity were not significantly affected by the increase in the fly ash utilization ratio in all the other mixtures, except for the mixture containing 1.5% HRWRA. However, it was found that the flow value increased with the increase of the fly ash content in the mortar mixtures containing 1.5% HRWRA. It was stated by Park et al. (2021) that this situation resulted from the decrease in the reaction degree of cement with the increase in fly ash utilization ratio. Similarly, in another study by Altun, (2021), it was emphasized that the flow value of the mixtures increased with the increase in the fly ash utilization ratio. At the end of 60 minutes, it was determined that the mixtures containing 2% HRWRA had the highest consistency retention capacity, regardless of fly ash fineness and utilization ratio. It was stated by the researchers that the adsorption of the negatively charged carboxylate molecules on the surface of the fly ash particles is quite low since the fly ash surface is negatively charged (Altun, 2021; Wang et al., 2021; Cangialosi et al., 2009). Thus, the cement surface is covered with more water-reducing admixture molecules and the consistency retention capacity of the mixtures increases (Wang et al., 2021).

## **Conclusion**

Within the scope of the study, the results obtained in line with the materials used and the tests applied are listed below:

- Regardless of fly ash fineness and utilization ratio;
  - It was observed that the flow values of the mixtures increased with the increase of the high range water-reducing admixture usage dosage.
  - It was determined that the highest consistency retention performance is in the mixtures containing 2% high range water-reducing admixture.
- The change in fly ash fineness had no significant effect on the flow value and consistency retention performance of the mortar mixtures.
- It was understood that the flow value of the mortar mixtures containing 1.5% high range water-reducing admixture increased with the increase of the fly ash utilization ratio.
- In terms of flow performance, it was determined that FA6K\_15\_2% mixture having 2% high range water-reducing admixture and 15% fly ash with 6000 cm<sup>2</sup>/g fineness was the best mixture.

## **Scientific Ethics Declaration**

The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

## **Acknowledgements**

\* This article was presented as an oral presentation at the International Conference on Technology, Engineering and Science ( [www.icontes.net](http://www.icontes.net) ) held in Antalya/Turkey on November 16-19, 2022.

\*The first author would like to acknowledge the scholarship provided by the Bursa Uludag University Science and Technology Centre (BAP) under grant number FAY-2021-579 Besides, the first author would like to acknowledge the scholarship provided by the Scientific and Technological Research Council of Turkey (TUBITAK) under grant number 219M425 and 2211-A program during her Ph.D. study. In addition, the authors would like to acknowledge Polisan Construction Chemicals Company, Oyak Factory-Bolu Quality Control Laboratory and Orhaneli Technical Power Plant for their kind assistance in providing the high range water reducing admixture, cement and fly ash as well as determining the technical properties of these products, respectively.

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**To cite this article:**

Sahin, H.G., Biricik, O., & Mardani, A. (2022). Investigation of high range water-reducing admixture requirement in cementitious systems containing fly ash with different utilization ratio and fineness. *The Eurasia Proceedings of Science, Technology, Engineering & Mathematics (EPSTEM)*, 21, 388-395.