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Perspectives of Using Lignin as Additive to Improve the Permeability of In-Situ Soils for Barrier Materials in Landfills

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Abstract: Very often, in-situ soil does not meet the requirements for landfill barriers; therefore, it is necessary to purchase the material from quarries. An increasing number of by-products have been proposed as alternative landfill barrier materials. The present study investigated the performance of two soils of Central Italy (alluvial and volcanic soils) with an organosolv lignin (sulfur-free lignin (SFL)), a widespread by-product in the world. Laboratory investigations indicated that the volcanic soil mixed with 10% in weight of lignin did not reach the permeability value required for landfill bottom liners, also showing high compressibility. On the contrary, the addition of 20% to 30% lignin to the alluvial soil reached the permeability value recommended for the top-sealing layer of landfills: scanning electron microscope analysis indicated that the improvement was due mainly to the physical binding. Large-scale investigations should be carried out to evaluate the long-term performance of the mixtures. The increasing production of organosolv lignin worldwide gives this by-product the opportunity to be used as an additive for the realization of the top-sealing layer. The approach can save the consumption of raw materials (clayey soils from quarries), giving lignin a potential new field of application and recovering in-situ soils.

Keywords: landfill barrier; permeability; by-product; lignin; fine-grained soils

1. Introduction

The use of recycled materials for improving physico-mechanical and hydraulic properties of compacted earth structures is nowadays one of the main targets in engineering and environmental geology. According to reference [1], when waste products or by-products (fly ash, rice husk ash, pulverized biomass, etc.) are used in place of raw materials (e.g., soils from quarries), natural resources and energy are preserved, reducing costs for disposals. This approach aligns with sustainable development, which requires the simultaneous achievement of environmental, economic, and social sustainability [2]. These concepts are particularly important in the construction of landfills, which require low-permeable soils (or equivalent materials) for bottom liners and final top barriers (capping system). The landfill EU directive [3] prescribes the minimal requirements for the construction of compacted mineral barriers, which have been enacted in some EU member states such as Italy, Germany, and the United Kingdom [4]. According to the Italian regulation [5], to ensure the landfill containment and to prevent gas emission and infiltration of rainfalls into the waste, compacted soil for non-hazardous waste should have the following properties:

- a bottom liner of compacted soil ≥ 1 m with hydraulic conductivity (k) $\leq 1 \times 10^{-7}$ cm/s and
- a top-sealing layer of compacted soil ≥ 0.5 m with hydraulic conductivity (k) $\leq 1 \times 10^{-6}$ cm/s.

The implementation of landfill barriers requires several tons of clayey soils per m². Soils that meet the hydraulic conductivity requirements are often not available locally; therefore, it is necessary to purchase the clayey soils extracted from quarries. This increases operational and development costs of containment landfills affecting sustainable development strategy in terms of its economic and environmental impacts. An increasing number of diverse secondary materials (residues from steel industry and incineration, from paper industry, etc.) have been suggested as alternative landfill barrier materials [6] to preserve natural resources, giving secondary materials a new field of application. In other words, one of the sustainable and economical solutions for landfill barriers could be the development of liner material with the required permeability by using local soil itself with some additives [7]. Among the by-products, which can be used as additives, lignin is a promising material because it is the most abundant aromatic biopolymer in nature and is the main by-product of the paper and pulp industries and bio-refinery processes [8]. According to references [9,10], different types of lignin are available which can be grouped in two main categories: sulfur-bearing (lignosulfonates, kraft lignins, and hydrolyzed lignins) and sulfur-free lignin (organosolv, soda lignin, and lignin from second-generation biorefinery process). Lignin shows a complex and heterogeneous chemical structure consisting of lignans subunits produced by enzyme-mediated radical polymerization of three monolignols: *p*-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol [11]. Lignin has interesting properties such as biodegradability, renewability, antioxidant activity, and UV shielding capability. It has attracted attention for its valorization and application in different fields [12] including absorbent for the selective removal of metals from water and other materials such as surfactants, pesticides, and phenols [13–18].

The annual production of lignin from different biomass separation processes currently accounts for about 70 million tons. As recently reported by reference [19], "millions of tons of lignin are produced by the paper pulping industry every year and are mostly (>95%) treated as waste or used in low-value applications such as fuels. Commercially used lignin is typically produced as lignosulfonates or kraft lignins". With respect to other types of lignin, the current production of organosolv lignin is not available [10] even though organosolv-like lignin production is increasing as cellulosic ethanol has been promoted as a substitute for fossil fuels [20]. The production of this type of lignin is expected to increase further, considering green energy policies.

Sulfur-bearing lignins, such as lignosulphonates and kraft lignins, have been used in the last decades as soil stabilizer, an important technique to enhance soil properties without damaging the environment [21]. Among the physico-mechanical properties of silty and clayey soils, the unconfined compressive strength, plasticity index (PI), and durability are the most affected by the addition of lignin [22–26]. As reported by reference [25], lignin has the most influence on PI and swelling potential of expansive soils compared to the other waste materials such as rice husk powder and rice husk ash.

According to reference [27], sulfur-free lignins (SFLs) are not well studied even though they represent a large portion of lignin waste. As reported by reference [28], SFL could yield comparable strength to fly ash treatment. Moreover, the soil stabilized with SFL may provide excellent moisture and frost resistance [24,29,30].

Compared to sulfur-bearing lignin, in-depth studies should be carried out to explore other areas of SFL application. Despite considerable progress in the development of new lignin-based materials, the potential use of lignin in landfill liners and capping systems, as an additive of in-situ soils, requires more focused studies. The adding of organosolv lignin to in-situ soils may improve the hydraulic properties of compacted materials, making them potentially useful in the realization of landfill bottom liners and capping systems. This hypothesis needs to be verified, investigating-at laboratory scale the effect of lignin on some soil types. For this purpose, two fine-grained soils widely outcropping in Central Italy were selected which do not fulfill the permeability requirements for landfill barriers. The main objective of the study was to demonstrate which soil was improved by the adding of the

organosolv lignin. This knowledge is not currently available in the literature, and it represents the first step for further evaluations addressed to the use of the by-product as an additive in the containment systems of landfills.

2. Materials and Methods

2.1. Organosolv Lignin

The lignin studied was supplied from Chemical Point (Oberhaching, Germany) and derived as a by-product from the sulfur-free organosolv pulping process containing 25% moisture, ≤0.5% residual sugar, and ≤5.0% ash. Lignin powder shows a brown color, pleasant smell, and water insolubility. In order to study the chemical structure of lignin, spectroscopic vibrational analysis was performed by attenuated total reflection infrared (ATR-IR) measurement. The analysis was carried out at room temperature with a Perkin−Elmer Spectrum One spectrometer (Waltham, MA, USA) equipped with an ATR-IR cell and the IR spectra were recorded by averaging 32 scans with a resolution of 4 cm^{-1}. The recorded infrared spectrum (Figure 1) showed the typical peaks of sulfur-free organosolv lignin, in accordance with data previously reported by reference [31]: C-H bending of methyl and methylene groups (1465.32 cm^{-1}), the C=O (1601.98 cm^{-1}), C-H (2936.93 cm^{-1}), and O-H stretching (3420.16 cm^{-1}) appeared to be the most representative vibrational bands.
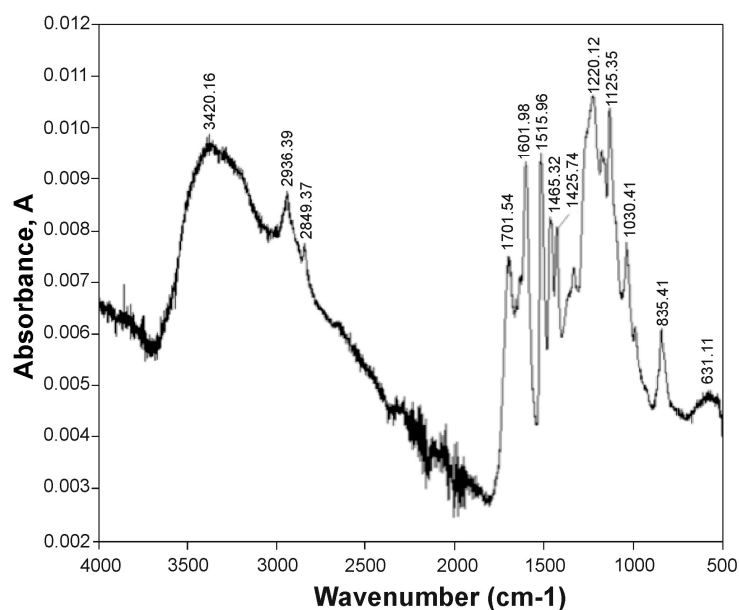

Figure 1. ATR FT-IR spectra (Attenuated Total Reflection Fourier Transform Infrared Spectroscopy) of sulfur-free organosolv lignin.

The qualitative and quantitative analyses of characteristic OH moieties were performed by the phosphorous nuclear magnetic resonance (^{31}P-NMR) [32]. We dried 15 mg of lignin by the oven for 24 h at 70 °C and then dissolved in pyridine/CDCl$_3$ (300 µL; 1.6/1.0 *v/v*), followed by addition of chrome (III) acetylacetonate solution (50 µL, 11.4 mg/mL) as a relaxing agent. Finally, freshly prepared phosphitylation reagent 2-chloro-4,4,5,5-tetramethyl-1,3,2-dioxaphospholane (200 µL) was added under magnetic stirrer and gentle heating at room temperature for 3.0 h [33]. ^{31}P-NMR analysis was performed in the presence of cholesterol as an internal standard using a Bruker 400 MHz apparatus (USA, Billerica).

The total amount of OH groups was evaluated by comparing the ^{31}P-NMR integral area of each specific signal with respect to the peak of the internal standard [32]. Figure 2 reports the characteristic NMR signals of the main lignin sub-units of the polymer; Table 1 shows the functional hydroxyl group distribution for the organosolv lignin. The OH *p*-hydroxyphenyl moieties largely prevailed over the

other possible phenolic groups. The ^{31}P-NMR spectra showed high peaks of carboxylic acids groups, confirming the hydrophobic properties of lignin. Moreover, the ATR FT-IR spectra (Figure 1) were comparable with those of the hydrophobic SFL lignin used by reference [31] to improve the engineering behavior of soils.
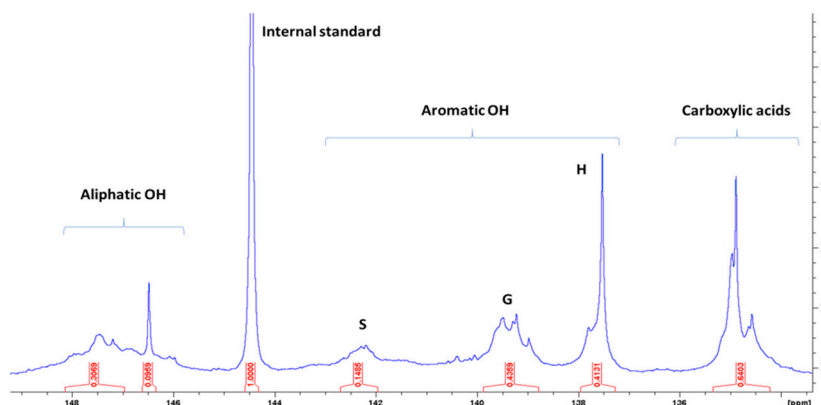

Figure 2. Quantitative phosphorous nuclear magnetic resonance (^{31}P-NMR) spectra and signal assignment of organosolv lignin. Typical lignin chemical shift (ppm): para-hydroxy phenolic groups (H) 138.5–138.8 ppm; guaiacyl groups (G) 138.7–140.1 ppm; condensed phenol groups 140.3–140.5 ppm; syringyl groups (S) 142.0–144.0 ppm; aliphatic hydroxyl groups 146.0–149.0 ppm; and carboxylic acids groups 134.0–135.5 ppm e.g., [32].

Table 1. Functional hydroxyl group distribution (mmol/gram) for organosolv lignin.

Aliphatic-OH	Carboxylic Acid	Phenolic-OH		
		Syringyl	Guaiacyl	p-Hydroxyphenyl
3.99	4.05	1.91	3.81	6.11

2.2. Soils

Two sampling sites (S_A and S_B) were selected in the Tiber River basin (Central Italy, Figure 3).

The site S_A is characterized by sandy silts belonging to the recent fluvial deposits of the upper Tiber River, a NW–SE trending intermountain depression flanked mainly by siliciclastic marine formations (flysch formations). The site was chosen because it is easily accessible to soil sampling, and it is characterized by fluvial deposits widely outcropping in Central Italy [33]. According to a Dynamic Probing Super Heavy test (DPSH) carried on the left bank of the Tiber River, these deposits are about 3 m thick and rest on the siliciclastic bedrock outcropping in the riverbed.

The site S_B is characterized by sandy silt soils originated by the weathering of tephritic-phonolitic and leucite-bearing lava belonging to the Vico Volcanic Complex [34]. The Vico Volcanic Complex is a strato-volcano with a central caldera depression housing Lake Vico. This volcano was active between 419,000 and 95,000 years ago and developed on a graben elongated NW–SE at the intersection with a NE–SW fracture [35]. As for site S_A, the S_B site is easily accessible for sampling; it is not disturbed by human activity and has known mineralogical characteristics of the soil, being located close to the EUR03 sampling site as reported in reference [36].

The mineralogy of the two soils is significantly different. As recently reported by reference [37], the S_A soil is mainly composed of quartz (50%), orthoclase (21%), calcite (15%), and albite (14%). Taking as reference EUR03 soils [28], the mineralogical composition of soils from S_B site can be synthesized as follow: biotite (10%), K-feldspars (39%), leucite (19%), plagioclase (3%), pyroxenes (22%), Fe-oxides (6%), and minor amounts of hydrated halloysite and zeolite minerals. Overall, the mineralogical characteristics of the two materials can be considered as representative of many fluvial-lacustrine and volcanic soils outcropping in Central Italy.
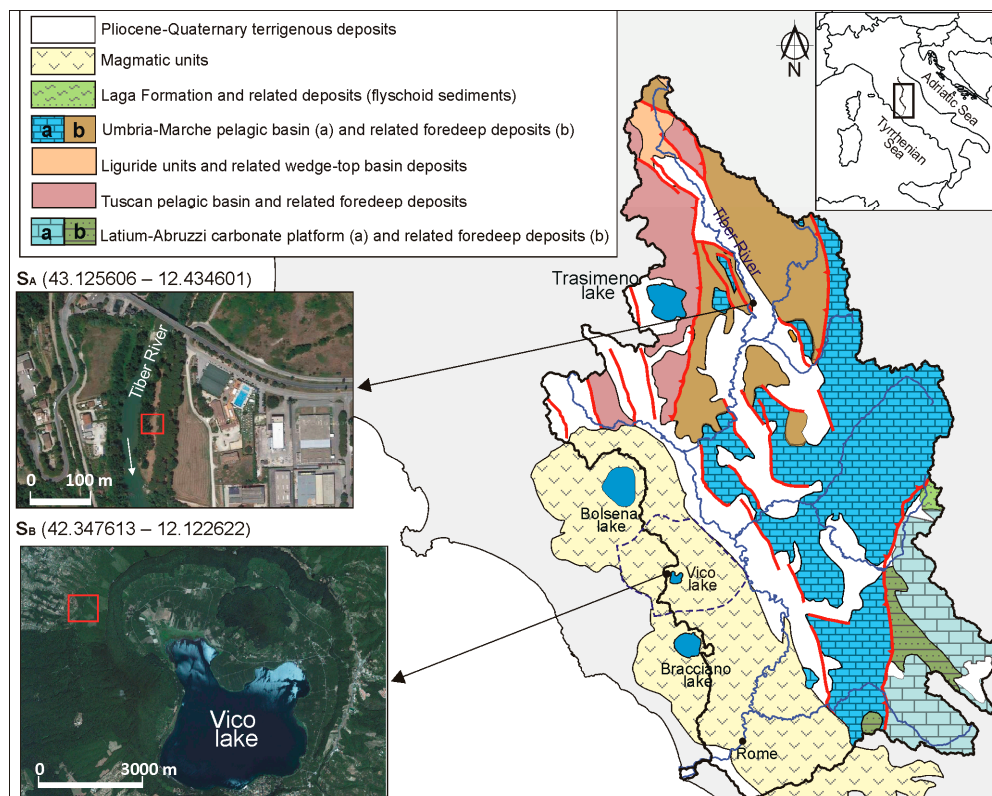
Figure 3. Geological map of the Tiber River basin with location of sampling sites.

2.3. Experimental Procedures and Soil Characteristics

In the construction of landfill liner, one of the main components is the mineral layer, which is selected and compacted to provide leachate containment. Several laboratory tests are carried out on remolded soil samples, useful to understand the suitability of soils for landfill barriers.

Basic soil properties were measured with standard methods, including particle size distribution [38], specific gravity (Gs) [39], standard compaction Proctor test [40], organic matter [41], and Atterberg limits [42]. The fall cone technique was used to determine the Liquid limit (LL) of soils because it is less operator-dependent than the Casagrande cup method [43–46]. Table 2 summarizes the main geotechnical properties of the two soils. Both soils are non-plastic, with fines content higher than 20%, and the fraction of gravels lower than 10%. To check the swelling potential and expansivity, the Free Swell Ratio (FSR, Equation (1)), i.e., "the ratio of the equilibrium sediment volume of 10-g oven-dried soil passing a sieve of 0.425 mm with distilled water (V_d) to that in carbon tetra chloride or kerosene (V_k)" [47], was used.

$$\text{FSR} = \frac{V_d}{V_k} \tag{1}$$

Table 2. Geotechnical properties of soils. CF—clay fraction; Gs—specific gravity; MDUW—maximum dry unit weight; OMC—optimum moisture content; PI—plasticity index; OM —organic matter; N.P.—non-plastic.

Soil	Grain Size (%)			CF (%)	G_s (−)	Compaction Properties		PI (%)	OM (%)
	Fines	**Sand**	**Gravel**			**MDUW (kN/m³)**	**OMC (%)**		
S_A	33.0	67.0	-	3	2.66	17.5	13.2	N.P.	1.5
S_B	48.5	42.7	8.8	8	2.51	11.1	18.4	N.P.	3.0

When FSR ≤ 1, soils are classified as no-swelling with negligible soil expansivity. Moreover, for non-plastic soils (N.P.), like those used here (Table 2), the shrinkage and swelling are negligible (soils are not active according to reference [48,49]).

Hydraulic conductivity measurements were carried out in an oedometer cell (Figure 4a) and in a falling-head permeability cell (Figure 4b), following references [50] and [51] standards, respectively. A scanning electron microscope (SEM) was used to investigate the micropores distribution of the soil. The SEM specimens were prepared at standard Proctor optimum conditions following the same procedures used to compact soil samples into the permeability mold. Images were taken using an FE SEM LEO 1525 ZEISS (Carl Zeiss Microscopy GmbH, Jena, Germany) with the GEMINI field emission column.
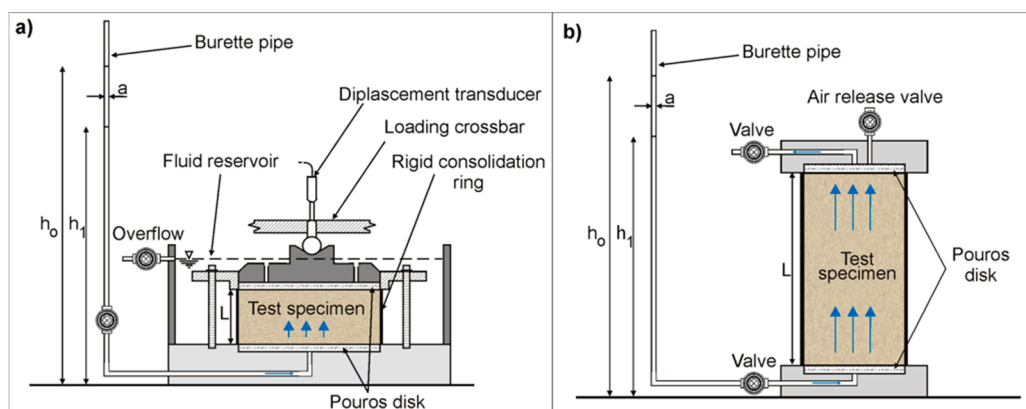

Figure 4. Schemes of equipment used for permeability tests (not to scale). (**a**) Oedometer (fixed-ring type) equipped with a falling-head device. (**b**) Falling-head permeability cell.

An oedometer test (one-dimensional consolidation) is useful to study the stress-deformation process of compacted landfill bottom liners [52]. The soil compressibility is particularly important because the vertical stress produced by wastes can reduce the volume of the landfill barrier, nullifying the overall containment system. As illustrated in the previous section, soil S_B contains some amounts of zeolite—well recognized for its high adsorption capacity for pollutants [53]—making this material potentially suitable for use as landfill bottom compacted liner. Oedometer tests were carried out on soil S_B and soil S_B + 10% of lignin (percent by weight). This amount was chosen with reference to the use of lignin as a soil stabilizer [25,54–56]. Soil and soils/lignin mixture were tested at optimum compaction characteristics (i.e., similar to field application conditions); thus, the materials had different initial dry unit weight and water content due to the standard Proctor compaction.

Samples for the oedometer tests (50 × 20 mm) were collected from the standard Proctor cores. Samples were sealed in a plastic bag to cure in the humidity room where the temperature was maintained at 20 ± 2 °C for seven days before performing oedometer tests. During this period, the water, uniformly distributed in the sample, allowed potential chemical–physical soil/lignin reactions. After this process, samples were placed in the oedometer cell, taking care with the assembly of test specimens into the equipment, avoiding the formation of cracking. Soils were saturated and then subjected to progressive effective stresses (12.5, 25, 50, 100, 200, and 400 kPa): each load increment was applied every 24 h and drainage was allowed from the bottom and top of the samples. According to reference [57], the stress range used simulates a moderate height landfill with up to 30 m of filling. The oedometer cell allowed checking hydraulic conductivity values at the end of each effective vertical stress increment.

Falling-head permeability tests were carried out on samples compacted directly in the permeability mold with a transparent Plexiglas body (150 mm height and 63 mm diameter). The falling-head test is suitable for fine-grained soils and is subject to minimal sources of errors [58–60]. Samples were cured for seven days in a controlled temperature–humidity room. The compaction system included the standard Proctor compaction rammer (2.5 kg) coupled with a brass cylinder (3.0 kg) resting on the soil

layer (Figure 5a). Each specimen was compacted in three layers into the mold until achieving at least 95% of maximum dry unit weight (MDUW) values (required degree of compaction of in-situ soils). The tests were performed at different weight percentages of the lignin in the mixture (0%, 10%, 20%, and 30%). Figure 4b shows the standardized number of blows provided by the compaction system for each sandy silt/lignin mixture with MDUW values obtained by the standard Proctor procedure. According to reference [61], Gs value of pure lignin is about 0.45, affecting both Gs and MDUW values of sandy silt/lignin mixtures: MDUW values decrease as the lignin content in the mixture increases (Figure 5b). Similar results were obtained by reference [27].
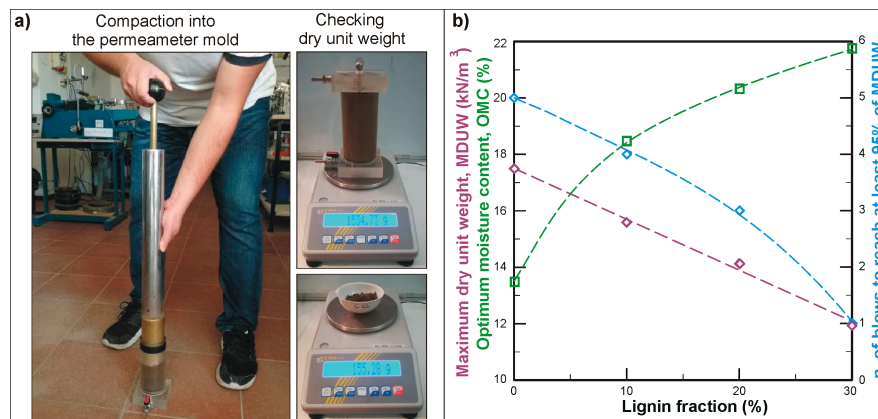

Figure 5. (**a**) Compaction procedure into the permeability cell (φ = 63 mm). (**b**) Verification of the number of blows needing to reach 95% of optimum compaction properties obtained by standard Proctor on the different S_A/lignin mixtures.

3. Results

3.1. Effect of Lignin on Consolidation and Hydraulic Conductivity of Soil SB

The saturated consolidation characteristic was investigated to give insight into the effect of 10% lignin content on compressibility and hydraulic conductivity of soil S_B. The FSR for soil S_B + 10% lignin was 0.75, indicating a negligible degree of swelling potential.

Samples were tested at standard optimum compaction properties: MDWU and optimum moisture content (OMC) for soil S_B and soil S_B + 10% lignin were 11.1 kN/m^3 ÷ 18.4% and 9.06 kN/m^3 OMC ÷ 18.8%, respectively.

Figure 6a shows settlement values (δ) with time for the different effective stress (σ') as resulted in soil S_B and S_B + 10% lignin. As expected, the vertical displacement of both materials increases as the consolidation pressure increases. The behavior of soil S_B + 10% lignin is particularly interesting: at low effective stress (σ' = 12.5 kPa), it showed a much higher settlement than that achieved by soil S_B alone. Moreover, as the effective stress increased, the gap between the two materials decreased. Steady-state settlement of S_B + 10% lignin at σ' = 100 kPa was fairly similar to that of soil S_B for σ' = 200 kPa. Soil S_B + 10% lignin reached 50% of the total settlement at 100 kPa. A possible explanation of high settlement of soil S_B + 10% lignin at low-stress value (up to 100 kPa) is that some stabilization occurs in the silty soil (electrostatic reaction, as well as ionic binding, took place). In other words, the soil tended to assume a "sandy behavior" and, as a result, even at low stress, there was a more marked reduction of its volume compared to the soil without lignin (S_B).

According to the theory of consolidation, the change in height (H) of samples corresponded to a reduction of the void index (e). The same apparatus was used to evaluate the hydraulic conductivity (*k*) with the increase of the effective stress (i.e., at different void index). The hydraulic conductivity of S_B soil showed the expected trend (Figure 6b), with low hydraulic conductivity for structure at higher effective stress (low void ratio). Although the hydraulic conductivity of soil S_B decreased one

order of magnitude passing from 12.5 kPa to 400 kPa, it did not meet the minimum permeability requirements for landfill bottom liner ($k < 10^{-7}$ cm/s). As the effective stress increased, the hydraulic conductivity of soil $S_B + 10\%$ lignin was also reduced, but the trend was much different than that of soil S_B. Up to $\sigma' = 50$ kPa, the mixture of soil and lignin showed low k values compared to soil S_B. For $\sigma' = 100$ kPa, the mixture showed similar hydraulic conductivity to soil S_B; after this effective stress, the hydraulic conductivity remained slightly higher than that of soil S_B. In other words, the addition of 10% in weight of lignin to soil S_B reduced the hydraulic conductivity only for low effective stress levels, making it not suitable to be used in landfill bottom liners.
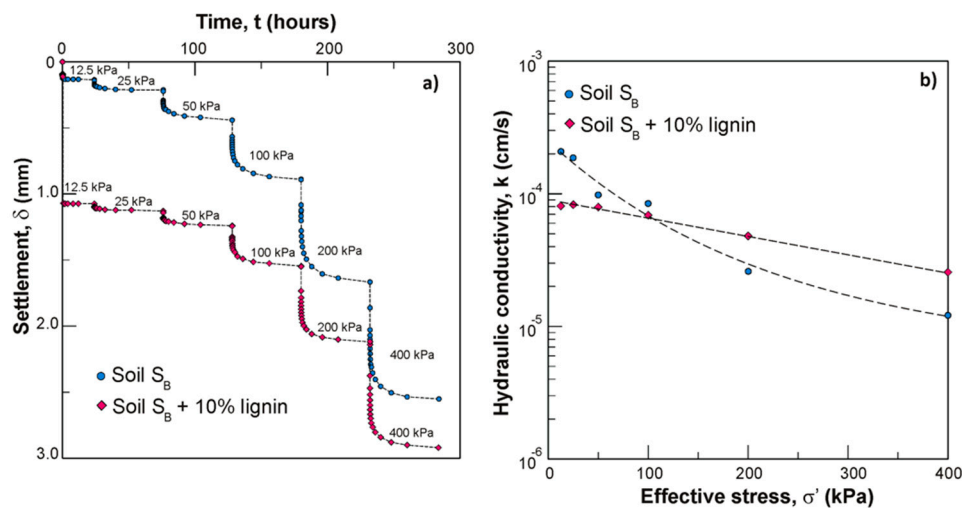

Figure 6. (**a**) Settlement values with time for soil S_B and $S_B + 10\%$ lignin. (**b**) Hydraulic conductivity of soil S_B and $S_B + 10\%$ lignin for different effective stress values.

3.2. Effect of Lignin on Hydraulic Conductivity of Soil SA

Soil S_A was tested for suitability as a component of the capping system (compacted sealing layer). Considering the top layer of the landfill is not subjected to excessive loads as is the case for the bottom layer, the compressibility of the mixture had a reduced influence in this case. The consolidation was not investigated here. We focused on the effect of lignin content on the hydraulic conductivity of the mixtures.

Four series of permeability tests were conducted on dynamically compacted and reconstituted specimens of S_A soil with different lignin content (0%, 10%, 20%, and 30% in weight). Initial dry unit weight and water content values for the different soil/lignin mixtures are reported in Figure 5. The hydraulic conductivity measurements were replicated several times in order to check the quality of data. Figure 7 shows the results for the different sandy silt/lignin mixtures. As the lignin content increased, a decrease in the hydraulic conductivity was observed, indicating an overall improvement of the performance of S_A soil. For a lignin content of about 28–30%, the soil met the minimum permeability requirements for compacted top-sealing layers ($k < 10^{-6}$ cm/s). For this mixture, the FSR was about 1.9: according to FSR classification [47], this value falls in the swelling class materials with moderate expansivity.

Some attempt was made to investigate the mechanism responsible for the reduction in soil permeability. Soil S_A and $S_A + 30\%$ lignin were analyzed by scanning electron microscope (SEM) to investigate micropores distribution. Figure 8a,b shows SEM images at a different scale. Widespread intergranular pores developed in soil S_A, which were mainly controlled by the compaction effect. The size of pores ranged from about 20 μm to about 80 μm; pores were well connected, producing a high soil hydraulic conductivity value (3×10^{-5} cm/s). On the contrary, S_A soil + 30% lignin showed a packed structure with small pores of 5–10 μm, reaching the minimum hydraulic conductivity value (4×10^{-7} cm/s). The adding of lignin affected the particle size distribution of soil: the fine fraction

(P200) increased to about 60%, affecting *k*-values. As shown in Figure 8, lignin fills the micropores formed by fine-grained particles indicating that the possible mechanism responsible for hydraulic conductivity reduction consists mainly of physical binding [62]. A similar mechanism has been recently proposed by reference [30], indicating that the stabilization mechanism differs from traditional soil stabilizers (i.e., lime).
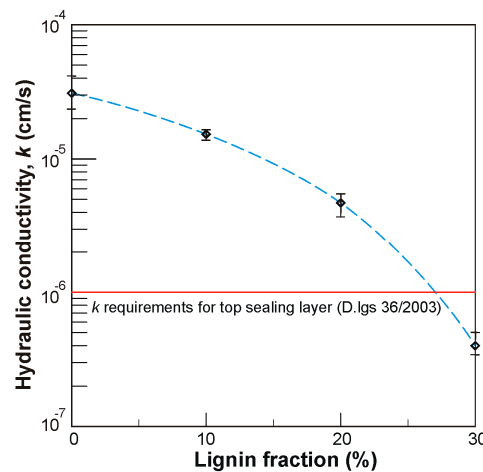

Figure 7. Hydraulic conductivity of soil S_A with different fraction of lignin (0%, 10%, 20%, and 30% in weight).
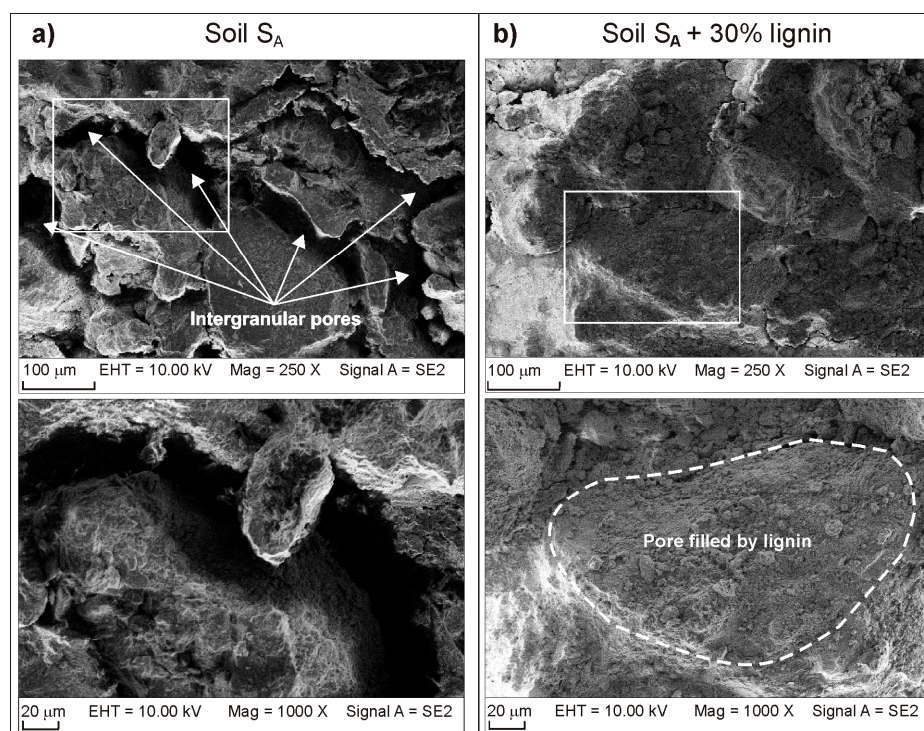

Figure 8. Scanning electron microscope (SEM) images. (**a**) S_A soil and (**b**) S_A soil + 30% lignin at two magnifications (250X and 1000X).

4. Discussions

The laboratory experiments provided some first indications about the use of a mixture of soil and lignin for the landfill barriers. The tests were mainly aimed at verifying the permeability of the soil/lignin mixture with some consideration on the compressibility.

The first type of experiment, conducted through oedometer tests, highlighted that by adding 10% in weight of lignin to S_B soil, the hydraulic conductivity of the mixture decreased. Nevertheless, the value required for landfill bottom liners was not reached ($k < 10^{-7}$ cm/s). In addition, the mixture showed compressibility higher than that of the soil, indicating that the mixture was not promising to ensure the stability of the landfill base.

The second type of experiment, conducted through a permeability cell on compacted not plastic S_A soil, indicated that mixtures with 28–30% in weight of lignin allowed the achievement of hydraulic conductivity values lower than $k < 10^{-6}$ cm/s (the threshold value for the top-sealing layer of the landfill by the Italian regulation). The use of the mixture soil/lignin for the top-sealing layer of landfills can represent an innovative solution with specific reference to the improvement of the hydraulic conductivity. However, the moderate expansivity and swelling behavior of the mixture suggested that further studies have to be carried out, checking whether the laboratory value can be reproduced in practice and how to ensure the long-term durability of the capping system. It is also a typical problem encountered with other additives, such as coal fly ash [63] or paper sludge [64]. It should be pointed out that the issue of swelling and shrinking of the capping system also occurs when traditional materials are used (e.g., clayey soils). As reported by reference [65], the addition of small quantities of bentonite has a greater impact on swelling index than on compressibility. These problems require designing and performing field experiments, which consider the best compaction techniques and local meteo-climate conditions. In this way, the maintaining of the water content of the barrier near to the optimum moisture content contributes to avoiding cracks produced by shrinkage and swelling [66,67].

Considering the hypothesis of increasing production of organosolv-like lignin in the future [20], new fields of application are to be expected. This by-product could be used as an additive for the realization of a landfill top-sealing layer. In this way, a reduction in the cost of containment construction system is presumed. The approach results in savings the raw materials, avoiding the costs for purchasing the clayey soils from quarries and recovering the in-situ soils, which should be disposed of if not used. According to reference [68], it is not possible to generalize the evaluation because "all the costs are tentative and an approximate indicator of cost which may change according to the location and may vary with time." Although advances in the optimizing of the organosolv process should be made [69], currently, the method could be considered convenient, especially for landfill sites located near the lignin production area (lignin availability with reduced transport costs). This preliminary consideration should be further explored in a more general and accurate context of the life cycle assessment (LCA), useful to assess the environmental impacts and resources used throughout a product's life cycle (i.e., from raw material acquisition, via production and use phases, to waste management [70]). Although the LCA is not the focus of the present work, the recycling of lignin is a key factor in also reaching environmental benefits considering its new value as amender at a large-scale application, reducing the overall impact on the environment (coast for lignin disposal, etc.).

5. Conclusions

Considering the hypothesis of the potential improvement of the permeability values of in-situ soils by adding the organosolv lignin, the objective of the study was to investigate the performance on two fine-grained soils widely outcropping in Central Italy, which do not meet the permeability requirements for landfill barriers. The following conclusions can be drawn from the study:

–   The hydraulic conductivity of soil S_B amended with 10% in weight of lignin did not reach the values required for landfill bottom liners ($k < 10^{-7}$ cm/s). In addition, the mixture showed compressibility higher than that of the soil alone, making it unsuitable to be used under the loads induced by the landfill.

–   For soil S_A, the increase in the ratio of lignin (28–30% in weight) may lead to an effective reduction in hydraulic conductivity, reaching values lower than $k < 10^{-6}$ cm/s as recommended for the top-sealing layer of landfills by the Italian regulation.

−   The improvement of k of soil S_A may be attributed mainly by physical binding produced by lignin, which filled the micropores formed by fine-grained particles.

The preliminary results of this study suggest that lignin has good suitability for improving in-situ soils for the top-sealing layer of landfills. Its use could be very convenient in terms of costs when compared with those relating to the use of raw materials. In the context of the circular economy, green policies and related incentives could increasingly encourage the use of by-products, the application of which cannot be achieved without in-depth laboratory studies and in-field verification. Further focused studies are needed to reproduce the hydraulic conductivities in the field similar to those found in the laboratory. In this framework, large-scale investigations could be useful to check the long-term performance of the mixtures, also monitoring in-field settlements and swelling.

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