



Article Response of High Swelling Montmorillonite Clays with Aqueous Polymer

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Abstract: Expansive clays containing mineral montmorillonite exhibit swelling and shrinkage due to variations in the moisture content, leading to significant distresses. There has been a growing interest in chemical and polymer additives treated for high swelling montmorillonite clays in recent years. However, limited attention has been paid to the effect of polyacrylamide on the soil's swelling behavior. Moreover, nontraditional methods of the soil treatment are applied for the rapid stabilization of soil. In this article, polyacrylamide polymer is used as an additive to expansive clays to control the swelling phenomenon. Three different percentages—2.5%, 5%, and 7.5%—of polymer are blended with oven-dried soil to determine Atterberg limits, compaction features, and swelling characteristics. Additionally, electrical impedance measurement is conducted on treated soil samples with different moisture contents. The electrical resistance of soils and polymer-treated soils is measured based on the electrical resistivity correlation of soils. Tests results for soils stabilized with polyacrylamide show that swelling is significantly reduced with increasing the additive content. Moreover, the addition of polymer improves resistivity of soil. Aqueous polyacrylamide can be utilized as an effective stabilization additive to enhance properties of expansive clays.

Keywords: polyacrylamide; electrical resistivity; expansive clays; swelling; shrinkage; sustainable material

1. Introduction

Expansive clay subgrades frequently severely impact the design and execution of infrastructures, especially highways [1,2]. The seasonal changes in different seasons' moisture that cause the expansive clays to regularly expand and contract in volume and negatively impact the service life of roadways are well known [3,4]. Highway engineers must usually decide between: (i) designing within the expansive subgrade's constraints, which mainly entails over-designing the topmost paving layers [5]; or (ii) trying to minimize the potential for swelling and shrinkage through soil stabilization to meet engineering requirements [6]. Since the first is not always feasible monetarily, logistically, or both, the latter is frequently chosen. Regular maintenance may be necessary to maintain the final performance for any choice. Soil stabilization is physically and chemically modifying a natural soil's composition to satisfy engineering criteria [7,8]. Historically, calcium-based binders such as Portland cement, calcium chloride, and lime have been used to stabilize expansive soils [9]. Introducing these additives to the soil water complex sets off a series of short- and long-term chemical reactions that promote the flocculation or aggregation of clay particles, considerably enhancing important soil properties such as strength, stiffness, permeability, compressibility, swelling and shrinkage [10]. Even though they are efficient at stabilizing materials, energy-intensive materials such as Portland cement and lime are not environmentally friendly because they



Citation: Panda, G.P.; Bahrami, A.; Nagaraju, T.V.; Isleem, H.F. Response of High Swelling Montmorillonite Clays with Aqueous Polymer. *Minerals* 2023, *13*, 933. https:// doi.org/10.3390/min13070933

Academic Editors: Mukuna Patrick Mubiayi, Adolph Anga Muleja and Thabo Falayi

Received: 12 May 2023 Revised: 29 June 2023 Accepted: 30 June 2023 Published: 13 July 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). emit large amounts of carbon during application [11,12]. This disadvantage emphasizes the need to reduce dependence on these binders.

In this context, substituting energy-intensive components with more environmentally friendly industrial and agricultural byproducts is a common solution [13,14]. In terms of geotechnical performance and sustainability, polymers, resins, and sulfonated oils are promising substitutes [15]. Non-traditional additives include enzymes, liquid polymer, resin, acids, silicate, ions, and lignin derivatives. Tingle and Santoni [16] summarized that non-traditional additives could include salt, acids, enzymes, lignosulfonates, petroleum emulsion, polymer, and tree resin. Clays are made of negatively charged aluminosilicate, which is kept together by cations. The important characteristic property of clay is its ability to absorb water between the layers. Swelling strongly depends upon the molecular packing of intercalated water. Soil with smectite swell, such as montmorillonite, swells by moisture absorption and shrinks when it loses moisture [17–20].

Polymer stabilization of soil, one of the non-traditional methods to improve soil properties, is widely applied for the soil treatment [21–24]. The different treatment method has another way of stabilizing clay. Sulfonated oils typically rely on hydrogen ion penetration into the clay lattice, which alters the clay structure by reducing the water-holding capacity. Smectite clay, which exhibits high swelling when treated with potassium ions, shows less swelling [25]. Potassium ions enter the clay lattice and make it less active [26]. Ammonium compounds are useful ion exchangers. Thus, they reduce the activity of clay. Inhibition of clay swelling requires materials that can prevent swelling of the interface layer and bond with the surface of layers. Aqueous polymers are best suited to coat clay particles, and several researchers have employed this technique to control dust, cracks in soil, and erosion [27-30]. Another advantage of the polymer treatment is that its adsorption onto clay is irreversible [30]. The addition of polymers to the soil water medium can cause the flocculation of the clay particles by triggering the appropriate clay–polymer interaction mechanisms, such as charge neutralization, van der Waals or ionic bonds, and cationic bridging for cationic, neutral, and anionic polymers, respectively [31]. Polyacrylamide appears to have a variety of beneficial soil stabilizing characteristics and hence merits more research among the myriad commercially produced and easily accessible polymeric stabilizers [32]. Polyacrylamide is the name given to a class of synthetic polymers made from acrylamide monomers; they are hydrophilic (and thus water-soluble) in nature and can be produced in anionic, neutral, or cationic forms [33]. In several industries, polyacrylamide-based additives have been successfully used. These applications include their use as a flocculant in sludge dewatering and water treatment processes and their adoption in the agriculture industry to improve soil water retention during drought conditions [34]. Applications for polyacrylamide in the geotechnical field include dewatering mining tailings, improving soil compaction effectiveness, reducing desiccation-induced clay cracking, boosting shear strength, and controlling seepage and erosion [30]. Despite being encouraging, the results from these experiments, particularly for expansive clays, still need to be improved to utilize polyacrylamide as an efficient binder in the expansive clays [31]. Factors such as porosity, ionic concentration of the pore fluid, the composition of solids, degree of saturation of soil, particle shape, and orientation affect the electrical properties of soil. Many researchers have attempted to establish a correlation in electrical properties due to changes in density, moisture content, and temperature [32,33]. Resistivity is a material property independent of the media's shape. The current frequency affects the electrical resistance and dielectric constant of soil in that zone. Measuring the electrical resistivity is an excellent monitoring tool with which to envisage the change inside the soil structure [34]. In addition, data that are currently available on polyacrylamide stabilized soils are mainly restricted to standard geotechnical laboratory tests such as Atterberg limits and strength characteristics, which, while valuable, have not yet been sufficient to inspire confidence in polyacrylamidetreated expansive clays for widespread use in highway projects. For example, in the context of expansive soils, a thorough review of the existing literature reveals that the swelling phenomenon and electrical resistivity of polyacrylamide-blended expansive soils, when

subjected to field conditions, have not yet been examined, implying the need for additional research to better understand polyacrylamide clay blends and, as a result, its true stabilization potentials and/or limitations [34,35]. The stabilization of an expansive soil from the United States is being investigated in this study as a future application of an anionic polyacrylamide-based material as an environmentally sustainable material. The main goals are to examine how applying polyacrylamide at various polyacrylamide percentages affects the behavior of soil in terms of swelling.

2. Materials and Methods

2.1. Soils

Clays were selected based on their high swelling behavior, which presents a significant opportunity for the adsorption of polymer molecules onto both their interlayer surfaces and particle surface. Laboratory tests were conducted to evaluate the impact of polymer solution on the compaction, swelling, and plasticity characteristics of three soils that were collected from Houston, United States. To create polymer-blended soils, field clayey soil samples were used. The geotechnical properties of all three types of clay were assessed based on ASTM Standards. The properties of clays are presented in Table 1. After analyzing the gradation and plasticity characteristics, it was determined that Soil-A was classified as CL, while Soil-B and Soil-C were identified as CH using the unified soil classification system (USCS). Table 1 demonstrates that Soil-B exhibits more swelling than the other soils, Soil-A and Soil-C, based on the free swell index values.

Table 1. Test methods and properties of soils.

Parameter	Test Method	Soil-A	Soil-B	Soil-C
Specific gravity	ASTM D854	2.67	2.66	2.66
Liquid limit (%)	ASTM D4318	36	86.41	53.91
Plastic limit (%)	ASTM D4318	18	22.5	18.5
Free swell index (%)	ASTM D4546-14	66	95	83
Optimum moisture content (%)	ASTM D698	22	18	17
Maximum dry density (g/cc)	ASTM D698	2.01	1.83	1.95
USCS classification	ASTM D2487	CL	СН	СН

The X-ray diffraction analysis (XRD) technique was used to determine the mineralogical properties of the expansive soil. Figure 1 depicts the analysis of the XRD patterns of Soil-A, Soil-B, and Soil-C. The data obtained from the XRD analysis indicate a distinctive peak for quartz, with the 20 position at 21.42°, 21.81°, 25.42°, 38.16°, 49.18°, 60.12°, and 69.45° matching the quartz spectra. The peaks at 20 values of approximately 19.45°, 23.16°, and 29.12° correspond to montmorillonite. Meanwhile, calcite was identified at 30.12°, 37.48°, and 40.14°, and Hematite at 26.18°, 33.14°, and 41.27°. Moreover, the common peak at 38.1° was observed for quartz, hematite, and calcite, whereas the illite clay mineral diffraction peak was seen at 20.85°. Based on the XRD results, it was concluded that the expansive soil contains a considerable amount of quartz while also containing montmorillonite, illite, hematite, and calcite.

2.2. Polymer Treatement

Commercially available polyacrylamide polymer was utilized in this study. The polymer solution was prepared with different percentages of polyacrylamide polymer. The effective portion of polymer added to clay soil was 2.5%, 5%, and 7.5% by weight of dry soil. Soil was pulverized and made dry and kept in oven; then, polymer was added to it. For proper dispersion of polymer over pulverized soil, the polymer solution was mixed thoroughly and allowed to dry for one day at room temperature. To prepare the polymer

solution, the following mix of polymer was employed. The polymer solution was prepared by mixing Mix-1 and Mix-2 (Table 2).



Figure 1. XRD patterns of expansive clays.

Table 2. Polymer preparation mix.

Ν	fix-1	Mi	ix-2
AV-100	2.5%, 5%, 7.5%	AV-102	0.5%
AV-101	0.5%		
Water	50 mL	Water	50 mL

This polymer solution was applied on the pulverized soil. The preparation process is straightforward. Firstly, clay particles are mixed with a dispersant polyacrylamide aqueous solution. Given the negatively charged surfaces of the clay particles and positively charged edges, the polyacrylamide anions are expected to adsorb onto the positively charged edges of the clay particles. Consequently, the clay particles are dispersed in water, and the resulting clay dispersion is gradually added to a polyacrylamide aqueous solution, which is then stirred to ensure proper mixing. Anionic polyacrylamide is also anticipated to adsorb onto the positively charged edges of the clay particles. A schematic view of the polyacrylamide treatment of clays is presented in Figure 2.

2.3. Testing

A standard compaction test was conducted to obtain the maximum dry density at the optimum moisture content. All samples were subjected to standard proctor compaction tests. The effect of polymer addition on the optimum moisture content was noted. The standard compaction test was done on 0%, 2.5%, 5%, and 7.5% polymer added to clay soil.

The swelling test was performed as per the ASTM standard D4546-14. The swelling test was carried out for soils and polymer-treated soils. The shrinkage–swelling test quantitatively determines the expansive potential of undisturbed and remolded clay soils. The expansion index (*EI*), according to ASTM D4829-11, is defined as Equation (1):

$$EI = \frac{\Delta H_1}{H} \tag{1}$$

where *H* is the initial height of the sample (in or mm), ΔH_1 is change in the height of the sample (in or mm). Meanwhile, the soil classification is done as per the value of the expansive index using a hyperbolic model (Equation (2)):

$$\delta = \frac{t}{A + B \times t} \tag{2}$$

where δ is deflection (in) at any given time (min), *t* is time (min), and model parameters, *A* and *B*, were used to predict the behavior of *EI* of soils.



Figure 2. Schematic view of polymer-treated clay mechanism.

The electrical resistivity of soil was measured using ASTM standard G187-18. The electrical resistivity of soil is a crucial parameter that reflects the soil's ability to resist the flow of the electrical current. It is a sensitive parameter that various factors, such as changes in moisture content, temperature, pore fluid conductivity, soil texture, and mineralogy, can influence. Therefore, treated soils undergoing stabilization techniques such as mixing, chemical, or thermal treatment may have different electrical resistivity values compared to untreated soils. These differences can occur owing to the changes in the soil structure, pore fluid conductivity, and chemical properties resulting from the treatment process. Consequently, understanding the electrical resistivity of treated soils is essential for assessing their suitability for various engineering applications [36–38]. Vipulanandan and Amani [39] studied different possible equivalent circuits models for composite materials with two probe measurements. For soil studies, Case-2 material behavior was selected. Case-2 behavior is observed when the capacitance behavior of Case-1 material is assumed to be negligible (Figure 3). The total impedance of the equivalent circuit for Case-2 (Z_2) is when the frequency of the applied signal is very low (Equation (3)):

$$Z_{2}(\sigma) = R_{b}(\sigma) + \frac{2R_{c}(\sigma)}{1 + \omega^{2}R_{c}^{2}C_{c}^{2}} - j\frac{2\omega R_{c}^{2}C_{c}(\sigma)}{1 + \omega^{2}R_{c}^{2}C_{c}^{2}}$$
(3)

When the frequency of the applied signal is very low, ($\omega \rightarrow 0$) $Z_2 = R_b + 2R_c$; and when it is very high, ($\omega \rightarrow \infty$) $Z_2 = R_b$.



Figure 3. Case-1 and Case-2 impedance models for blended materials.

The material response trends employing the two-probe method for monitoring are well known [34]. Figure 4 displays the mechanism of the two-probe method. Testing the polymer-treated clays (composites) revealed Case-2; hence, the overall material's characteristics can be conveyed through resistivity and characterized using the two-probe method at a frequency of 300 kHz. The main goal was to investigate the influence of adding polyacrylamide to high-swelling clays.



Figure 4. Mechanism of two-probe method.

3. Results and Discussion

3.1. Index Properties

After the polymer treatment onto the dry soil, the liquid limit (LL) and plastic limit (PL) tests were repeated on the treated soil. The tests results are summarized in Table 3. LL and PL of Soil-A, Soil-B, and Soil-C indicated a similar trend. With the addition of 2.5%, 5%, and 7.5% polymer to Soil-A, reduction of LL was observed by 4.16%, 9.72%, and 15.21%, respectively. The addition of 2.5%, 5%, and 7.5% polymer to Soil-B reduced LL by 20.36%, 35.13%, and 40.93%, respectively, and the same polymer addition to Soil-C decreased LL by 7.75%, 12.38%, and 16.78%, respectively. Similar trends were observed in the case of PL. Reduction in LL and

PL leads to a decrease in PI. PI was reduced by 6.1%, 13.8%, and 23.6%, respectively for Soil-A, 24.2%, 42.3%, and 48.3%, respectively for Soil-B, and 16.09%, 24.5%, and 33.88%, respectively for Soil-C when 2.5%, 5%, and 7.5% polymer were respectively added to these soils.

Percentage of Polymer Content		Soil-A			Soil-B			Soil-C	
	LL	PL	PI	LL	PL	PI	LL	PL	PI
0	36	18	18	86.41	22.5	63.91	53.91	18.5	35.41
2.5	34.5	17.6	16.9	68.81	20.5	48.31	47.21	17.5	29.71
5	32.5	17	15.5	56.05	19.5	36.55	43.21	16.5	26.71
7.5	30.5	16.75	13.75	51.04	18	33.04	39.41	16	23.41

Table 3. Index properties of soil after polymer treatment.

From the Atterberg tests, it was clearly witnessed that the polymer addition affected the index properties of soil (for instance Soil-B), as can be seen in Figure 5. The addition of polymer coats the clay particles and inhibits their ability to coagulate. A coat formation occurs on top of the clay particles. Based on PI, the soil expansivity can be classified using Holtz–Gibbs and Chen criteria (Table 4).



Figure 5. Plasticity characteristics of Soil-B blended with various dosages of additive.

Table 4. E	Expansivity	characterization	based on	Holtz-0	Gibbs and	Chen	criteria
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Soil-A	Holtz–Gibbs Criteria	Chen Criteria	Soil-B	Holtz–Gibbs Criteria	Chen Criteria	Soil-C	Holtz–Gibbs Criteria	Chen Criteria
0%	Low	Low	0%	Very high	Very high	0%	Very high	Very high
2.5%	Low	Low	2.5%	Very high	Very high	2.5%	Medium	Medium
5%	Low	Low	5%	Very high	Medium	5%	Medium	Medium
7.5%	Low	Low	7.5%	Medium	Medium	7.5%	Medium	Medium

3.2. Compaction Characteristics

The compaction test of soil samples before and after the polymer treatment has been analyzed, as illustrated in Figure 6. In the case of Soil-A, the maximum dry density for

untreated samples and treated samples with 0%, 2.5%, 5%, and 7.5% polymer by weight was 2.01 (g/cc), 1.92 (g/cc), 1.85 (g/cc), and 1.79 (g/cc), with the optimum moisture content of 22, 25, 29, and 32, respectively. In the case of Soil-B, the maximum dry density of soil treated with polymer at 0%, 2.5%, 5%, and 7.5% was 1.83 (g/cc), 1.79 (g/cc), 1.74 (g/cc), and 1.63 (g/cc) with the optimum moisture content of 18%, 24%, 28%, and 33%, respectively. For Soil-C, the maximum dry density was 1.95 (g/cc), 1.85 (g/cc), 1.78 (g/cc), and 1.72 (g/cc) with the optimum moisture content of 17%, 19%, 22%, and 24%, respectively. The polymer

3.3. Swelling Behavior of Treated Soils

content was observed.

The swelling index test was carried out in accordance with ASTM D4829-11. The sample for the swelling index test was performed keeping the dry side of the optimum moisture content condition for 7 hours. Using the hyperbolic model, the expansion of clay was predicted with actual results. As expected, the maximum deflection was witnessed in the case of untreated soil, and the swelling behavior was reduced considerably with the addition of polymer. Figure 7 depicts the swelling behavior of treated clays. In the case of Soil-A, the addition of 2.5%, 5%, and 7.5% polymer reduced the swelling index by 65%, 77%, and 88%, respectively. For Soil-B and Soil-C, the addition of 2.5%, 5%, and 7.5% polymer resulted in the reduction of the swelling index as 62%, 72%, and 83%, and also 67%, 78%, and 89%, respectively. The swelling behavior of soil is influenced by the type and percentage of clay minerals present in clay. Polymer-treated soil demonstrated less swelling as polymer particles covered the soil particles and prevented it from swelling. Additionally, the ionic nature of polymer made it effective in controlling the swelling potential of clayey soils. Using the hyperbolic model, the swelling behavior of soil was modeled. The model parameters are listed in Table 5. The model parameters can be used to predict the swelling behavior of soil. For Soil-A, Soil-B, and Soil-C, it was observed that with the increment of polymer, the model parameters also increased.

treatment reduced the density of treated soil, and an increase in the optimum moisture

3.4. Electrical Impedance of Treated Soils

The electrical properties of soil are measured with different polymer percentages and moisture contents (Table 6). The impedance and resistivity of all soils with varying moisture contents were measured and the highest was found to be Case-2. AC measurement was used to measure the change in electrical properties of soil due to the polymer additions. The electrical impedance curve was plotted for soil treated with polymer at 10% moisture content. The impedance curves displayed a definitive trend. The impedance curves of Soil-A, Soil-B, and Soil-C with different percentages of polymer are shown in Figure 8.

In the soil samples, it was seen that the addition of polymer increased resistivity of soils. Similar trends were observed for all soils. With increasing the moisture content, resistivity of soils dropped. Increasing the moisture content in polymer-treated soils reduced resistivity. The polymer treatment of soils increases resistivity by coating the soil particles with polymer. Measuring resistivity of soils after the polymer treatment can be used as a quality control measure in field applications. The addition of polymer to soils enhanced the electrical resistivity. All soils indicated a similar pattern with the increased polymer content. Electrical measurement can be used as a method for the detection and measurement of polymer-treated soils.

3.5. Micro-Structural Analysis

The SEM images presented in Figures 9 and 10 illustrate the results of untreated Soil-B and Soil-B blended with 7.5% polyacrylamide, respectively. Upon comparison of different magnifications, it is evident that the addition of the aqueous polymer resulted in the formation of numerous fiberous and reticular structures that are entwined around the clay particles. This phenomenon enhanced the connection between the clay particles, leading to a stronger spatial structure system. As a result, the scattered clay particles transformed into

a compact clay particle spatial structure system. The polymer not only adsorbed onto the surface layer of clay but also plugged clay with filiform linkages [40,41]. Consequently, the aqueous polymer solidified, clustering the clay particles and strengthening the chemical bonds established by the clay–polymer interaction. The primary reason for this spatial structure system was the firm bonding of polyacrylamide molecules with the clay particles, resulting in a denser skeleton that significantly improved the strength and stability of the stabilized clay.



Figure 6. Compaction characteristics of various soils blended with additive.



Figure 7. Swelling behavior of various soils blended with additive.

Several previous studies have explored the microstructural characteristics of soils treated with polyacrylamide through qualitative analysis of SEM images. For example, Amiri et al. [42] demonstrated the ability of polyacrylamide to improve particle cohesion, while Lie et al. [43] found that polyacrylamide adsorption resulted in a glue-like bonding between soil particles. Lentz [44] used microstructure images to demonstrate that clayey soils were more prone to aggregation than silty gravel ones. Similarly, Huang et al. [33] utilized an anionic polymer to treat a gravel, sand, silt, and clay mixture and observed that the ionic mechanism facilitated aggregation and microscopic density enhancement. These studies suggested that the strong attractions of heterogeneous charges are the most

important factor in stabilizing the soil structure for negatively charged polymers. Therefore, soils with high clay content, high cation exchange capacities, and rich divalent cations are more suitable for stabilizing polyacrylamide.

Soil Type	Polymer Addition (%)	Α	В	R ²
Soil A	0	11.86	0.58	0.99
Soil A	2.5	26.75	1.74	0.99
Soil A	5	41.57	2.55	0.98
Soil A	7.5	148.28	4.83	0.98
Soil B	0	0.81	0.18	0.98
Soil B	2.5	8.70	0.50	0.98
Soil B	5	18.42	0.62	0.99
Soil B	7.5	15	1.09	0.97
Soil C	0	4.40	0.25	0.95
Soil C	2.5	42.10	0.79	0.98
Soil C	5	67.01	1.17	0.99
Soil C	7.5	146.81	4.85	0.96

Table 5. Model parameters of swelling soil.

Table 6. Electrical resistivity of soil treated with polymer at different moisture contents.

Soil Type	D (Electrical Resistivity (Ω .m)					
	of Polymer Content	10% Moisture Content	20% Moisture Content	30% Moisture Content	40% Moisture Content		
	0	35.33	8.38	5.71	3.22		
C - 1 A	2.5	528.51	17.61	7.84	3.65		
5011-A	5	650.94	20.05	10.56	7.27		
	7.5	786.96	23.66	12.16	8.45		
	0	43.80	7.40	4.40	1.80		
Cail D	2.5	521.20	44.30	9.50	3.40		
5011-В	5	861.30	56.00	11.40	5.30		
	7.5	1185.70	72.00	13.30	6.10		
Soil-C	0	27.41	7.78	6.51	2.82		
	2.5	687.11	30.10	10.96	4.29		
	5	1177.88	41.82	12.16	5.95		
	7.5	1468.75	60.38	16.12	7.50		



Figure 8. Impedance curves of various soils blended with different percentages of additive keeping 10% moisture content.



Figure 9. SEM images (various magnifications) of untreated Soil-B.



Figure 10. SEM images (various magnifications) of Soil-B blended with 7.5% polyacrylamide.

4. Conclusions

The polymer treatment of soil remarkably affected the index properties of soil. The polymer treatment led to a reduction in PI of soil.

The polymer treatment of soil influenced the dry density of soil and the optimum moisture content of treated soil. A regular trend was found in the case of the dry density and the optimum moisture content of soil. Polymer particles coated soil particles, which led to an increased density at higher optimum moisture content.

The swelling test on polymer-treated soil revealed that the polymer treatment was extremely effective in reducing the modified swelling index of soil. In the polymer treatment, the addition of 2.5%, 5%, and 7.5% polymer to Soil-A reduced swelling by 65%, 77%, 88%, respectively; to Soil-B, a reduction of 62%, 72%, and 83% was respectively observed in swelling; and to Soil-C, a reduction of 67%, 78%, and 89% was respectively witnessed.

The electrical measurements indicated that all soils treated with polymer exhibited Case-2 behavior. The addition of polymer improved the electrical impedance curve. Similar trends were seen for Soil-A, Soil-B, and Soil-C.

The resistance measured through impedance can be utilized to determine resistivity of soil, which is a material property. Resistivity of soil increased with the addition of polymer at various moisture contents. Furthermore, resistivity can be employed as a quality control tool in field applications.

Author Contributions: G.P.P.: Conceptualization, Methodology, Investigation, Writing—original draft preparation. A.B.: Conceptualization, Methodology, Investigation, Validation, Formal analysis, Resources, Writing—original draft preparation, Writing—review and editing, Project administration. T.V.N.: Conceptualization, Methodology, Investigation, Validation, Writing—original draft preparation. H.F.I.: Validation, Formal analysis. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: Not applicable.

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

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