Influence of black liquor on concrete performance: a sustainable approach

Nasser A. M. BARAKAT ▪ Chemical Engineering Department, Minia University, Minia, Egypt
▪ nasbarakat@mu.edu.eg
Mamdouh M. NASSAR ▪ Chemical Engineering Department, Minia University, Minia, Egypt
Taha E. FARRAG ▪ Chemical Engineering Department, Port Said University, Port Said, Egypt
Hamdy A. A. MOHAMED ▪ Qena Paper Company, Qena, Egypt
Mohamed S. MAHMOUD ▪ University of Technology and Applied Sciences-Suhar, College of engineering and technology, Department of Engineering, Sultanate of Oman
▪ mohamed.mohamed@utas.edu.om

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Abstract

The integration of industrial by-products into concrete mixtures has gained significant attention as a sustainable approach to improving material performance while reducing environmental impact. This study investigates the effects of black liquor (BL), a lignin-rich by-product of the paper industry, as a novel admixture in concrete. Concrete samples with varying BL concentrations (0–4 wt.%) were prepared and evaluated for mechanical strength, workability, density, porosity, and microstructural modifications. The results indicate that the incorporation of 2 wt.% BL optimally enhances concrete properties, yielding a 25% increase in compressive strength (from 40 MPa to 50 MPa) at 28 days. Furthermore, splitting tensile and flexural tensile strengths peaked at 3.6 MPa and 15 MPa, respectively, at this concentration. Workability improved significantly, as evidenced by a 200% increase in slump value compared to BL-free concrete. Additionally, bulk density reached its maximum at 2.28 kg/L, while apparent porosity exhibited a notable decline to 9%, indicating matrix densification. Scanning electron microscopy (SEM) confirmed the refinement of pore structure and enhanced cementitious bonding at 2 wt.% BL. However, excessive BL content (≥3 wt.%) led to reduced performance due to increased porosity and disruption of cement hydration. These findings highlight the potential of black liquor as an effective and sustainable concrete admixture, offering enhanced mechanical properties and improved durability while promoting industrial waste reutilization.

Keywords: Black liquor; Sustainable concrete; Mechanical properties; Microstructure; Porosity reduction
Kulcsszavak: fekete lúg, fenntartható beton, mechanikai tulajdonságok, mikrostruktúra, porozitáscsökkenés

Nasser A. M. BARAKAT
Nasser A. M. Barakat is a renowned Professor of Chemical Engineering at Minia University, Egypt. He earned his Ph.D. in Chemometrics from Hunan University, China, and has held academic positions at Chonbuk National University in South Korea. His research focuses on advanced materials, including electrospun nanofibers, energy applications, and water treatment technologies. Professor Barakat has published extensively, with significant contributions to the fields of nanotechnology and environmental engineering. His work has garnered numerous citations and awards, reflecting his impact and leadership in chemical engineering

Mamdouh M. NASSAR
Professor Emeritus of Chemical Engineering at Minia University, Egypt. He earned his Ph.D. in Chemical Engineering from the Technical University of Norway in 1975, following a Diploma in Pulping Technology and an M.Sc. from Alexandria University. With a career span of over five decades, Professor Nassar has held various academic and administrative positions, including Vice Dean of Student Affairs and Head of the Chemical Engineering Department at Minia University. His research focuses on mass transfer operations, adsorption engineering, and effluent treatment, contributing significantly to the field through numerous publications and collaborations

Taha E. FARRAG
Is a distinguished Professor of Chemical Engineering at Port Said University, Egypt. He currently serves as the Dean of the Faculty of Engineering. With a robust academic background and extensive research experience, Professor Farrag specializes in transport phenomena and the application of mass transfer techniques for wastewater treatment. His scholarly contributions include numerous publications on topics such as adsorption processes and water desalination. Professor Farrag's work has significantly advanced the field of chemical engineering, earning him recognition and citations in various academic circles

Hamdy A. A. MOHAMED
Ph.D. student at Department of Chemical Engineering, Minia University, El Minia, Egypt. Senior head of operation section at Qena Paper industry, Qena, Egypt. Member, Egyptian Society of Engineers. His expertise in industrial operations and commitment to excellence have played a crucial role in maintaining the high standards of Quena Paper Factory, one of the leading manufacturers of printing and writing paper in the region

Mohamed S. MAHMOUD
Associate Professor at the Chemical Engineering Department of Minia University, Egypt. He also serves as an Assistant Professor at the University of Technology and Applied Sciences (UTAS) in Suhar, Oman, where he is the Head of the Scientific Research Department. With a strong academic background and extensive research experience, Dr. Mahmoud specializes in areas such as high temperature reactions, wastewater treatment, nanotechnology, green hydrogen production and carbon. His contributions to the field are reflected in numerous publications and his leadership roles in both academic institutions

1. Introduction

Concrete is the most widely used construction material globally due to its versatility, durability, and cost-effectiveness. However, the environmental impact of concrete production, primarily attributed to the carbon emissions associated with cement manufacturing, has driven researchers to explore sustainable alternatives and admixtures. One such approach is the incorporation of industrial by-products to improve concrete properties while minimizing environmental harm. Among these, black liquor (BL), a by-product of the paper industry, has gained attention for its potential as a sustainable admixture.

The global production of cement, a critical component of concrete, accounts for approximately 8% of total carbon dioxide emissions annually [1, 2]. Efforts to mitigate this environmental footprint have focused on reducing clinker content, utilizing supplementary cementitious materials (SCMs), and incorporating waste products. Black liquor, which is rich in lignin and organic compounds, presents an opportunity to address these challenges [3]. It is generated in large quantities during the kraft pulping process, with global production exceeding 60 million tons per year [4]. Without proper utilization, black liquor poses significant disposal and environmental issues, including water pollution and toxicity [5].

The chemical composition of black liquor makes it a promising candidate for concrete admixture. It contains lignin, hemicellulose, and other organic compounds, which exhibit pozzolanic activity and can influence the hydration process of cement [6, 7]. Previous studies have demonstrated that lignin-based materials can enhance workability, reduce water demand, and improve the durability of concrete [8, 9]. Additionally, black liquor's ability to act as a retarder has been reported to extend setting times, providing greater flexibility during construction [10].

The incorporation of black liquor in concrete has been shown to improve mechanical properties under optimal conditions. For example, Kemal *et al* [11] observed that organic compounds in black liquor interact with calcium hydroxide (CH) to form additional calcium silicate hydrate (C-S-H) gels, which densify the matrix and enhance strength. Hassan *et al* [12] reported that admixtures derived from lignin improve the compressive strength and reduce porosity, which are critical for durability. However, excessive black liquor content can lead to increased porosity and reduced strength due to the disruption of hydration and bonding [13].

Utilizing black liquor in concrete not only enhances its properties but also aligns with the principles of sustainable development. By repurposing a waste product, this approach reduces the reliance on synthetic admixtures and minimizes industrial waste [14]. Additionally, it supports circular economy practices in the paper industry by creating value from by-products [15]. Previous research has highlighted the potential of black liquor to significantly reduce the carbon footprint of construction materials while addressing waste management issues [16, 17].

While several studies have explored the use of lignin and its derivatives in concrete, the specific effects of black liquor on mechanical properties, microstructure, and setting times remain underexplored. This study aims to fill this gap by systematically investigating the impact of varying black liquor contents (0–4 wt.%) on concrete properties. By integrating experimental findings with microstructural insights, this study seeks to establish black liquor as a viable and sustainable admixture for concrete production. The results have implications for both the construction and paper industries, offering a pathway toward more sustainable and high-performance.

2. Materials and methods

2.1 Materials

The primary materials used in this study include Ordinary Portland Cement (OPC), natural sand, crushed gravel, and black liquor (BL).

- Cement: OPC (Grade 42.5) was used as the binder, conforming to ASTM C150 standards.
- Sand: Natural river sand with a fineness modulus of 2.6 was used as fine aggregate.
- Gravel: Crushed gravel with a maximum size of 20 mm was employed as coarse aggregate.
- Black Liquor: BL was obtained from Qena Paper Industry Company, Quse, Egypt. Its chemical composition includes lignin, hemicellulose, and inorganic compounds, contributing to its pozzolanic activity.
- Water: Potable water was used for mixing and curing.

2.2 Sample preparation

Concrete mixes were prepared with varying BL contents of 0%, 1%, 2%, 3%, and 4% by weight of cement. Three different concrete samples (S1, S2, S3) were formulated based on *Table 1*:

Sample code	Ingredients			
	Cement (kg/m³)	Gravel (kg/m³)	Sand (kg/m³)	Water/cement ratio
S1	360	1410	705	0.47
S2	380	1385	692	0.47
S3	400	1360	680	0.47

Table 1 Prepared samples composition
1. táblázat Az előkészített minták összetétele

The mixing process followed ASTM C192, involving the sequential addition of aggregates, cement, water, and BL to ensure uniform distribution.

2.3 Testing methods

2.3.1 Mechanical properties

- *Compressive strength:* Tested according to ASTM C39 using a 2000 kN capacity compression testing machine. Specimens were cured for 1, 3, 7, 14, 28, and 90 days, with results recorded as the average of three specimens.
- *Splitting tensile strength:* Measured using ASTM C496 standards on cylindrical specimens. The load was applied diametrically using a 1000 kN capacity testing machine.
- *Flexural strength:* Conducted on prismatic beams following ASTM C78 standards. A three-point loading system was applied.

2.3.2 Durability and physical properties

- *Bulk density:* Determined by dividing the dry weight of specimens by their volume. Measured for each curing period.
- *Apparent porosity:* Evaluated using Archimedes' principle by measuring the saturated and dry weights of the specimens.
- *Initial and final setting times:* Assessed using Vicat apparatus per ASTM C191. BL's impact on hydration dynamics was observed for each mix.

2.3.3 Workability (Slump Test)

Slump value was measured using the ASTM C143 standard slump cone test. The effect of BL content (0%, 1%, 2%, 3%, 4%) on workability was evaluated by determining the slump value for each mix. Measurements were recorded to the nearest millimeter, highlighting changes in flowability with varying BL concentrations.

The slump test was conducted following ASTM C143 standards. The procedure is as follows:

1. *Equipment:* A standard slump cone (300 mm in height, 200 mm bottom diameter, 100 mm top diameter), a tamping rod, and a flat base plate were used.

2. *Preparation:* The slump cone was placed on the flat base plate, and the internal surface was lightly oiled to prevent sticking.
3. *Filling:* The cone was filled with concrete in three layers, each approximately one-third of the cone's height.
4. *Compaction*: Each layer was compacted using 25 strokes of the tamping rod, uniformly distributed across the surface.
5. *Leveling:* After the third layer, the excess concrete was struck off to level the surface with the top of the cone.
6. *Lifting the Cone:* The cone was lifted vertically and steadily within 5–10 seconds to avoid lateral displacement of the concrete.
7. *Measurement:* The slump value was determined by measuring the vertical displacement (difference in height) between the top of the slump cone and the highest point of the slumped concrete.

2.3.3 Microstructural analysis

Scanning Electron Microscopy (SEM) images were captured using a JEOL JSM-6510LV microscope. Samples from 0% and 2% BL mixes were analyzed to evaluate the microstructural changes.

All tests were conducted under controlled laboratory conditions, with an ambient temperature of 25 ± 2°C and relative humidity of 50%. Each test was repeated three times to ensure reproducibility and accuracy. Data were statistically analyzed using ANOVA to determine the significance of the observed differences among the mixes. A significance level of 0.05 was considered for all analyses.

3. Results and discussion

Fig. 1 shows the influence of black liquor content on the compression strength of the prepared concrete mixtures. As shown, in general, the addition of black liquor (BL) at different concentrations (0–4 wt.%) significantly affects the compressive strength of concrete samples (S1, S2, S3) over time (1, 3, 7, 14, and 28 days). Across all samples:

- The compressive strength increases with curing time, reaching its maximum at 28 days.
- Optimal black liquor content lies at 2 wt.%, where the compressive strength is at its peak. Beyond this concentration, the strength diminishes. Numerically, the determined compression strengths at 2 wt.% BL after 28 days aging time were 46, 46.8 and 48.8 MPa for S1, S2 and S3 samples, respectively.

Black liquor contains organic compounds and lignin, which may act as retarders or pozzolanic additives, influencing hydration, setting time, and strength development. The observed effects can be attributed to the pozzolanic activity. At optimal concentrations (2 wt.%), black liquor contributes to secondary hydration reactions, forming additional calcium silicate hydrates (C-S-H), which enhance strength. Studies confirm that lignin and other organic substances in black liquor react with calcium hydroxide to produce C-S-H gels [7]. The addition of black liquor improves the workability of concrete, potentially resulting in better compaction and reduced porosity. This is in line with research on admixtures derived from lignin-based substances [8]. Excessive black liquor (3–4 wt.%) likely introduces too much organic matter, which interferes with the cement hydration process, increases porosity, and weakens the concrete matrix [18].

Comparison of Samples (S1, S2, S3)

- *S1 (Low cement content; Fig. 1A):* Shows lower compressive strength overall. The addition of black liquor is beneficial but limited by the lower cement content, reducing the extent of hydration and pozzolanic reactions.
- *S2 (Moderate cement content; Fig. 1B):* Exhibits higher strength compared to S1 due to increased cement content, which supports more extensive hydration and secondary reactions facilitated by black liquor.
- *S3 (High cement content; Fig. 1C)*: Achieves the highest compressive strength among the samples. The improved hydration and densified matrix due to the interaction of black liquor with the high cement content explain its performance.

The strength development over time aligns with the typical hydration process. For instance, at 1 day, hydration is minimal, and black liquor primarily acts as a plasticizer, slightly improving strength. By 7 and 14 days, secondary hydration becomes more pronounced, and black liquor's pozzolanic activity maximizes strength. At 28 days, the matrix becomes denser, and the strength stabilizes, highlighting the long-term effects of black liquor.

In conclusion, using black liquor at 2 wt.% offers a sustainable way to enhance concrete properties, leveraging waste materials while reducing cement usage. Incorporating black liquor reduces industrial waste and promotes eco-friendly construction practices. However, the performance is sensitive to the dosage of black liquor and the initial composition of the concrete mix.

Fig. 2 shows the influence of aging time on the compression strength at the optimum black liquor content (2 wt.%). The data for sample S1 (as a model, the other two samples show similar behavior (data are not shown). As shown in the figure, the compressive strength increases significantly within the first 28 days, stabilizing thereafter with only a slight increase observed at 90 days: 43.5 and 46 MPa after 28 and 90 days, respectively. This pattern aligns with the general hydration process of cementitious materials, where the rate of hydration slows over time as the reaction nears completion.

The observed results can be attributed to the interplay between hydration, pozzolanic activity, and the role of black liquor. During the initial 28 days, the hydration of cement generates calcium silicate hydrates (C-S-H) and calcium hydroxide (CH), which are the primary contributors to strength development. Black liquor at 2 wt.% optimally enhances the formation of dense C-S-H gel, filling pores and reducing micro-cracks [6].

Black liquor introduces lignin and other organic compounds that act as secondary reactants. These compounds react with CH to form additional C-S-H gels, especially prominent in the first 28 days. This aligns with studies showing lignin-based materials' capability to improve strength through enhanced pozzolanic reactions [9]. Beyond 28 days, most CH is consumed, leading to a plateau in compressive strength.

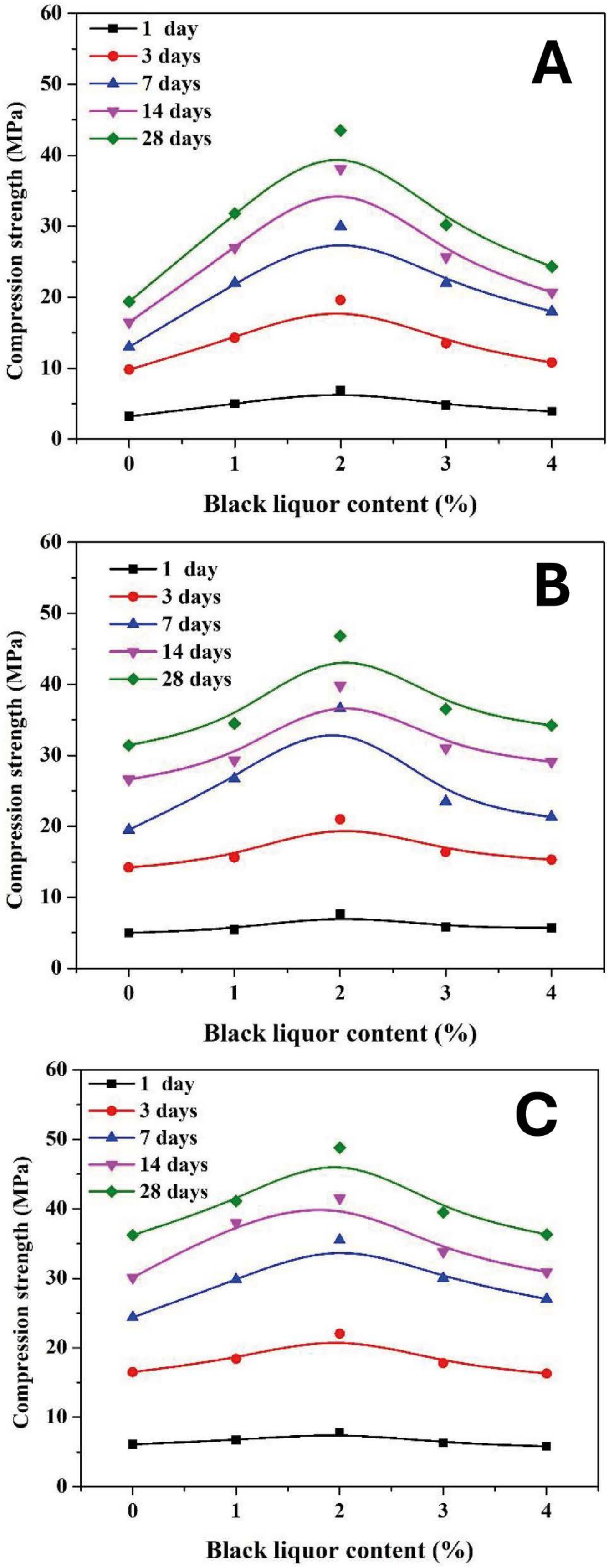

Fig. 1 Effect of black liquor content on the compression strength of the prepared concrete samples: S1; (A), S2; (B) and S3; (C)

1. ábra A fekete lúg tartalom hatása az előállított betonminták nyomósziládságára S1; (A), S2; (B) and S3; (C)

The optimal black liquor content helps refine the microstructure by reducing porosity and enhancing particle packing, as confirmed by studies on similar lignin-based admixtures [18]. The slight strength gains between 28 and 90 days likely results from the continued slow hydration of unreacted clinker phases and secondary pozzolanic reactions.

In conventional concrete without black liquor, compressive strength typically exhibits a slower rate of gain, particularly beyond 28 days. The introduction of black liquor accelerates early strength gain while maintaining long-term performance. This improvement is consistent with studies emphasizing the role of organic admixtures in promoting early strength while ensuring sustainability [18].

The accelerated strength development within 28 days makes black liquor-enhanced concrete suitable for time-sensitive construction projects where early loading is required. Utilizing black liquor, a waste product from the paper industry, promotes eco-friendly practices by reducing waste and minimizing cement usage, thereby lowering the carbon footprint. The stability of compressive strength at 90 days indicates the material's durability and long-term reliability.

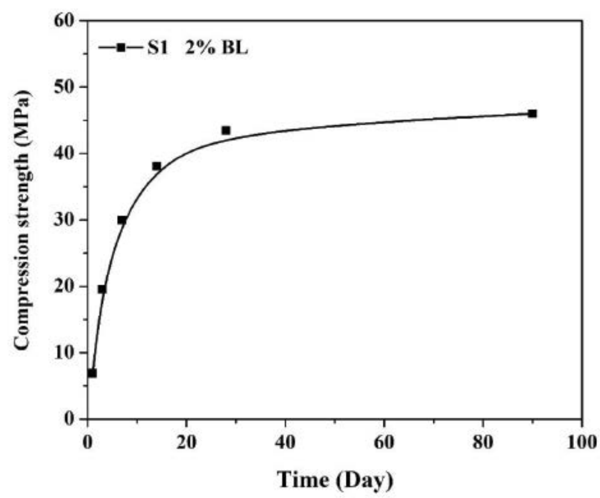

Fig. 2 Effect of residence time on the compression strength of S1 containing 2 % black liquor

2. ábra A tartózkodási idő hatása az S1 minták nyomószilárdságára 2%-os fekete lúg tartalom mellett

Fig. 3A shows the influence of black liquor content on the splitting tensile strength of the utilized three samples. As shown, the splitting tensile strength of the concrete samples (S1, S2, S3) exhibits a parabolic trend with varying black liquor (BL) content. A steady increase in tensile strength is observed with BL content up to 2 wt.%, followed by a gradual decline beyond this concentration. Sample S3 demonstrates the highest splitting tensile strength, followed by S2 and S1, in alignment with their respective cement contents. Numerically, the determined splitting tensile strengths for the three samples were 2.83, 3.24 and 3.52 MPa for S1, S2 and S3 samples, respectively.

The splitting tensile strength of concrete is influenced by its composition, microstructure, and the interaction of additives like black liquor with the cement matrix. As aforementioned, black liquor's lignin and organic compounds contribute to pozzolanic reactions, consuming calcium hydroxide (CH) and forming additional calcium silicate hydrate (C-S-H) gels. These gels enhance the bond strength within the concrete, improving its tensile properties [19]. At 2 wt.% BL, the organic components likely improve workability and compaction, reducing voids and enhancing the tensile load-bearing capacity [18]. At higher BL concentrations (3–4 wt.%), excessive organic compounds disrupt the hydration process and increase porosity, leading to weaker tensile strength. This phenomenon aligns with previous findings on the detrimental effects of over-admixturing in concrete [20]. The inclusion of BL at 2 wt.% may result in improved aggregate-cement paste adhesion, critical for tensile strength. Improved adhesion reduces crack propagation under tensile loads, explaining the observed peak at 2 wt.% [6].

Lowest splitting tensile strength was observed with the lowest cement content sample (S1) due to limited cement content and reduced hydration product formation. The addition of BL provides some improvement but is constrained by the overall composition. For S2 sample, the sample exhibits better tensile strength than S1, reflecting the synergistic effect of BL and higher cement content. BL optimally interacts with the higher volume of cementitious materials to enhance tensile properties. However, S3 achieves the highest tensile strength, benefiting from the ample cement content and the optimized interaction with BL. The matrix is denser, with fewer micro-cracks, leading to superior performance.

Adding black liquor up to 2 wt.% can enhance splitting tensile strength, making concrete more suitable for applications requiring tensile stress resistance, such as pavements and slabs. Black liquor, being a by-product of the paper industry, offers a sustainable and cost-effective alternative to synthetic additives, contributing to waste management and reduced cement consumption. Careful control of BL dosage is necessary to avoid the negative impacts of excess organic material on concrete's tensile properties.

Fig. 3B displays the effect of the black liquor content on the flexural tensile strength of the three concrete samples (S1, S2, S3). As shown, the flexural tensile strength follows a parabolic trend similar to that observed in compressive and splitting tensile strength. The strength improves with increasing black liquor (BL) content up to 2 wt.%, where it reaches a maximum, and declines beyond this concentration. Sample S3 exhibits the highest flexural tensile strength due to its higher cement content, followed by S2 and S1, consistent with the trends observed for other mechanical properties. Numerically, the determined flexural tensile strengths were 13.3, 14.5 and 15.2 MPa for S1, S2 and S3 samples, respectively.

Flexural tensile strength is governed by the concrete matrix's resistance to bending stresses, which depends on its composition, microstructure, and the distribution of stresses across the section. At 2 wt.% BL, the organic compounds in black liquor act as pozzolanic additives, promoting the formation of additional C-S-H gels. These gels fill voids and enhance the bond between aggregates and the cement paste, resulting in higher flexural strength [3, 7]. The improved microstructure at this concentration minimizes crack initiation and propagation under bending stresses, leading to higher flexural performance [6].

At higher BL contents (3–4 wt.%), excess lignin and organic compounds increase porosity and reduce the effectiveness of the cement hydration process. This weakens the matrix and reduces its ability to resist bending stresses [18]. The observed flexural tensile strength trends are closely linked to compressive and splitting tensile strength. The peak performance at 2 wt.% BL is consistent across all strength tests, confirming this concentration as optimal for enhancing the concrete's mechanical properties. The decline beyond 2 wt.% BL highlights the diminishing returns of excess organic material in all strength parameters, underscoring the importance of controlled admixture dosages. Sample hierarchy (S3 > S2 > S1) trend reflects the critical role of cement content in enabling the matrix to utilize the beneficial effects of BL effectively. The same hierarchy was observed for compressive and splitting tensile strengths, emphasizing the interplay between composition and admixture performance.

Flexural tensile strength is particularly sensitive to microstructural properties such as aggregate – cement paste bonding and porosity. The role of BL in reducing voids and improving cohesion between matrix components at 2 wt.% contributes significantly to the observed improvements. The use of BL at 2 wt.% improves the flexural capacity of concrete, making it suitable for applications subject to bending stresses, such as beams, slabs, and pavements. The simultaneous enhancement of compressive, splitting tensile, and flexural tensile strengths highlights the versatility of BL as a sustainable additive. Utilizing BL, a waste by-product of the paper industry, aligns with eco-friendly construction practices by reducing cement consumption and waste disposal issues.

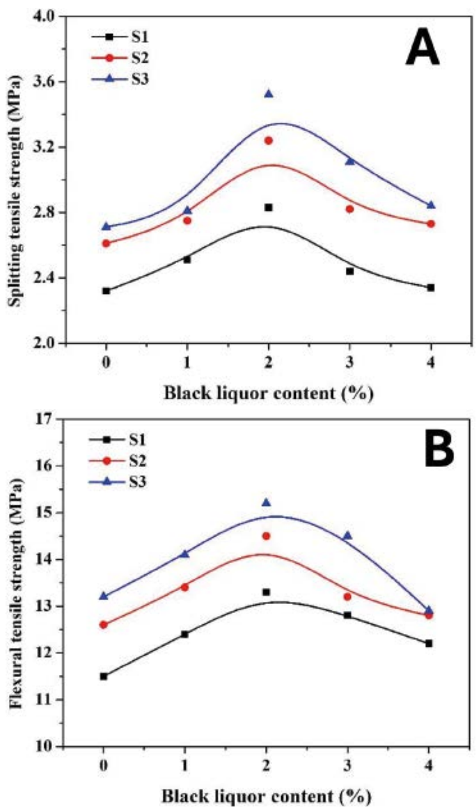

Fig. 3 Effect of black liquor content on the splitting; (A) and flexural; (B) tensile strength of the prepared concrete samples

3. ábra A fekete lúg tartalom hatása az előállított betonminták hasító- (A) és hajlító- (B) húzószilárdságára

Fig. 4A represents the effect of the BL content on the initial setting time of the prepared concrete samples. As shown, the initial setting time of the concrete samples (S1, S2, S3) increases with the addition of black liquor (BL) content:

- At 0 wt.% BL, the initial setting time is approximately 80 minutes for all samples.
- At 2 wt.% BL (optimal content for strength properties), the initial setting time increases to about 140 minutes.
- At 4 wt.% BL, the setting time reaches around 190 minutes, indicating a significant delay.

This trend suggests that black liquor acts as a retarding admixture, slowing down the hydration process and extending the time before the concrete begins to set.

Black liquor contains organic compounds like lignin and sugars, which are known to adsorb onto cement particles. This adsorption creates a barrier that delays the hydration reaction of cement, leading to an increase in setting time [6, 21].

At lower BL concentrations (1–2 wt.%), the retarding effect is moderate, allowing sufficient hydration to develop strength while extending workability. At higher concentrations (3–4 wt.%), the abundance of organic molecules excessively hinders hydration, causing prolonged delays in setting time.

The increase in setting time at 2 wt.% BL aligns with the enhanced mechanical properties (compressive, splitting tensile, and flexural tensile strengths). The delayed setting provides additional time for particle rearrangement and densification of the concrete matrix, contributing to improved strength development. This balance between delayed setting and optimal hydration underscores the beneficial effects of controlled BL addition. At 3–4 wt.% BL, the extended setting time correlates with reduced strength properties. Excessive retardation limits the formation of hydration products within a reasonable timeframe, resulting in weaker concrete [18].

S1 exhibits slightly higher setting times compared to S2 and S3 across all BL concentrations, likely due to its lower cement content and reduced hydration rate. However, S2 and S3 samples show comparable setting times, with S3 demonstrating marginally shorter times at higher BL contents. This reflects the influence of higher cement content in promoting faster hydration despite the presence of BL.

The increased setting time with BL addition enhances workability, making the concrete easier to handle and place, particularly in complex construction projects. The extended setting time at higher BL concentrations could delay construction processes, requiring adjustments in scheduling. By incorporating BL as a retarding admixture, the reliance on synthetic retarders can be reduced, promoting eco-friendly and cost-effective practices.

Fig. 4B displays the influence of the BL content on the final setting time of the prepared concrete samples. As shown, the final setting time of concrete samples (S1, S2, S3) increases linearly with the addition of black liquor (BL) content:

- At 0 wt.% BL, the final setting time is approximately 150 minutes for all samples.
- At 2 wt.% BL (optimal content for strength properties), the final setting time increases to around 270 minutes.
- At 4 wt.% BL, the setting time reaches approximately 400 minutes, indicating a significant delay.

This result parallels the trend observed for the initial setting time, confirming black liquor's role as a retarding admixture. Similar to the initial setting time, the presence of lignin and other organic substances in black liquor delays hydration by forming a temporary barrier around cement particles. This results in a prolonged final setting time. At low BL concentrations (1–2 wt.%), the retarding effect is controlled, ensuring sufficient hydration while extending workability. Higher BL concentrations (3–4 wt.%) exacerbate the delay, slowing hydration excessively and prolonging the final setting process.

The relationship between initial and final setting times remains consistent across BL concentrations. At 0 wt.% BL, the time gap between initial and final setting is approximately 70 minutes (80–150 minutes). At 2 wt.% BL, the gap increases to 130 minutes (140–270 minutes), reflecting a greater overall retardation effect. At 4 wt.% BL, the gap further widens to 210 minutes (190–400 minutes). This proportional increase suggests that black liquor uniformly influences both stages of the setting process, delaying the onset and progression of hydration reactions.

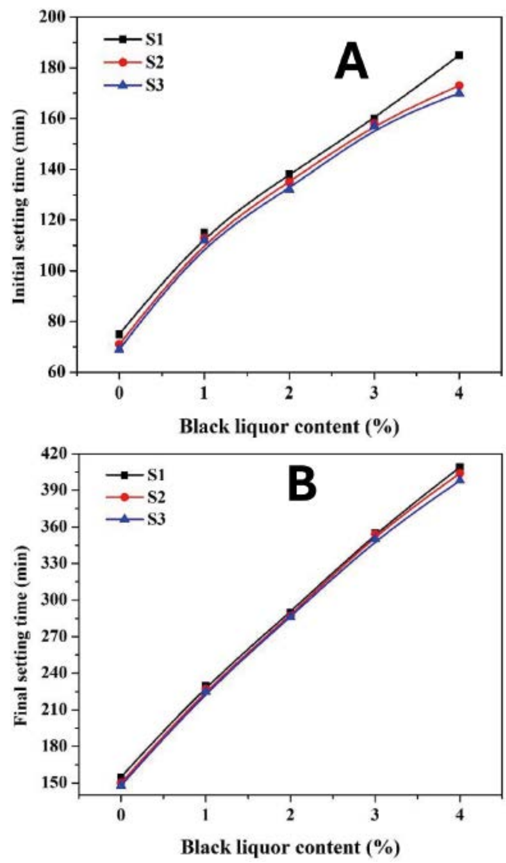

Fig. 4 Effect of black liquor content on the initial and final setting times of the prepared concrete samples.
4. ábra A fekete lúg tartalom hatása az előállított betonminták kötési idejének kezdetére és a teljes kötési időre

At 2 wt.% BL, the extended final setting time allows for better compaction and hydration, contributing to enhanced mechanical properties, as observed in compressive, splitting tensile, and flexural tensile strengths. The balance between delayed setting and optimized hydration underscores the suitability of this concentration for practical applications. Excessive delay in final setting at these concentrations correlates with reduced mechanical properties. The extended hydration period limits the timely formation of hydration products, weakening the overall matrix. The extended setting times improve workability, allowing for better handling and placement of concrete in complex structures. Excessive delays at higher BL contents may disrupt construction schedules, necessitating careful control of dosage. Black liquor provides an eco-friendly alternative to synthetic retarders, reducing environmental impact while enhancing concrete properties.

Fig. 5 demonstrates the impact of the BL content on the bulk density of the prepared concrete samples. As shown, the bulk density of the concrete samples increases with black liquor (BL) content up to 2 wt.%, followed by a slight decline at higher concentrations. Numerically, at 0 wt.% BL, the bulk density is approximately 2.16 kg/l at 1 day and gradually increases over time. At 2 wt.% BL, the bulk density peaks around 2.25–2.28 kg/l (depending on curing time), corresponding to the optimal BL content observed for mechanical properties. Beyond 2 wt.% BL, the bulk density decreases slightly, stabilizing at approximately 2.22–2.23 kg/l at 4 wt.% BL for long curing times (90 days).

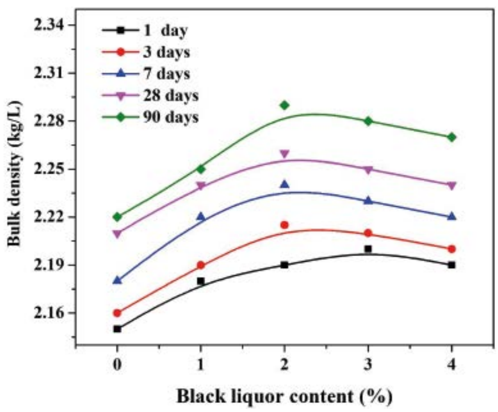

Fig. 5 Effect of black liquor content on the bulk density of the concrete of S1 sample
5. ábra A fekete lúg tartalom hatása az S1 betonminta testsűrűségére

The introduction of black liquor improves compaction and reduces voids in the concrete matrix up to 2 wt.%, resulting in higher bulk density. This is supported by the enhanced particle packing and matrix densification due to secondary pozzolanic reactions that generate additional C-S-H gels. At higher BL contents (3–4 wt.%), the organic components increase porosity by disrupting the hydration process and forming weak zones within the matrix. This reduces the bulk density despite prolonged curing times. At the early stage (1 to 7 days), The bulk density increases rapidly during the first week, reflecting the hydration of cement and the contribution of black liquor to early matrix densification. For instance, at 2 wt.% BL, the bulk density increases from 2.18 kg/l (1 day) to approximately 2.25 kg/l (7 days). Over time, the hydration slows, and the bulk density stabilizes. At 90 days, the bulk density for samples with 2 wt.% BL reaches its peak (~2.28 kg/l), indicating a fully developed matrix.

The peak bulk density at 2 wt.% BL aligns with the highest values observed for compressive, splitting tensile, and flexural tensile strengths. This highlights the role of dense matrix formation in enhancing mechanical performance. The improved bulk density at this concentration reduces porosity, ensuring better load distribution and crack resistance. The decrease in bulk density beyond 2 wt.% correlates with reduced mechanical strengths, emphasizing the detrimental effect of excessive organic content on matrix integrity.

Increased bulk density up to 2 wt.% BL indicates improved workability and durability, making the material suitable for applications requiring dense and durable concrete. The decline in bulk density at higher BL concentrations suggests limitations for structural applications where strength and compactness are critical.

Fig. 6 displays the influence of BL content on the apparent porosity of the prepared concrete. As shown, the apparent porosity of concrete samples decreases significantly with the addition of black liquor (BL) content up to 2 wt.% and increases slightly beyond this concentration. At 0 wt.% BL, the porosity is highest, approximately 16% at 1 day and reduces to about 13% at 90 days due to hydration and matrix densification. At 2 wt.% BL, the porosity reaches its lowest point, approximately 10.5% at 1 day and 9% at 90 days, reflecting optimal densification. Beyond 2 wt.% BL, the porosity increases slightly, stabilizing around 11% at 90 days for 4 wt.% BL. This trend confirms black liquor's ability to reduce porosity by enhancing matrix densification and particle packing up to an optimal concentration.

The organic compounds in black liquor act as a plasticizer, improving the workability of the concrete and allowing better compaction. This results in reduced voids and a denser microstructure at optimal concentrations [22, 6]. The secondary pozzolanic reactions triggered by black liquor produce additional C-S-H gels, filling the pores and further reducing porosity. At higher BL concentrations (3–4 wt.%), excessive organic material disrupts hydration and may lead to poor bonding within the matrix. This creates microvoids and weak zones, increasing porosity.

The decrease in apparent porosity at 2 wt.% BL aligns with the peak in bulk density (~2.28 kg/l). This correlation highlights the role of reduced porosity in improving density and overall compactness. At higher BL contents, the increase in porosity correlates with the slight decline in bulk density, confirming the inverse relationship between these properties. The lowest porosity at 2 wt.% BL contributes to the highest compressive, splitting tensile, and flexural tensile strengths. The denser matrix reduces crack initiation and propagation, resulting in superior mechanical performance. At higher BL concentrations, increased porosity weakens the matrix, leading to reduced mechanical properties.

Apparent porosity decreases rapidly during the first week due to early hydration and the formation of C-S-H gels. For example, at 2 wt.% BL, porosity decreases from 10.5% (1 day) to 9.5% (7 days). Long-term curing further reduces porosity, particularly at optimal BL content. At 90 days, porosity for 2 wt.% BL reaches its minimum (~9%), reflecting a fully developed matrix.

Reduced porosity at 2 wt.% BL improves durability by minimizing water ingress and resistance to environmental degradation. The results emphasize the importance of using BL at its optimal concentration (2 wt.%) to balance porosity reduction with mechanical performance.

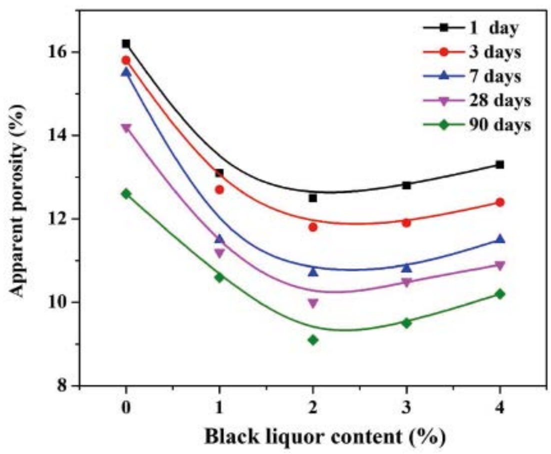

Fig. 6 Effect of black liquor content on the apparent porosity of S1 sample
6. ábra A fekete lúg tartalom hatása az S1 minta látszólagos porozitására

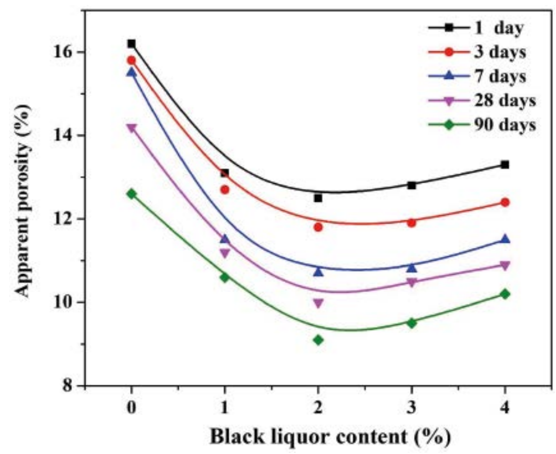

Fig. 7 Effect of black liquor content on the slump value of the prepared concrete samples
7. ábra A fekete lúg tartalom hatása az előállított betonminták roskadási értékére

The slump value of concrete samples (S1, S2, S3) (shown in *Fig. 7*) increases with the addition of black liquor (BL) content across all tested mixes. Typically, at 0 wt.% BL, the slump value is approximately 25 mm, indicating relatively low workability. At 2 wt.% BL, the slump value increases to around 100 mm, reflecting a significant improvement in workability. At 4 wt.% BL, the slump value reaches its maximum, approximately 200 mm for S1, with similarly high values observed for S2 and S3. This trend demonstrates that black liquor effectively enhances the workability of concrete by increasing its slump value.

Black liquor contains lignin and other organic compounds that act as natural plasticizers, reducing the internal friction between particles and allowing the concrete to flow more freely [3]. These compounds also improve the dispersion of cement particles, reducing water demand and enhancing fluidity [23]. Black liquor's organic content enhances the water-retaining capacity of the mix, which contributes to higher slump values [24]. The improved lubrication within the mix reduces resistance to flow, increasing workability without compromising the water-to-cement ratio.

The first sample (S1) exhibits the highest slump values across all BL concentrations, likely due to its lower cement content, which reduces the viscosity of the mix. However, both S2 and S3 samples show slightly lower slump values compared to S1, consistent with their higher cement content, which increases the paste viscosity and slightly restricts flow.

The increased workability at higher BL concentrations is consistent with the observed extension in setting times. The retarding effect of BL allows for greater slump values, improving handling and placement in construction. While higher slump values improve workability, excessive BL content (3–4 wt.%) can compromise mechanical properties due to increased porosity and disrupted hydration dynamics, as previously discussed. The enhanced workability provided by BL addition makes the concrete easier to handle, place, and compact, particularly in complex or heavily reinforced structures. While 4 wt.% BL maximizes the slump value, 2 wt.% BL provides a balanced improvement in workability and mechanical properties, making it the optimal concentration for practical applications.

The SEM images (*Fig. 8*) of the S2 sample with 0% BL and 2% BL provide direct evidence of the microstructural changes induced by the addition of black liquor. For the BL-free sample (*Fig. 8B*), the microstructure shows larger voids and less cohesive matrix bonding. The cement particles appear less integrated, with visible gaps that correlate with higher porosity (~13% at 90 days) and lower bulk density (~2.22 kg/L). On the other hand, for BL-containing sample (*Fig. 8A*), a denser and more compact microstructure is observed, with fewer voids and a more cohesive cement matrix. The enhanced bonding between aggregates and the cement paste supports the lowest porosity (~9% at 90 days) and highest bulk density (~2.28 kg/L), directly contributing to superior mechanical properties.

The SEM image for 2% BL shows the formation of additional C-S-H gels, filling the voids and reducing porosity. This aligns with the observed decrease in apparent porosity from 13% (0% BL) to 9% (2% BL) and the corresponding increase in bulk density from 2.22 kg/L to 2.28 kg/L. The dense microstructure minimizes pathways for crack initiation and water ingress, contributing to improved durability.

The enhanced interfacial bonding visible in the SEM image for 2% BL correlates with the peak compressive strength (~50 MPa at 28 days), splitting tensile strength (~3.6 MPa), and flexural tensile strength (~15 MPa). This demonstrates the role of a compact microstructure in resisting applied stresses. The SEM image for 0% BL highlights a less compact structure with larger voids, which weakens the concrete. This correlates with lower compressive strength (~40 MPa at 28 days) and increased porosity, reducing durability and mechanical performance.

The additional C-S-H gel formation, evidenced in the 2% BL image, results from the pozzolanic reaction of black liquor's organic compounds with calcium hydroxide. This improves matrix densification and reduces porosity [2]. The smoother and more uniform surface of the matrix at 2% BL reflects improved workability and compaction during the mixing process. While not visible in these images, higher BL concentrations (3–4%) are known to disrupt hydration and lead to micro-voids, reducing density and mechanical properties.

The SEM images reinforce the conclusion that 2% BL is the optimal concentration for enhancing both microstructural and macroscopic properties. The denser matrix observed at 2% BL suggests improved resistance to environmental degradation, such as freeze-thaw cycles and chloride ingress.

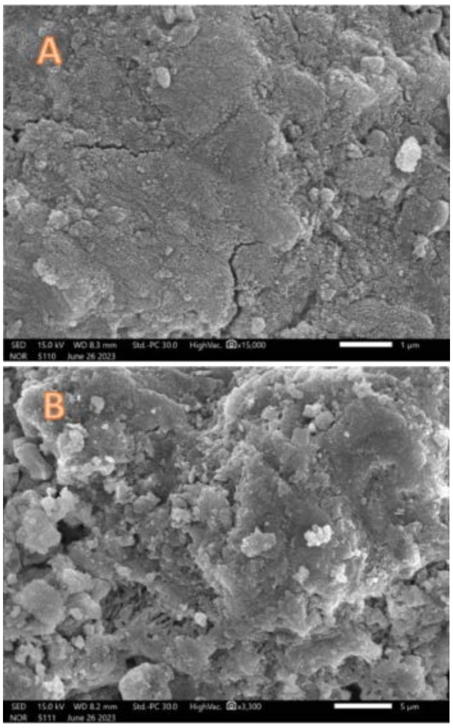

Fig. 8 SEM images for the BL containing (S2, 2% BL); (A) and pristine (S2); (B) concrete samples
8. ábra SEM felvételek a fekete lúg tartalmú (S2, 2% BL); (A) és eredeti (S2); (B) betonmintákról

4. Conclusion

This study demonstrates the potential of black liquor (BL) as an effective and sustainable admixture for concrete. Incorporating BL at an optimal concentration of 2 wt.% significantly enhances the mechanical properties, including compressive strength (66.7% 50 MPa in this study compared to 20~40 MPa in conventional concrete), splitting tensile strength (20% 3.6 MPa in this study compared to 2~4 MPa in conventional concrete), and flexural tensile strength (300% 15 MPa in this study compared to 5 MPa in conventional concrete). Additionally, this concentration results in a denser matrix with reduced porosity (9%) and increased bulk density (2.28 kg/l), as confirmed by microstructural analysis using SEM. The extended setting times observed with BL addition provide improved workability, making it suitable for practical applications. However, excessive BL content (3-4 wt.%) negatively impacts performance due to increased porosity and disrupted hydration dynamics. These findings highlight the dual benefits of performance enhancement and sustainability through the utilization of industrial by-products.

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