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Preferred Chemical Agent for Electrochemical Modification of Physical and Mechanical Parameters of Mudstone

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Abstract: To study the influence of electrochemically modified mediums on the physical and mechanical parameters of mudstone samples, focusing on electrolyte solutions and electrode materials, this paper combines theoretical analysis and experimental research. It analyzes the modification mechanism of mudstone through electrochemical techniques, clarifying that the main factors improving the strength of mudstone are electro-osmotic drainage consolidation and electrochemical reaction cementation. The mudstone was electrochemically modified using the controlled variable method. The mudstone sample's hydraulic properties and shear strength were measured before and after modification. The study compared and analyzed the effectiveness of different modified materials. The results indicated that the liquid limit of the modified mudstone samples decreased by 7.874%, while the plastic limit increased by 9.499%. The type of ions introduced by the electrolyte solution influenced the cementation strength of the mudstone. AlCl3 solutions with a 10% mass fraction and CaCl2 solutions with a 25% mass fraction both effectively modify the reinforcement; however, the AlCl3 solution with a 10% mass fraction is the most effective for modifying mudstone samples. The electrochemical modification of mudstone samples with the three electrode materials (graphite, iron and aluminum) revealed that the samples modified with graphite electrodes had the highest shear strength, while those modified with aluminum electrodes had the lowest shear strength. The internal friction angle of graphite electrode-modified mudstone specimens was 26.7°, compared to the original value of 23.9°, and the cohesion was 34.4 kPa, compared to the original value of 12.3 kPa, nearly three times the original value. It is recommended to use graphite electrodes and a 10% mass fraction of AlCl3 for the electrochemical modification of this type of mudstone in engineering applications.

Keywords: electrochemical modification; mudstone; electro-osmotic drainage consolidation; electrochemical reaction cementation; graphite

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Citation: Chen, X.; Han, L.; Tovele, G.S.V.; Kong, J.; Yang, H. Preferred Chemical Agent for Electrochemical Modification of Physical and Mechanical Parameters of Mudstone. *Appl. Sci.* **2024**, *14*, 11789. https://doi.org/10.3390/app142411789

Academic Editor: Angeles Sanroman Braga

Received: 22 November 2024
Revised: 11 December 2024
Accepted: 13 December 2024
Published: 17 December 2024

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1. Introduction

Mudstone is one of the more common rocks in Chinese open-pit coal mine slopes, which is characterized by a short rock formation time, structural incompleteness, and poor cementation [1]. This leads to its softness, obvious creep after stress, and sensitive hydrological properties when exposed to water; thus, the presence of mudstone layers can have a serious impact on slope stability [2].

Solving the stability problem of mudstone-containing slopes is of great significance to engineering practice. However, the special nature of mudstone makes it more difficult to solve the problem of mudstone reinforcement in engineering, especially mudstone with poor permeability, obvious softening in water, and easily creeping deformation. For example, the slope of Xiaolongtan open-pit mine contains three layers of low-strength mudstone interlayers, and the slope stabilization strength tends to decrease significantly under the influence of long-term mining operation activities [3]. The landslide on the north slope of the Fushun West open-pit mine was caused by precipitation infiltration that

increased the water content of the mudstone layer in the slope, leading to accelerated creep deformation of the north slope [4].

Grouting reinforcement [5] and anchor cable anchoring [6] are the main conventional methods to reinforce slopes. However, the permeability of mudstone is extremely poor, and soft plastic creep deformation predominates over fracture damage [7]. Considering the construction cost and the timeliness of open-pit mine slopes, the above methods are seldom applied to the reinforcement of mudstone-containing slopes in open-pit mines [8]. The solution to the problem of reinforcing slopes containing mudstone in open-pit mines can start from the mudstone itself by changing its hydrophysical or mechanical properties to increase the shear strength of the mudstone and then improving the stability of the slope [9]. Alzo'Ubi et al. investigated the mechanical behaviour of mudstone under temperature and found that the linear Mohr–Coulomb criterion can characterize the shear behaviour of mudstone before and after heat treatment [10]. A small reduction of about 4% in the peak friction angle was observed after thermal treatment at 500 °C, while the cohesion increased by about 47%. Xu et al. found that the addition of lime basalt fibres significantly improved the water sensitivity and water swelling of red mudstone, with the cohesion being more than 14 times higher than that of untreated red mudstone when the fibre content was 0.2% and the lime content was 4% [11]. Feng et al. found that laser irradiation can improve the strength of the soft rock [12].

Compared with the above modification methods, the electrochemical modification technique causes less disturbance and damage to the original slope morphology, and the reinforcement effect is targeted. In the 1930s, electrochemical technology began to be introduced into the reinforcement of weak rock and soil, using electric current fields to achieve electroosmotic drainage and electrode reactions to form anti-slip piles for reinforcement, thereby improving the bearing capacity of weak rock and soil [13]. In 2014, Jones et al. first proposed using electrochemical technology to reinforce slopes at the Fourth International Conference on Surface Geological Improvement Systems, highlighting the method's superiority in environmental protection and economy compared to traditional methods [14]. Lamont et al. carried out electrochemically modified reinforcement on a slope in England, which improved the slope angle compared to the soil nail support method used the previous year [15]. A 29% reduction in construction costs and a 40% reduction in carbon emissions demonstrated that the method can be applied in engineering practice. This method becomes even more energy-efficient and environmentally friendly if renewable energy sources such as wind and solar power can be used to drive the electrochemical modification of the reinforcement.

Electrode materials and electrolyte solutions are important influencing factors of the electrochemical modification effect. Zhou et al. comparatively investigated the effect of iron, graphite, copper, and aluminum electrodes on the electrochemical modification of silt and suggested iron as the preferred electrode material [16]. Chai et al. studied the modification effect of copper, iron, and aluminum electrodes on the compressive strength of coal-bearing mudstone. The results showed a significant improvement in the compressive strength of mudstone, but rapid electrode corrosion remained an issue [17]. In the electrochemical modification process, OU et al. injected calcium chloride and then sodium silicate into the anode and achieved good technical application results [18].

Currently, there are few studies on electrode materials and electrolyte solutions in electrochemical modification, and most of the existing studies have been conducted on silt, sandstone, or soft soil roadbed slopes. Due to the unique properties of mudstone, it is crucial to study the mechanical properties of mudstone after electrochemical modification when reinforcing mudstone-bearing slopes in open-pit mines. Accordingly, the shear strength of mudstone is identified as the primary index in this study. The self-made test equipment was used to study the mechanical characteristics of the mudstone of the open-pit mine after electrochemical modification under the conditions of different electrode materials and electrolyte solutions. The electrode material and electrolyte solution are preferred to

provide a certain basis for the reinforcement management of mudstone-containing slopes in surface coal mines.

2. Research Program and Methodology

2.1. Theoretical Basis for Electrochemical Modification of Mudstones

Mudstone is a sedimentary rock formed by the weathering and deposition of original rock, primarily composed of three clay minerals: illite, kaolinite, and montmorillonite. The clay minerals in mudstone contain aluminum–oxygen octahedra and silica–oxygen tetrahedra that undergo lattice substitution, imparting a certain amount of negative charge to the clay particles [19]. This negative charge causes a large number of hydrated cations to aggregate around the particles. These adsorbed cations can be divided into a fixed layer (adsorption layer) and a diffusion layer based on their adsorption strength, forming a double-layer structure. Electrochemical technology can modify and reinforce mudstone precisely because of the existence of this double electric layer structure in clay minerals [20]. The electrification characteristics and double electric layer structure resulting from lattice substitution in clay mineral particles provide a theoretical basis for the electrochemical reinforcement of mudstone.

2.2. Mechanism of Action of Electrochemically Modified Mudstone

Electrochemical methods mainly improve the properties of mudstone by improving both physical and chemical aspects [21]. The physical improvement is achieved by driving the water inside the mudstone to be fully discharged through electro-osmosis, resulting in drainage and consolidation. The chemical action involves reactions between the electrode, electrolyte, and clay mineral components, forming cement in the particle gaps and improving the mechanical strength of the mudstone.

2.2.1. Electro-Osmotic Drainage Consolidation

Pore water in denser soft clay soils is mainly controlled by capillary forces, so it is difficult to remove it by conventional drainage methods [22]. Applying electric osmotic pressure to the water in the soil not only removes the pore water between the soil particles but also removes the bound water with a strong binding force on the surface of the soil particles. Pore water has a large number of metal cations, and after applying DC voltage to the soil, the pore water will produce electro-osmotic flow from anode to cathode with metal cations [23]. This effect effectively avoids the problem of poor permeability of mudstone and is more conducive to the realization of electrochemical reinforcement of mudstone.

During the energization process, the voltage drives the hydrated cations in the diffusion layer in their free state to move the water molecules toward the cathode for drainage [24]. Ion replacement occurs simultaneously with the drainage process. The high-valence cations in the injected electrolyte solution replace the low-valence cations in the pore water. This results in a decrease in the number of high-valence cations required for charge equilibrium, a reduction in the thickness of the diffusion layer, and subsequently a reduction in the thickness of the water layer surrounding the lattice. Electro-osmosis action effectively eliminates the pore water and bound water inside the mudstone, reduces the overall water content, and ultimately increases the particle density and effective stress within the mudstone [25].

2.2.2. Chemical Reaction Cementation

The migration of cations from the mudstone to the cathode forms particle agglomerates with the soil particles in their path. This phenomenon modifies the plasticity of clay minerals and increases cohesion. It also enhances the frictional properties of soil particles, thereby increasing the internal friction angle [26].

Using iron electrodes as an example, during the electrochemical stabilization of mudstone, iron electrodes undergo redox reactions to introduce iron ions into the clay minerals and produce new cementation materials through hydration reactions, achieving the cemen-

tation of mineral particles. The cementation reaction process when using iron electrodes for the electrochemical stabilization of mudstone is as follows.

Iron electrodes form large amounts of divalent iron ions when energized:

$$Fe - 2e^- = Fe^{2+} \quad (1)$$

Fe^{2+} will hydrolyze with OH^- in water to form the transiently present compound $Fe(OH)_2$:

$$Fe^{2+} + 2H_2O = Fe(OH)_2 + 2H^+ \quad (2)$$

$Fe(OH)_2$ is extremely unstable in air and will all react with the combined action of oxygen and water to form $Fe(OH)_3$:

$$4Fe(OH)_2 + O_2 + 2H_2O = 4Fe(OH)_3 \quad (3)$$

Some of the $Fe(OH)_3$ will weather in the air and undergo a dehydration reaction to form ferric trioxide:

$$2Fe(OH)_3 = Fe_2O_3 + 3H_2O \quad (4)$$

The electrochemical consolidation process described above effectively consolidates mudstone by converting its natural cementation to iron cementation, a method widely accepted by field engineers. The choice of different electrode materials or electrolyte compositions can result in various cementation states, significantly influencing the effectiveness of electrochemical reinforcement.

Mudstone, a widely occurring rock in nature, often appears on slopes. The shear strength of mudstone is a critical factor affecting slope stability. Electrochemical modification can achieve drainage consolidation of mudstone and improve its shear strength parameters, significantly enhancing the stability of slopes containing mudstone. Therefore, this study used the shear strength of mudstone after electrochemical modification as a comparative indicator to select the optimal electrode material, electrolyte solution, and concentration. This achievement has important value for controlling mudstone deformation and improving slope stability.

2.3. Preferred Solution for Electrochemical Modification of Mudstone Agents

The choice of mudstone as the object of electrochemical modification requires a study of its microstructure and physical and mechanical parameters separately, determining whether mudstone possesses the basic properties of electrochemical modification [27], to figure out how the physical and mechanical parameters of the mudstone improve during electrochemical modification [24] and to provide a theoretical basis for the design of the experiment.

In the electrochemical modification research program, the physical and mechanical properties of the original mudstone and the electrochemically modified mudstone are compared, with liquid/plastic limit and shear strength among the physical and mechanical parameters to be tested. The direct shear test is used to measure the shear strength (cohesion C, internal friction angle φ). The experimental outcomes were compared and analyzed to determine the impact of electrochemical modification on mudstone's physical and mechanical properties. The factors influencing the reinforcement process in the electrochemical experiments are investigated further. The control variates were used to constrain the test conditions, and the electrodes of different materials, electrolyte solutions of different compositions, and concentrations were used to conduct electrochemical modification tests on mudstones [28]. The physical and mechanical parameters of the modified mudstone were measured to compare the enhancement of the modified strength of the mudstone by various electrode materials and the modification effect of different electrolyte solutions and concentrations. The optimal modification test conditions from the electrode and electrolyte solutions included in the experiment were selected. The logical structure of this research plan is shown in Figure 1.

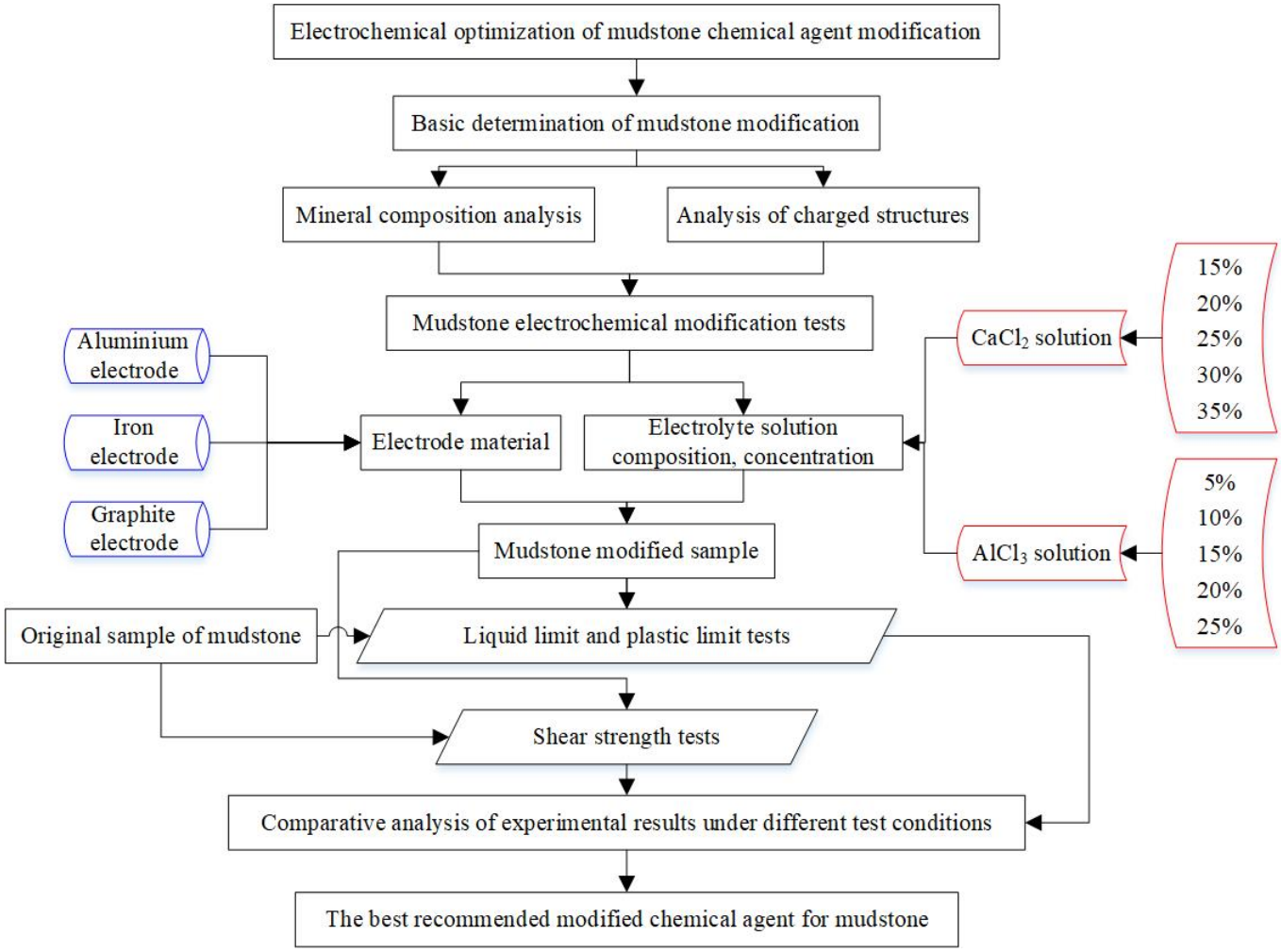


Figure 1. Structure of the study on the preferential method of electrochemical modification of chemical agents.

3. Mudstone Electrochemical Modification Tests Program

3.1. Mudstone Samples Characteristics

The sample used in this experiment is mudstone taken from the north slope of the quarry at the Shengli West No. 1 open-pit coal mine of Guoneng Beidian Shengli Energy in Inner Mongolia Autonomous Region, China. The mineral composition of the mudstone samples was determined using a SmartLab X-ray powder diffractometer, revealing the contents of montmorillonite, kaolinite, illite, and quartz to be 31%, 35%, 16%, and 18%, respectively. The montmorillonite, kaolinite, and illite in these mineral components have good water absorption properties. Montmorillonite and illite expand upon exposure to water, whereas kaolinite, while capable of absorbing water, does not undergo significant expansion. If this type of mudstone exists at a dip angle in the slope, it can significantly contribute to rock deformation and landslide formation.

Mudstone has low permeability and good water absorption. During this experimental study, the water content of the mudstone was controlled at 25%, making it unsaturated. The motion state of water driven by no voltage can be described by Richard's equation for the continuity of water seepage in unsaturated soil and rock. The soil hydraulic characteristics of unsaturated mudstone include the soil moisture characteristic curve and the permeability coefficient curve. The soil water characteristic curve (SWCC) describes the variation in the soil water matrix potential with soil water content, while the permeability coefficient curve represents the relationship between the permeability coefficient and matrix suction. The soil

water characteristic curve provides a foundation for studying the strength, permeability, and constitutive theory of unsaturated soil.

In this study, the mudstone sample's soil water characteristic curve was fitted using the VG parameters. As the moisture content decreases, the soil water characteristic curve transitions through different stages: initially dominated by capillary action, followed by water film adsorption and finally firm adsorption. The electrochemical drainage consolidation and modification of mudstone will further alter its soil water characteristic curve, a topic for future exploration in this study.

3.2. Test Condition Setting

To explore the specific effects of electrochemical modification on mudstone while ensuring the single-variable principle of controlled experiments, the mudstone was processed into standardized samples of the same specifications. A set of mudstone samples for electrochemical modification were selected. After modification, the physical and mechanical parameters of the original mudstone samples were compared to those of the modified samples to analyze the effects of electrochemical modification. The main influencing factors in the electrochemical modification experiment (electrode material, electrolyte solution type, and concentration) were used as variable conditions for this experiment.

There are two types of electrode materials: metallic electrodes and non-metallic electrodes. Metal electrodes produce metal cations in the chemical reaction to achieve the cementation and stabilization of the soil particles. Non-metal electrodes do not undergo electrolytic reactions and thus do not produce cementation that blocks drainage channels, which are more conducive to achieving electro-osmotic drainage. To compare how different electrode materials affect electrochemistry, aluminum and iron, which are commonly used as metal electrodes, were chosen for their high electrical conductivity and low costs [29]. In addition, graphite electrodes were chosen as a control for electrodes made of oxidizable metals, which are characterized by a high specific surface area, strong oxidation resistance, corrosion resistance, and high electrical conductivity [30].

The influence of the electrolyte composition on the electrochemical stabilization effect is similar to that of electrode materials. The reinforcement effect is achieved by introducing cations to produce cement. Depending on the cations introduced, the cement's strength can vary significantly. The concentration of the electrolyte solution affects the total amount of cement produced and the time and drainage consolidation efficiency of the electrochemically modified mudstone. To precisely examine the effect of electrolyte solutions on the electrochemical modification of mudstone, variable test conditions were regulated, and only graphite electrodes were used for comparative electrolyte composition and concentration tests. Mudstone conditions in the laboratory usually change after 20 h, so the electrolyte time factor was not examined separately in the experiments.

3.3. Test Setup and Procedure

Experimental equipment: The experimental setup included an MS602D DC power supply (maximum voltage of 60 V, maximum current of 2A, real-time display of voltage and current), lead wires, three shear sample cylinders (inner diameter 61 mm, length 10 cm, round tube shape), three uniaxial compression sample cylinders (inner diameter 50 mm, length 20 cm, round tube shape), sealed rubber gaskets (diameter 6 cm $\times$ 5 cm), a digital scale, a measuring cup, and a drying box.

Experimental materials: Mudstone specimens, electrolytes ($CaCl_2$ and $AlCl_3$), electrode sheets (graphite, aluminum, and iron), and pure water.

Preparation process of mudstone specimens: Select the mudstone specimens for weighing, crush the specimens, and place them in an oven for drying. Weigh the required amount of mudstone for one specimen, and add electrolyte solution to achieve a 25% moisture content remoulded specimen. The 25% moisture content is higher than the plastic limit of the sample, meeting the requirements for both size and morphology.

Experimental setup: Figure 2 depicts the experimental setup, in which a parting funnel containing the electrolyte solution is placed above the specimen barrel so that the electrolyte solution can flow into the mudstone specimen by gravity. A water collection dish is placed under the sample barrel to catch the solution that drains from the sample cathode.

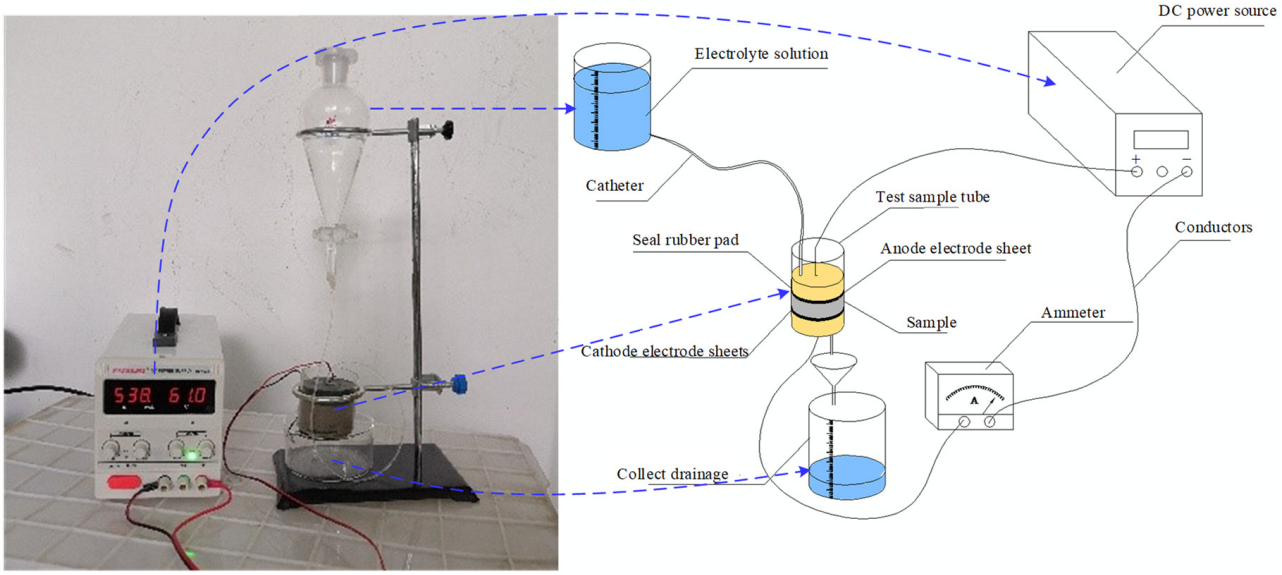


Figure 2. Experimental device for electrochemical modification of mudstone specimens.

After assembling the experimental device [31], connect the power to start the electrochemical modification test. Continuously inject electrochemical solution from the solution conduit at the upper end, and determine the experimental progress by monitoring the current changes during the electrochemical modification process. When the current stabilizes for more than 5 h, the electrochemical modification of mudstone is considered complete.

Experimental design: Experiments were conducted to preferentially select electrochemical modification agents for mudstone based on two variable factors: electrolyte (CaCl2, AlCl3) and electrode materials (Fe, Al, graphite). As shown in Table 1, the orthogonal experimental method was used to design six sets of electrochemical modification tests. Only graphite electrodes were used in the tests for the preferential selection of the electrolyte solution concentration, and the concentrations (mass fractions) of the two electrolytes were set according to Table 1.

Table 1. Electrochemical modification test protocol.

Electrode	Electrolyte		Mass Fraction of $CaCl_2$ Solution					Mass Fraction of $AlCl_3$ Solution				
	$CaCl_2$	$AlCl_3$										
Iron	Test 1	Test 2	-	-	-	-	-	-	-	-	-	-
Aluminum	Test 3	Test 4	-	-	-	-	-	-	-	-	-	-
Graphite	Test 5	Test 6	15%	20%	25%	30%	35%	5%	10%	15%	20%	25%

3.4. Determination of the Electrolyte Solution Amount

During the test of the electrochemical modification of mudstone, the volume of electrolytes injected into the sample via the separatory funnel needs to be determined through repeated modification procedures. The specific determination method is as follows:

(1) Begin with a given initial volume of electrolytes for testing. When the current stabilizes at zero and no longer changes, remove the sample, weigh it to determine the decrease in mass, and then conduct a direct shear test on the sample.

(2) Replenish the electrolyte solution with the same mass as the initial mass reduction in the specimen. Inject the electrolyte into the specimen that has completed the initial

modification 4–5 times, allowing it to stand so that the electrolyte is uniformly distributed. Conduct the electrochemical modification again until the current stabilizes at zero. To determine the mass reduction value, remove the secondary modified specimen and weigh it. Maintain the same vertical direction as the initial test specimen and conduct a direct shear test on the secondary specimen.

(3) Repeat the above process until the modification experiment no longer changes the shear strength of the mudstone. The total amount of electrolyte consumed in this repeated process is the volume required for a mudstone sample to achieve complete modification.

Prepare mudstone samples, conduct electrochemical modification tests according to the above experimental conditions, and test the specimen's liquid and plastic limits and shear strength parameters. Compare these properties to evaluate the electrochemical modification effects under different experimental conditions.

4. Results

4.1. Comparison of Mudstone Liquid/Plastic Limit Modification

4.1.1. Determination of the In Situ Mudstone Liquid/Plastic Limit

To investigate the effect of electrochemical modification on the liquid and plastic limit of mudstone, the relevant parameters of mudstone before modification were first determined experimentally. The tests were carried out using the falling cone method with cone depths of 17 mm and 2 mm at the liquid and plastic limits, respectively.

First, prepare three samples with different water contents, corresponding to the mudstone's flow state, plasticity state, and intermediate state, as shown in Figure 3a. The actual moisture content for these three states is calculated using the drying method after determining the liquid and plastic limits. The experimental process and results are shown in Figure 3.

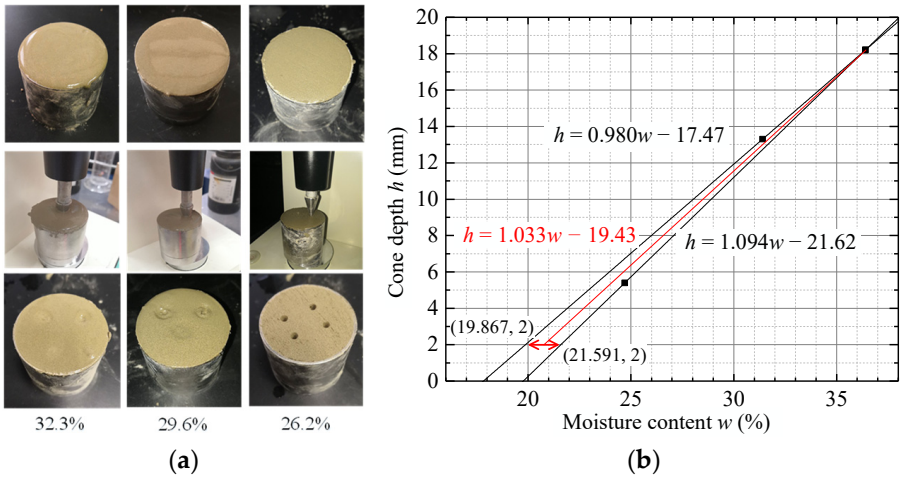


Figure 3. The experiment and results of the combined determination method of Atterberg limits. (**a**) Three moisture content samples; (**b**) test results.

Figure 3b indicates that the difference in water content at a cone depth of 2 mm is 21.591% − 19.867% = 1.72%, which is less than 2%, indicating that the results of the test are reliable. The plastic limit water content of the mudstone before modification was calculated to be 20.745% and the liquid limit water content to be 35.266%.

4.1.2. Changes in the Liquid/Plastic Limit of the Modified Mudstone

As a fundamental property of mudstone, the liquid/plastic limit is extraordinarily sensitive to internal factors. The cementation and electrophoresis produced by electrochemical modification increase the plasticity index of the mudstone to a certain degree because the introduced ions affect the particle size composition and chemical composition of the

soil. Figure 4 shows the joint test results to find the limit between liquid and plastic for modified specimens.

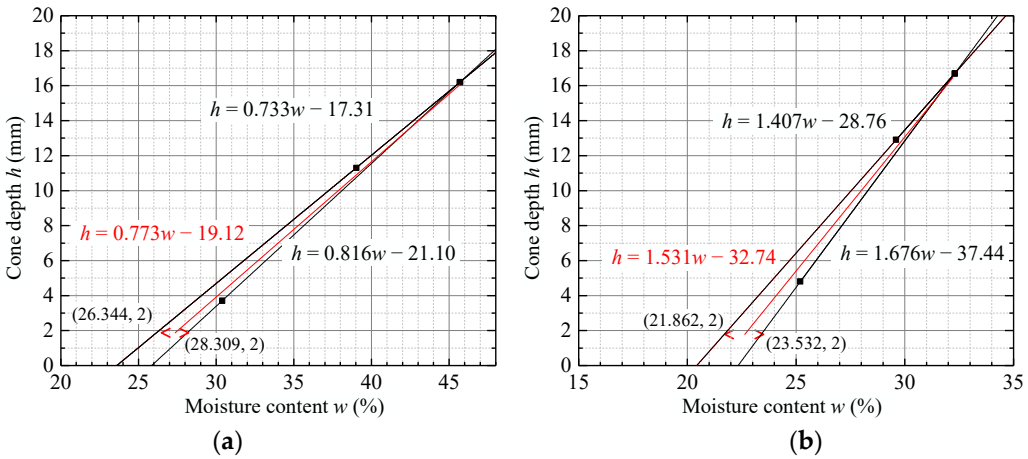


Figure 4. The combined determination method of Atterberg limits the results of modified specimens. (**a**) unmodified by the addition of 10% $AlCl_3$ solution; (**b**) modified by the addition of 10% $AlCl_3$ solution.

At a cone depth of 2 mm, the difference in water content between Figure 4a,b is less than 2%, indicating that the experimental data are reliable. After the addition of 10% AlCl3 solution, the water content of the unmodified specimen was 27.322% of the plastic limit and 46.727% of the liquid limit. After modification, the water content of the plastic limit is 22.691%, and the water content of the liquid limit is 32.489%.

The addition of the $AlCl_3$ solution significantly improved the liquid limit water content of the soil because polymeric $AlCl_3$, as a flocculant, can hydrolyze with water and produce a certain degree of aggregation of soil particles, resulting in the formation of large soil particles and water-stable agglomerates to improve the soil's water holding capacity. The liquid limit moisture content of the modified sample has decreased to a certain extent. The plastic limit has slightly increased, which is caused by the dual effects of ion introduction and electrochemical reaction.

4.2. Modification Analysis of the Mudstone Shear Strength

4.2.1. Determination of In Situ Mudstone Shear Strength

Prepare multiple sets of mudstone samples with different water contents and use a quadruple direct shear tester to test the shear strength of mudstone at these varying water contents. Conduct direct shear experiments on four samples with the same moisture content in each group. The results are shown in Figure 5.

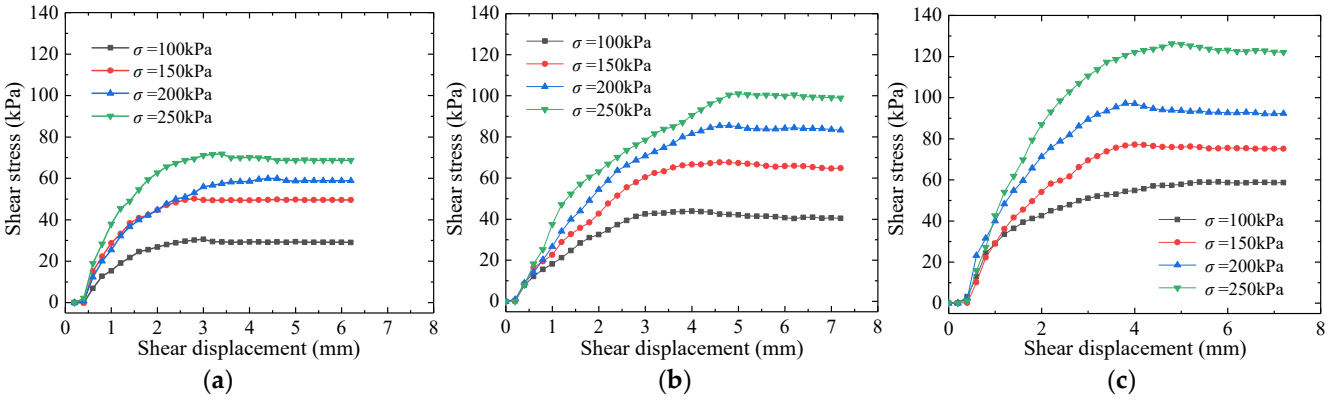


Figure 5. Shear stress–shear displacement curves of mudstone under different water content. (**a**) w = 25%; (**b**) w = 20%; (**c**) w = 15%.

As the water content decreases, both the shear strength and residual shear strength show an upward trend, and the shear stress on the specimen increases at an accelerated rate. The shear stress–shear displacement charts for the three mudstones with varying water content follow essentially the same loading path, with a short elastic phase (rapid increase in shear stress), a long plastic phase (slow increase in shear stress), and a residual shear phase (basic shear stress stability).

According to Figure 5, the τ-σ curves of mudstone under different water content conditions were plotted and linearly fitted to obtain the equations between shear stress and normal stress, as shown in Figure 6.

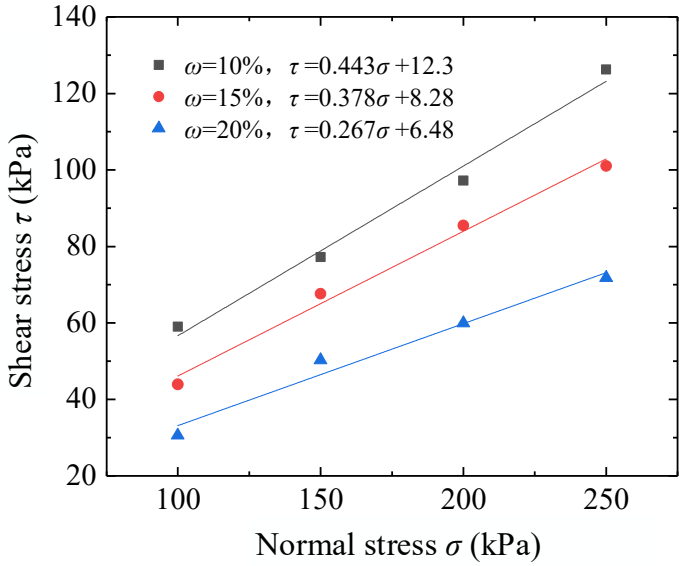


Figure 6. The τ-σ curves of mudstone under different water content.

According to the fitted equations, the C and φ of mudstone under three water content conditions can be obtained, as shown in Table 2.

Table 2. Shear strength parameters for in situ mudstone.

Water Content		**15%**	**20%**	**25%**
Shear strength parameters	φ (°)	23.9	20.7	14.9
	C (kPa)	12.3	8.28	6.48

When the specimen's water content increased from 15% to 20%, the shear strength parameter cohesion C and the internal friction angle φ decreased by 32.7% and 13.3%, respectively. When the water content increased from 20% to 25%, the internal friction angle φ value decreased to 14.9°, a decrease of 28%, while the decrease in cohesion C was 21.7%, which slowed down compared to the previous range. This indicates that the relationship between mudstone's C and φ values and its water content is nonlinear.

4.2.2. Variation in the Shear Strength of Modified Mudstone

The shear strength index of the mudstone specimens under different modification conditions is a combination of the cementing effect of electrochemical modification and the drainage consolidation effect. In this study, the direct shear test was conducted with the normal stress of σ = 200 kPa, and the shear specimens were mudstone samples that had undergone various electrolytic modifications. Table 3 displays the electrolytic conditions of the specimens. Figure 7 depicts the process curves for the three shear tests.

Table 3. Electrolytic conditions for direct shear specimens.

Specimens	Electrolytic Conditions I	Electrolytic Conditions II
Specimen 1	Samples with 25% water content, electrolysis time 3.5 h	-
Specimen 2	Samples with 25% water content, electrolysis time 3.5 h	25 mL of solution in 3 drops, electrolysis over 5.5 h
Specimen 3	Samples with 25% water content, electrolysis time 3.5 h	50 mL of solution in 6 drops, electrolysis over 7.5 h

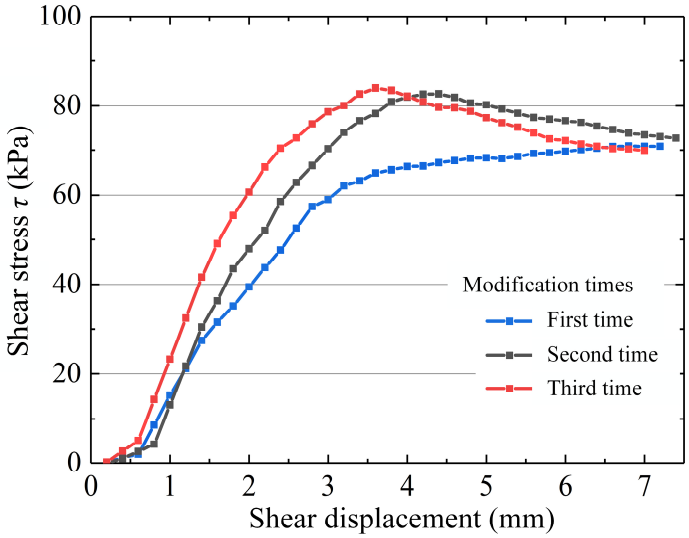


Figure 7. The relationship between the modification times and the shear strength of mudstone.

Figure 7 demonstrates that electrochemical modification significantly increased the shear strength of the mudstone specimens. At a vertical pressure of 200 kPa, the maximum shear strength of unmodified mudstone specimens with a 25% water content was only τ_0 = 59 kPa. As the amount of electrolyte increased, the shear strength of the electrochemically modified mudstone specimens increased further. The maximum shear strength of specimen 1 was τ_1 = 70 kPa, whereas specimens 2 and 3 had maximum shear strengths of τ_2 = 82 kPa and τ_3 = 83 kPa, respectively. The increase in shear strength of the mudstone under the electrochemical modification conditions for specimen 3 was not statistically significant compared to specimen 2, primarily because the electrolyte solution injected during the electrochemical modification of specimen 3 had already reached the quantity necessary for complete mudstone specimen modification. Even if the amount of electrolyte solution continues to increase indefinitely, it will be difficult to further enhance the shear strength of the mudstone.

The shear stress–shear displacement curve can be divided into five stages, which are the compaction stage, linear elasticity stage, yield stage, peak shear stress stage, and post-peak softening stage [32]. Generally speaking, the shear stress of clay soft rock reaches its peak value and then gradually decreases to the residual strength (post-peak softening stage), and the curve shows a continuous process of change. At this time, the specimen displays shear damage, plastic damage, and a residual strength that was slightly less than or approximately equal to the peak strength. It can be seen from Figure 7 that the shear stress–shear displacement curve of the mudstone sample after electro-osmotic consolidation and drainage still conforms to the characteristics of clay soft rock.

5. Discussion

Alzo'Ubi et al. investigated the mechanical properties of mudstone under the influence of temperature and found that the peak shear strength of mudstone increased significantly with increasing temperature [10]. Xu et al. found that the addition of lime–basalt fibre significantly improved the water sensitivity and shear strength of mudstone [11]. Nevertheless, considering the financial and topographical constraints, it is difficult to apply

measures such as high-temperature treatment or the addition of modified materials to mudstone to improve its shear strength. In contrast, electrochemical modification methods have greater applicability, and this paper proposes the utilization of electrochemical modification to enhance the shear strength of mudstone. Therefore, the electrolyte solutions and electrode materials used in the electrochemical modification are investigated and optimized in this paper.

5.1. Electrolyte Solution Optimization

According to the research plan designed in Figure 1, the electrochemical medium selection process is conducted in steps. First, the best modification effect is selected from the two electrolyte solutions, $CaCl_2$ and $AlCl_3$, and the optimal solution concentration is determined. Based on this, the mudstone was modified with the optimal electrolyte solution and electrodes made of graphite, aluminum, and iron. The optimal electrode material was then selected based on the shear strength of the modified mudstone. From the above research results, a set of optimal electrode materials, electrolyte solutions, and concentrations was selected. Multiple concentrations were established for each solution, with mass fractions of 15%, 20%, 25%, 30%, and 35% for the $CaCl_2$ solution and 5%, 10%, 15%, 20%, and 25% for the $AlCl_3$ solution. The specimens were modified with the two electrolyte solutions of different concentrations. The modified samples were subjected to a direct shear test, and the shear strength of the modified specimens under 200 kPa vertical pressure was obtained, as shown in Figure 8.

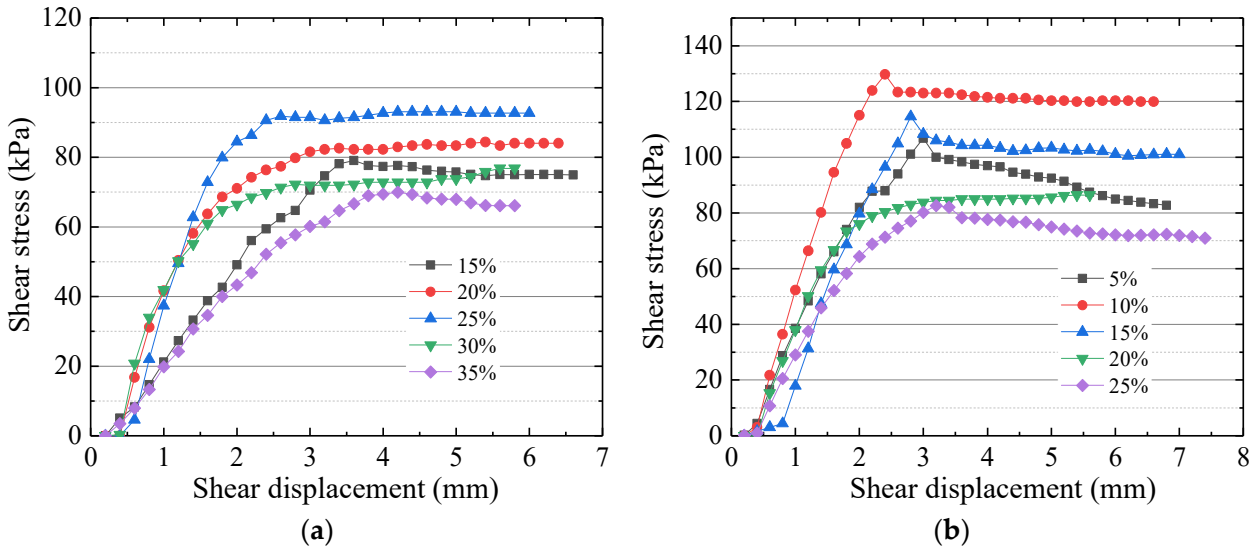


Figure 8. Shear strength of mudstone corresponds to different mass fraction electrolyte solutions (σ = 200 kPa). (**a**) $CaCl_2$; (**b**) $AlCl_3$.

According to the shear strength test results, the electrolyte modification and its solution concentration were ranked in terms of their effectiveness. $CaCl_2$ solutions with a mass fraction of 25% and $AlCl_3$ solutions with a mass fraction of 10% modify the reinforcement more effectively. The enhancement effect of the $AlCl_3$ solution on the shear strength of the specimens is greater than that of the $CaCl_2$ solution; consequently, the 10% $AlCl_3$ solution is the most effective in modifying the reinforcement. During the shear process, specimens modified with $CaCl_2$ solution exhibited significant soft plasticity. The peak shear strength and residual shear strength were essentially the same, and there was no significant decrease in the post-peak strength. The specimens modified with 5%, 10%, and 15% mass fractions of $AlCl_3$ solution exhibited brittle damage during shear, with obvious strain softening characteristics; the 5% $AlCl_3$ solution-modified specimens also exhibited a continuous decrease in residual strength as shear progressed, whereas the specimens modified with

20% and 25% mass fractions exhibited no brittle damage during shear and continued to exhibit soft plasticity.

From Figure 8a, it can be seen that the mudstone samples modified with 15% to 35% $CaCl_2$ solution still maintain the soft plastic characteristics of the clay rock and there is a gradual change from peak strength to residual strength. The residual shear strength of the modified mudstone samples is significantly improved compared to the original mudstone samples or those subjected only to electro-osmotic drainage. The residual shear strength of the mudstone sample with a 25% moisture content after electro-osmotic drainage is 70 kPa. After modification with a 25% $CaCl_2$ solution, the residual shear strength of the mudstone sample with the same moisture content reaches 93 kPa, indicating an increase of more than 20 kPa. The electrochemical modification effect is significant.

After the mudstone sample modification by $AlCl_3$ solution, its structure changes from cohesion soil to sandy soil, as shown in the shear strength chart in Figure 8b. The shear process exhibits brittle failure, where the peak strength suddenly decreases to the residual strength without a gradual decrease. This experimental result indicates that during the electrochemical modification process of mudstone, the addition of $AlCl_3$ solution changes the lattice structure and bonding state inside the mudstone, improves the bonding state between the powder crystal structures and results in the peak value and residual shear strength of the modified mudstone being better than that of the 25% $CaCl_2$ solution modification.

The electrochemical modification of mudstones has a long research history and an abundance of results. Studies on the modification of various types of electrolytes have primarily focused on comparing changes in parameters such as tensile strength, water absorption, swelling, and resistivity. Many researchers have investigated the modification of coastal calcareous soils using $CaCl_2$ as the electrolyte and the modification of illite and kaolinite [33]. $CaCl_2$ and $Al_2(SO_4)_3 \bullet 18H_2O$ have been demonstrated to be effective cementing agents [34]. However, comparative studies on the efficacy of modifications with different electrolyte solutions are scarce. $AlCl_3$ electrolyte was found to be more effective in modifying mudstone than $CaCl_2$ electrolyte, resulting in a significant increase in shear strength for the selected mudstone specimens. The $CaCl_2$-based electrolyte solution was selected for the modification test because the introduction of calcium ions tends to result in the calcification of the cemented state of the mudstone, thereby increasing the mudstone's mechanical strength.

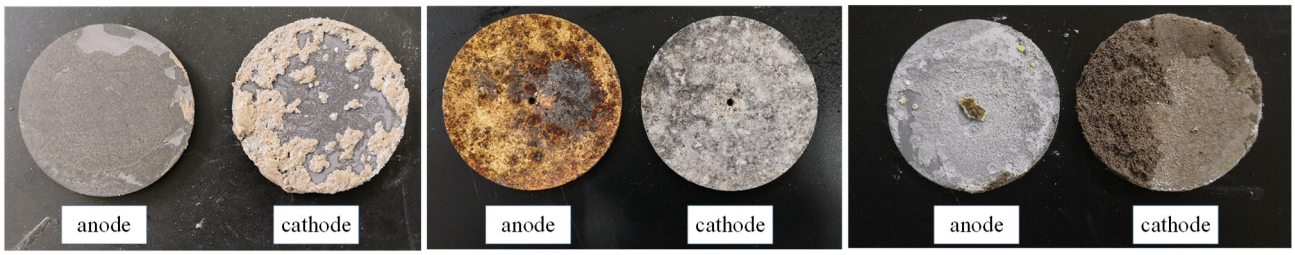
As the application scenario of this study is a soft rock slope, the mudstone shear strength and hydraulic properties were utilized as comparison variables. The test results indicated that a $CaCl_2$ solution with a mass fraction of 25% was the most effective at altering the mudstone samples. This result contradicts the results of Shang et al. [33], who discovered that a 15% $CaCl_2$ solution was the most effective for the electrochemical modification of offshore calcareous soils. The mudstone specimens chosen for this study had a mineral composition dominated by kaolinite. The calcareous soils studied by Shang et al. (2004) were inherently rich in Ca^{2+} and thus required less Ca^{2+} to achieve optimal electrochemical modification than in this study. In addition, Shang et al. [33] also found that a 10% solution of $Al_2(SO_4)_3 \bullet 18H_2O$ was the most effective for the electrochemical modification of offshore calcareous soils, and this study also found that a 10% solution of $AlCl_3$ was the most effective for mudstone specimen modification. The results of the two studies are similar and it was found that a 10% solution concentration is best for the electrochemical modification of soils or mudstones that do not have Al^{3+}.

This study focuses on the changes in the shear strength of mudstone before and after electrochemical modification. Lo et al. conducted field tests on the electrochemical reinforcement of soft, sensitive clays using the electro-penetration method and discovered a roughly 50% increase in the undrained shear strength of the clay in the electrode region [35]. Chien et al. found an average increase in the undrained shear strength of approximately 195% for soils close to the cathode [36]. Chien et al. observed an average increase of 125%–130% from the anode to the cathode [37]. Shang et al. reported an increase in undrained shear strength of soils by 38% to 138%, with the effective cohesion increasing from 0 to 11 kPa [33].

In this study, electrochemical modification increased the undrained shear strength of the mudstone. The peak shear strength of the mudstone specimens modified with 25% $CaCl_2$ solution was τ_4 = 93 kPa, and the peak shear strength of the specimens modified with 10% $AlCl_3$ solution was τ_5 = 130 kPa. These represent increases of 158% and 220%, respectively, compared to the shear strength before modification (τ_0 = 59 kPa). Compared to the shear strength of mudstone specimens with only electro-osmotic drainage (τ_1 = 70 kPa), the shear strengths increased by 133% and 186%, respectively. Thus, electrochemical modification significantly improved the shear strength of mudstone, with the $AlCl_3$ solution having a superior effect.

5.2. Electrode Material Optimization

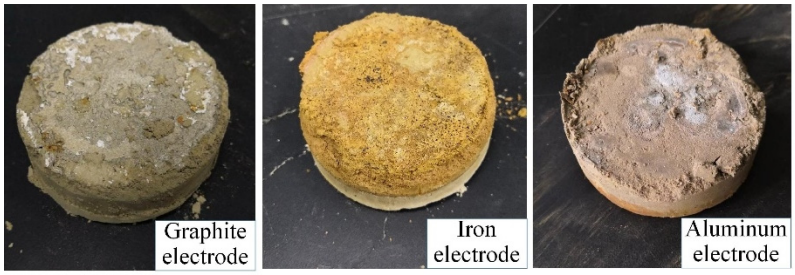
In this study, electrochemical experiments using three electrode materials (graphite, aluminum, and iron) were conducted, and their shear strength was evaluated after the experiments. Figure 9 depicts the modifications of the electrode sheets at the end of the electrochemical modification test. The graphite electrode underwent no significant changes, whereas the anode of the iron electrode corroded severely, with new areas of rust and yellowish-brown rust (Fe_2O_3) and small black areas (Fe_3O_4) visible on its surface. The anode corrosion of the aluminum electrode was also severe, with a significant amount of white material being lost (Al_2O_3). The cathodes of all three electrode materials experienced minimal corrosion, with more mudstone particles adhering to their surfaces due to the cathodes absorbing cations from the solution and mudstone particles around the clumped cations when energized.



(**a**) (**b**) (**c**)

Figure 9. Electrode after electrochemical reaction. (**a**) Graphite electrode. (**b**) Iron electrode. (**c**) Aluminum electrode.

Figures 10 and 11 depict the results of the modified specimens' shear strength tests with the three electrodes at the optimal electrolyte concentration (10% $AlCl_3$). The effect of the aluminum electrode on the specimens was relatively minor and the specimens remained soft and plastic, indicating that the specimen still had a high water content; in the iron electrode group, more iron ions penetrated the surface of the anode area and the penetration depth nearly reached the cathode. The sample surface changed colour, which makes it clear that it is not the same colour as the surface of the cathode.



(**a**) (**b**) (**c**)

Figure 10. Surface of specimens at 3 electrode conditions (10% $AlCl_3$). (**a**) Graphite electrode. (**b**) Iron electrode. (**c**) Aluminum electrode.

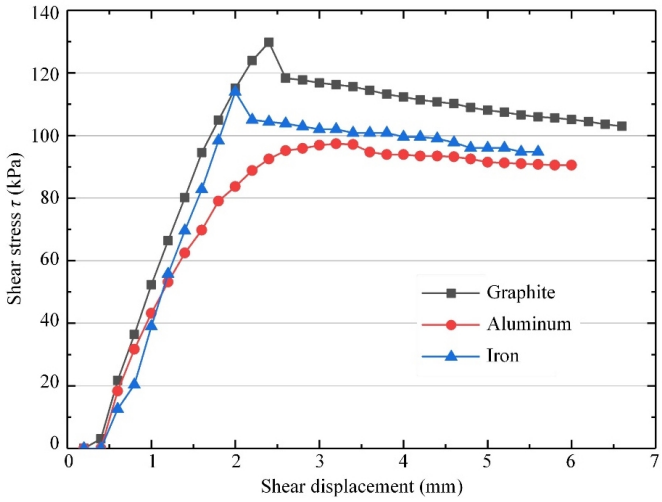


Figure 11. Shear strength of specimens at 3 electrode conditions (10% $AlCl_3$).

Three electrode materials were used for the electrochemical modification of mudstone, resulting in mudstone samples with different shear failure characteristics, as shown in Figure 11. The mudstone samples modified with graphite and iron electrodes exhibited brittle fracture characteristics, whereas those modified with aluminum electrodes still exhibited plastic fracture characteristics. Using the same 10% $AlCl_3$ solution, the aluminum electrode-modified samples showed plastic damage, which conflicts with the results of the electrolyte solution comparison mentioned earlier.

By comparing Figure 8b, it can be seen that when the concentration of Al^{3+} in the solution reaches 25%, the shear failure characteristics of the modified sample align well with the shear test curve of the aluminum electrode-modified mudstone sample in Figure 11. This indicates that when aluminum electrodes and an $AlCl_3$ solution are used to modify mudstone, the concentration of Al^{3+} in the solution increases, leading to a decrease in the shear strength of the modified mudstone. This further confirms that the modification effect of the 10% $AlCl_3$ solution is better. The other two electrode materials did not change the concentration of the electrolyte solution. The shear test curve characteristics of the modified samples were consistent, and the graphite electrode showed a better modification effect on mudstone.

As the electrode material influences the electrochemical modification of mudstone, this study found that the graphite electrode has the best modification effect. Experts and academics have also conducted relevant research in this field. Zhou et al. [16] performed a laboratory electrochemical modification of silt using electrodes made of iron, graphite, copper, and aluminum. The experimental results demonstrated that the effects of different electrode materials were directly reflected in voltage loss, with comprehensive comparisons identifying iron as the optimal electrode material for electro-osmosis. Their investigation revealed that electro-osmosis relies on the ions in the original soil rather than those produced by the electrode reaction. Zhou et al. [16] screened iron as the optimal electrode material by comparing the electro-osmosis effect and ionic strength, which is the primary reason for the discrepancy with the results of this study.

In contrast, this study used the shear strength of the mudstone as a screening criterion and found graphite electrodes to be the most effective. Our findings differ from those of Zhou et al. [16] due to disparities in target orientation and application scenarios. In their study of the effects of the electrode material and current interval on the electro-osmotic permeability coefficient and voltage loss at the soil–electrode interface, Shang et al. discovered that the voltage loss was related to the anode material, with metal anodes exhibiting less loss than carbon anodes [33]. These studies ranked electrode materials according to different objectives, resulting in varying conclusions.

In a study conducted by Bergado et al. on the electro-osmotic consolidation of clay soils in Bangkok using copper and carbon electrodes along with prefabricated vertical drains, it was discovered that electro-osmotic drainage consolidation was superior with carbon electrodes, resulting in 9% greater electro-osmotic drainage than copper electrodes and a 144% increase in the shear strength of clay soils [38]. In contrast, Tonnizam et al. investigated the effect of electrode type on the electro-osmosis of Malaysian soils and found that different types of electrodes produced the same cumulative drainage. However, the effect on shear strength was dependent on the electrode type [39].

Considering that more types of electrodes are compared in this study, including graphite, iron and aluminum, the results still indicate that graphite electrodes are the best material. Bergado et al. [38] increased the shear strength of clay by 144% without the addition of any electrolyte solution. In the present study, the shear strength increased from τ_0 = 59 kPa to τ_1 = 70 kPa, an increase of 118%, when only electro-osmotic drainage was applied. Bergado et al. [38] used a maximum voltage difference of 120 V/m, while in the present study, the maximum voltage difference was only 60 V, which is the primary reason for the difference in the shear strength increment.

As shown in Table 4, the shear-damaged specimens modified with graphite, iron, and aluminum electrodes were dried to determine the moisture content of each specimen. The graphite electrode group exhibited the best drainage effect and the highest degree of specimen hardening. The relatively poor drainage effect of the two metal electrode groups is attributed to the formation of a large number of metal oxides at the anode after oxidation due to energization. These oxides, being insulating, cause the passivation of the metal electrodes, thereby reducing the potential difference between the cathode and anode, affecting the efficiency of the energized modified drainage.

Table 4. The water content of the three electrode-modified specimens (samples).

Electrode	Sample Mass/g	Sample Mass After Drying/g	Moisture Content/%
Graphite	87.535	79.433	10.2
Iron	85.427	73.809	15.7
Aluminum	91.168	76.855	18.6

The oxides formed by the iron electrode primarily penetrate the soil, whereas those formed by the aluminum electrode are mostly adsorbed on the surface of the electrode, significantly impacting the energizing efficiency of the modification experiment. Consequently, the iron electrode is superior to the aluminum electrode in terms of the drainage effect. On the other hand, non-metallic electrodes, such as graphite, do not oxidize when energized, resulting in the most effective drainage consolidation effect. Figure 12 depicts the results of straight shear experiments on graphite electrode-modified specimens.

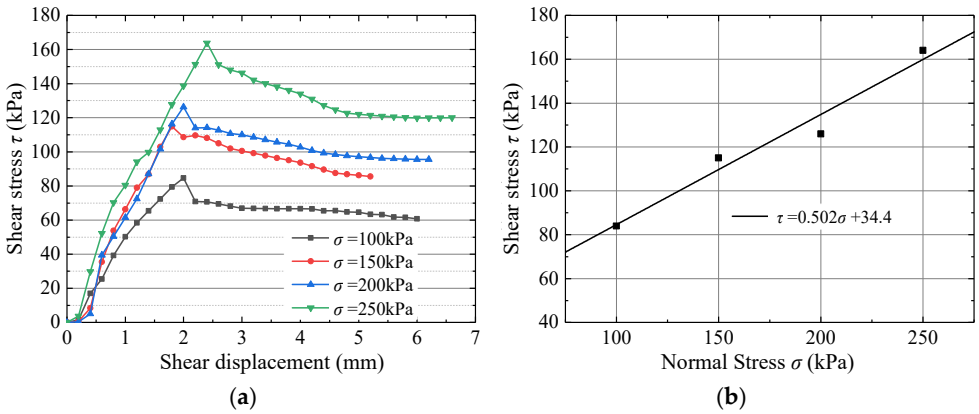


Figure 12. Shear strength parameters of graphite electrode modified specimens. (**a**) Shear stress–displacement curve; (**b**) shear stress–normal stress curve.

The internal friction angle of graphite electrode-modified mudstone specimens was 26.7°, compared to the original value of 23.9°, and the cohesion was 34.4 kPa, compared to the original value of 12.3 kPa, nearly three times the original value.

The findings of our investigation are of great importance to researchers in various fields such as the slope stabilization and electrochemical modification of open-pit mines. As alluded to in the Introduction Section, mudstone is one of the more common rocks on the slopes of open-pit coal mines in China. Its presence can have a serious impact on slope stability because of its special properties such as softness and creep, and conventional reinforcement methods do not apply to mudstone-containing slopes. Therefore, it becomes imperative to study the electrochemical modification of mudstone reinforcement methods from the mudstone itself. The results of this study are particularly important when selecting the optimal electrode material and electrolyte solution, as the selection of electrode material and electrolyte solution has a significant effect on the modification effect. From the test results, for the mudstone of Shengli No.1 open-pit mine, electrochemical modification using graphite electrodes with 10% $AlCl_3$ electrolyte can achieve better results.

6. Conclusions

Through theoretical analysis and experimentation, we conducted experiments on electro-osmotic drainage and the electrochemical modification of mudstone, with a focus on elucidating the differences in the modification effects of electrolyte solutions and electrode materials on the liquid and plastic limits and shear strength of mudstone. The following conclusions were reached.

(1) From the standpoint of theoretical analysis, the mechanism of the modification effect of electrochemical technology on mudstone is explained, focusing primarily on electro-osmotic drainage consolidation and electrochemical reaction cementation. After testing and comparing the liquid and plastic limits of mudstone samples before and after modification, it was found that the liquid limit of the modified mudstone samples decreased by 7.874%, while the plastic limit increased by 9.499%.

(2) Using a method with controlled variables, the laws of the electrolyte solution and electrode materials on the shear strength of modified mudstone were investigated. It was found that the type of ions added to the electrolyte solution affected the cementation strength of the soil. The concentration of ions in the electrolyte solution affected the electro-osmotic efficiency and the amount of residual ions in the soil after electrolysis. The best solution to match the mudstone samples in the test was an $AlCl_3$ solution with a mass fraction of 10%.

(3) Three common electrode materials were chosen for electrochemical modification tests: graphite, iron, and aluminum. The electrolytically oxidizable metal electrodes (iron and aluminum) produced passivation, resulting in higher water content in the modified mudstone specimens compared to the graphite electrode group. The internal friction angle of the graphite electrode-modified mudstone specimen was 26.7° and the cohesion was 34.4 kPa, which was nearly three times that of the original mudstone specimen with 15% water content. For real-world applications involving electrochemical changes to mudstones in the study area, it is suggested to use electrodes made of non-metallic materials and 10% $AlCl_3$.

Author Contributions: Conceptualization and methodology, X.C.; writing—original draft preparation, X.C. and L.H.; writing—review and editing, X.C. and G.S.V.T.; validation, G.S.V.T.; formal analysis, L.H.; data curation, J.K.; supervision, H.Y. and L.H. All authors have read and agreed to the published version of the manuscript.

Funding: This work was financially supported by the National Natural Science Foundation of China (No. 52374144), Natural Science Foundation of Liaoning Province (CN) (Grant No. 2022-KF-23-10), and Xinjiang Uygur Autonomous Region Science and Technology Major Program (No. 2023A01002).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The datasets analyzed during the current study are available from the corresponding author on reasonable request.

Conflicts of Interest: The authors declare no conflicts of interest.

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