



Hydrogeological Properties of Gypseous soils in South Africa

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Abstract

Gypseous soils occur in the western arid and semi-arid regions of South Africa and Namibia. These soils exhibit a complex nature and abnormal behaviour due to their gypsum content and as such they have become more prevalent in research. As these soils are finding more use in industry, an astute understanding of their hydrogeological properties and behaviour is required. Powdery gypseous soil samples collected from the Northern Cape (Geelvloer) and Western Cape (Rooiberg and R355) Provinces, as well as a prepared sample, are subject to XRD analysis, particle size distribution determination and falling-head permeability tests using both water and brine. The testing served as preliminary research to guide further studies into the topic. The prepared sample, with 19% fines, comprises 35% gypsum and 65% sand. Geelvloer samples, with 91.95% gypsum content, are comprised mostly of sand-sized particles with 45% fines. Rooiberg samples contain 75% fines with a slightly lower gypsum content of 83.25%, while R355 samples have 50% fines with 75.35% gypsum. It is generally understood that particle size distribution contributes to the hydraulic conductivity of soils, where a higher portion fines will result in a lower conductivity. In the case of gypseous soils, the solubility is of importance as well, as it may have long term effects. With the intent of evaluating the effect of the aforementioned factors on the hydraulic conductivity of gypseous soils in South Africa, the samples taken represent differences in particle size distribution and origin. Geelvloer had k -values in the order of 8.82×10^{-6} m/s, with the brine sample giving 9.43×10^{-6} m/s, while the k -values for Rooiberg and R355 were in the order of 3.90×10^{-6} m/s and 5.87×10^{-6} m/s, respectively. The brine resulted in 5.63×10^{-6} m/s for Rooiberg and 9.90×10^{-6} m/s for the R355 sample. The made sample, having less fines, had k values in the order of 2.15×10^{-5} m/s, and 4.19×10^{-5} m/s for the brine. The differences between the results are largely negligible and show that despite what is believed to influence the hydraulic conductivity, in the case of gypseous soils in South Africa, on a small scale, it remained unaffected.

Introduction

Gypsum-rich soils are defined as soils where there has been a significant accumulation and/or replacement of a pre-existing soil or mineral by gypsum due to processes within a zone of weathering (Aref, 2003). The term 'gypsum-rich soils' is a broad classification of soils that contain enough gypsum to alter its properties, considering the degree of alteration could vary depending on the gypsum content. The terms 'gypsiferous' and 'gypseous' are commonly encountered in literature. Although there is no official classification, it is understood that 'gypsiferous' denotes soils which are considered to be 'gypsum

bearing' with a gypsum content between 1 and 40% and 'gypseous' refers to soils where gypsum is the main constituent with a gypsum content greater than 40% (Boyadgiev and Verheye, 1996; Herrero and Porta, 2000; Pearson et al., 2015). For the purpose of this research, all gypsum-rich soils will be referred to as gypseous soils.

Geology, climatic conditions and parent material are the most important pedogenic factors governing the formation of gypseous soils in South Africa. These soils most commonly occur in semi-arid and arid regions, where the nature of the

soil varies greatly in terms of gypsum content, crystal habit and size, and degree of cementation (Chen, 1997). This is as a result of various genetic and diagenetic processes that lead to the formation of gypseous soils or gypcrete (analogous to the terms calcrete or silcrete, denoting induration to some degree), where dissolution and recrystallization results in textures similar to chemical evaporites with the exception that they can exist at different topographic elevations (Aref, 2003).

The solubility of gypsum as well as the effect of heat and water on the chemical structure, leads to soils which are ultimately challenging to understand with properties that are difficult to characterise. Minimal published data exists in South Africa with regards to the geotechnical and hydrogeological properties of these soils, despite their wide application. Gypseous soils are commonly and successfully used abroad for purposes beyond the manufacture of Plaster of Paris or cement, where gypseous soils have been found to make reliable road building materials

Interest in these soils is spurred on by the fact that the gypsum, a complex mineral present as a main component of the soil, will control the behaviour of the soil.

Occurrence

Gypsum in South Africa mainly occurs in clay as crystal aggregates or as discontinuous horizontal layers of powdery material, where its occurrence in surficial and shallow pedogenic settings is restricted to areas where evaporation exceeds precipitation (Eckardt and Spiro, 1999). Gypsum also occurs as secondary aeolian deposits (FAO, 1990). In Southern Africa, large gypsum deposits are found mainly along the west coast, being particularly prominent in Namibia, and stretching south into the Northern Cape Province of South Africa (Figure 1). This coincides with areas of the country which are considered semi-arid or arid.

The gypsum is distributed as a component of clay and sand in sediments of the Cenozoic Era between Alexander Bay and Port Nolloth. Further south in the Western Cape Province, near Yzerfontein and Saldanha, some deposits can be found in multiple salt pans (Brabers, 1976). Large deposits also exist throughout the Knersvlakte near Vanrhynsdorp, where it is present in the form of selenite crystals, powdery gypsum and alabaster hosted in residual clay, which originated from the Louisrus Formation, Little Namaqualand Suite, Gariep Supergroup and Vanrhynsdorp Group (Cole et al., 2014).

Gypsum deposits found inland are most commonly associated with the Whitehill Formation, an extensive Lower Permian unit, which forms part of the Ecca Group (Cole et al., 2014). It often outcrops in a thin band stretching west from the Eastern Cape Province, into the Western Cape where it extends northwards into the Northern Cape and eastwards across the province, where most of South Africa's largest deposits occur. A chemical reaction in the weathered zone between calcareous materials, believed to be calcrete or calcareous nodules in shale, particularly shales of the upper Dwyka Group, and sulphuric acid led to the formation of these gypsum deposits. The oxidation of pyrite, a prominent

mineral in the Whitehill Formation, serves as the source of sulphuric acid (Cole et al., 2014).

Properties of gypseous soils

Mineralogy

The characteristics and behaviour of gypseous soils is governed by the content and type of gypsum within the soil. Gypsum is a moderately soluble mineral represented by calcium sulphate dihydrate ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), with a hardness of two on the Mohs scale (Deer et al., 1992). Gypsum occurs in various shapes depending on the formation processes. Common morphological forms of gypsum found in nature include; alabaster (fine-grained and massive), satin spar (fibrous) and selenite (clusters of crystals) (Brabers, 1976). The different morphologies are relevant as they develop different soil fabrics that affect soil behaviour (Shahid and Abdelfattah, 2009).

The structure of gypsum crystals consists of adjacent parallel layers of calcium ions (Ca^{2+}) bonded to tetrahedral sulphate ions (SO_4^{2-}). The layers are separated by sheets of water molecules that are weakly bonded to neighbouring sheets (Klein and Dutrow, 2007).

Due to the bound water in the structure, gypsum crystals undergo dehydration at relatively low temperatures, resulting in the formation of several progressive phases of metastable calcium sulphate accompanied by a change in chemical structure and volume (Posnjak, 1938; McConnell et al., 1987; Horta, 1989; Carbone et al., 2008; Jacques et al., 2009; Khasanov et al., 2008). There are three recognisable phases in the $\text{CaSO}_4 - \text{H}_2\text{O}$ system; namely gypsum, hemihydrate (commonly known as bassanite used as Plaster of Paris, $\text{CaSO}_4 \cdot \frac{1}{2}\text{H}_2\text{O}$) and anhydrite (CaSO_4) (Seufert et al., 2009). During the process of dehydration it is unclear if there is a complete structural change or only a morphological and dimensional alteration (Carbon et al., 2008). The dehydration process implies that in areas where rainfall is minimal and temperatures are high, the surface deposits could potentially experience mineralogical changes throughout a day.

The reversibility of the entire process is equally important to understand. Both dehydration phases readily take up water to re-hydrate, eventually reforming gypsum (Jha and Sivapullaiah, 2017). Hemihydrate reacts with water relatively quickly, a phase transition that is particularly notable as it undergoes setting and hardening resulting in a solid mass (Sirokman, 2014). This effect could lead to the development of hardening between soil particles, reducing the ability of water to penetrate and flow through the soil, resulting in variability in the permeability of gypseous soil (Kamei et al., 2013). The hardening of gypsum is a property that led to widespread use in the manufacturing of cement. When rainfall occurs, the initial downpour will not infiltrate the soil profile, rather be taken up by the minerals in the soil, if a phase change occurred.

Solubility

Although moderately soluble, gypsum will dissolve which could lead to soil mass loss due to leaching (Asghari et al., 2014). The solubility varies depending on particle size, the presence of

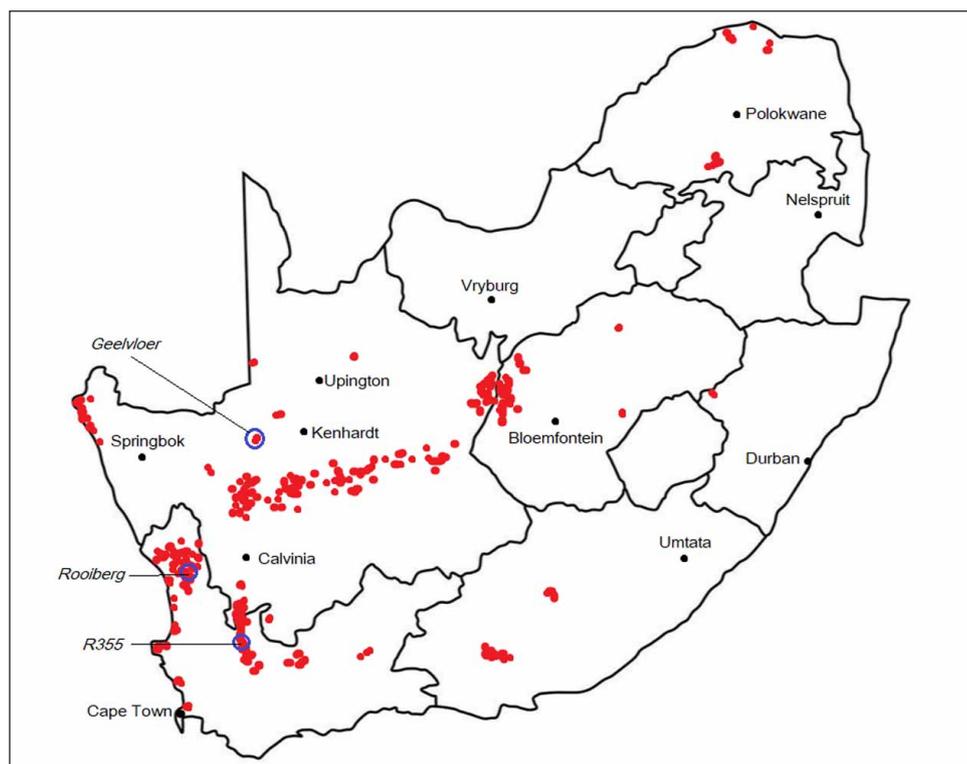


Figure 1. Gypsum Deposits in South Africa (adapted from Council for Geoscience, 2002).

other salts (sodium and magnesium chlorides, in particular, increase the solubility of gypsum), and the flow velocity of the solution moving through the soil (Kuttah and Sato, 2015). Gypsum dissolution leads to an increased void ratio in gypseous soils, resulting in cavities developing between particles ultimately forming channels for easier water flow (Al-Dabbagh et al. 1990, as cited in Kuttah and Sato, 2015). Continued leaching of gypsum will lead to larger cavities, which could result in sudden collapse (Cooper, 1989).

The presence of gypsum affects the hydraulic conductivity (k) of the soil (often referred to as the coefficient of permeability), with the extent largely dependent upon the quantity and size of the particles with respect to the soil particles (Kuttah and Sato, 2015). Keren et al. (1980) state that the hydraulic conductivity is reduced when the gypsum particles are very fine due to the closing of pore spaces between fine textured soils. This is not the case with coarser textured soils, as gypsum is seen to have minimal influence. Kuttah and Sato (2015) concluded that the soil permeability increases with increasing gypsum content if the gypsum particles are larger than the soil particles and a decrease in permeability occurs when the gypsum particles are smaller than the soil particles as they block flow paths.

Materials and methods

Sampling

Gypseous soil samples were collected from one location in the Northern Cape Province, a salt pan which is situated west of Kenhardt called Geelvloer, and from two locations in

the Western Cape Province, an abandoned mine north-west of Vanrhynsdorp, called Rooiberg, and a farm situated approximately 60 km south of Tankwa Karoo National Park along the R355, as seen in Figure 1.

All the samples contained powdery gypsum (Figure 2). The samples were dried at 40°C to prevent any changes in mineralogy upon re-hydration, which would lead to possible minor cementation between particles, ultimately affecting the results. An additional soil sample was prepared using a mixture of 35% powdery gypsum (sponsored by Pretoria Portland Cement Company Limited) and 65% clean, washed sand, as it is relatively inert.

Particle size distribution and XRD

The particle size distribution was done using SANS 3001-GR1, which details the wet preparation and analysis of soils.

The geochemistry of the gypseous soils was analysed using X-ray Diffraction (XRD). The samples were milled and prepared according to the standardised Panalytical backloading system, for analyses using a PANalytical X'Pert-Pro Powder Diffractometer in θ - θ configuration with an X'Celerator detector. The phases were identified and quantities estimated using X'Pert Highscore Plus software.

Falling-head tests

Several falling-head tests were conducted on the samples in order to establish the hydraulic conductivity of the soil. The

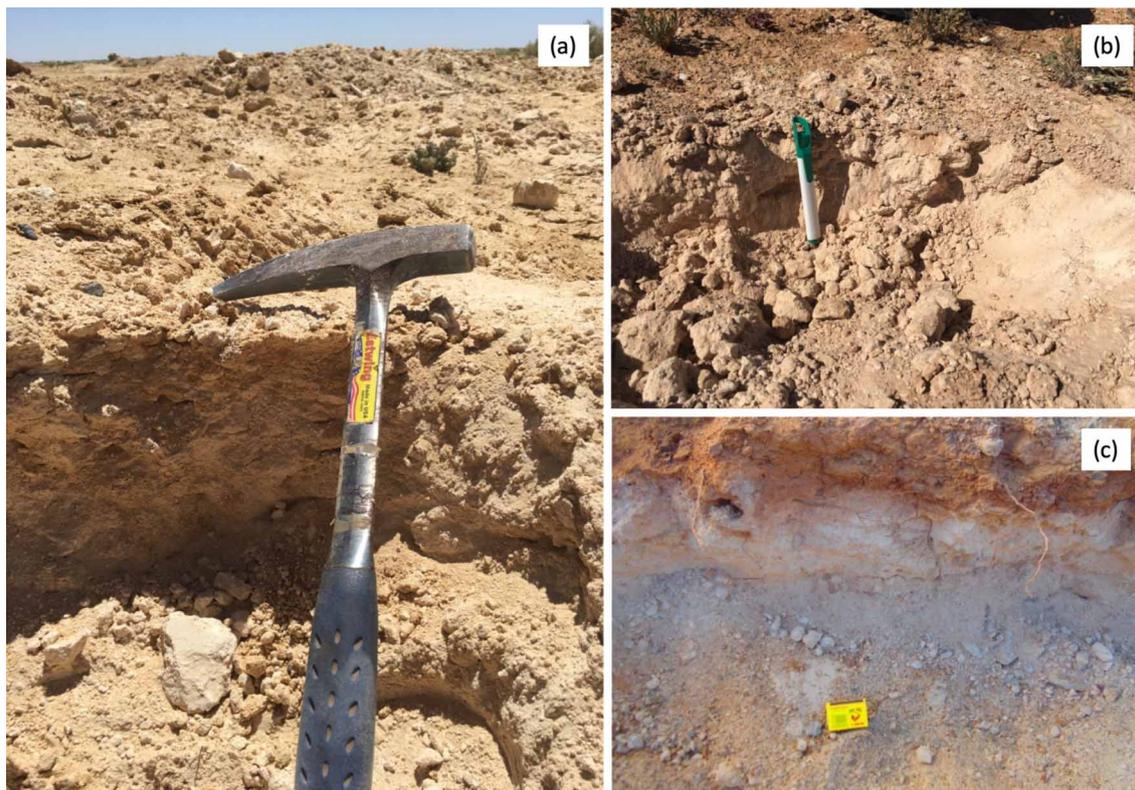


Figure 2. (a) Surface deposit of gypseous soil in Geelvloer. (b) Surface deposit of gypseous soil in Rooiberg. (c) Surface deposit of gypseous soil along the R355.

standard methodologies for falling-head tests were adapted for the purpose of this study (ASTM D5084, 2003).

Disturbed samples were packed in a perspex column, sandwiched between two coarse-grained quartz filters with a length of 40 mm each. The samples were 150 mm in length with a diameter of 60 mm (Figure 3). The initial stage entailed introducing an influx of water large enough to create a

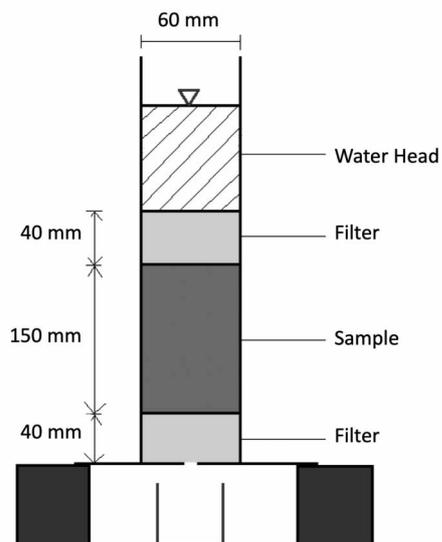


Figure 3. Schematic of experimental setup of falling-head permeability tests.

significant head, which was allowed to flow through the sample with the intent of creating uniform conditions, hydrodynamic consolidation and assumed saturation.

Once all the initial water had drained, the test was started by adding water (500 ml), increasing the head to that of the initial water influx, and allowing the water to drain through the material. The drop in head was measured every 15 minutes until all the gravitational water had flowed through the sample. The sample was then allowed to dry partially and the permeability test was run again, to assume natural field conditions. This was done for three cycles; producing three sets of permeability test results for each sample from the three locations and the prepared sample. The electrical conductivity (EC) of the water was measured after each cycle to assess the ionic character of the solution. An additional permeability test was done on each of the samples using a 3% brine solution instead of water to evaluate the effect of sodium chloride on the gypseous soils. All data from the falling-head tests were interpreted using Darcy's Law.

Results

The XRD analysis results and particle size distribution, following SANS 3001-GR1, of the samples are summarised and shown in Figure 4.

The natural samples all contain more than 75% gypsum, while the prepared sample contains 34.11%. Geelvloer and R355 samples have similar fines content with 45 and 50%, respectively.

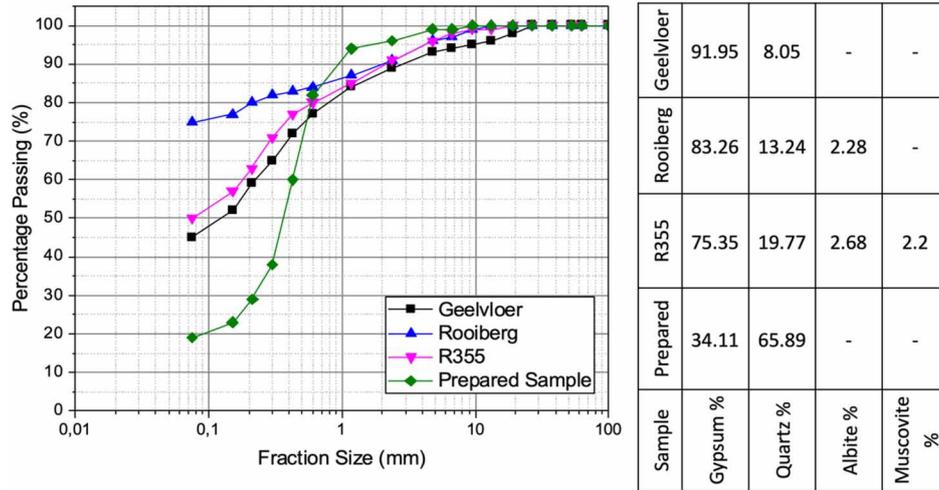


Figure 4. XRD analyses and grading curves for the gypseous soil samples.

Rooiberg, with 75% fines, and the prepared sample, with 19%, show more variation with the particle size distribution.

The change in head over time for all the samples are represented by graphs shown in Figure 5, which were interpreted and summarised in Table 1, along with the electrical

conductivity (EC) of the water that had drained through each sample. It should be noted that the graph in Figure 5d has a larger scale along the Δh -axis than the graphs in Figure 5a, b and c, as it is considered more suitable.

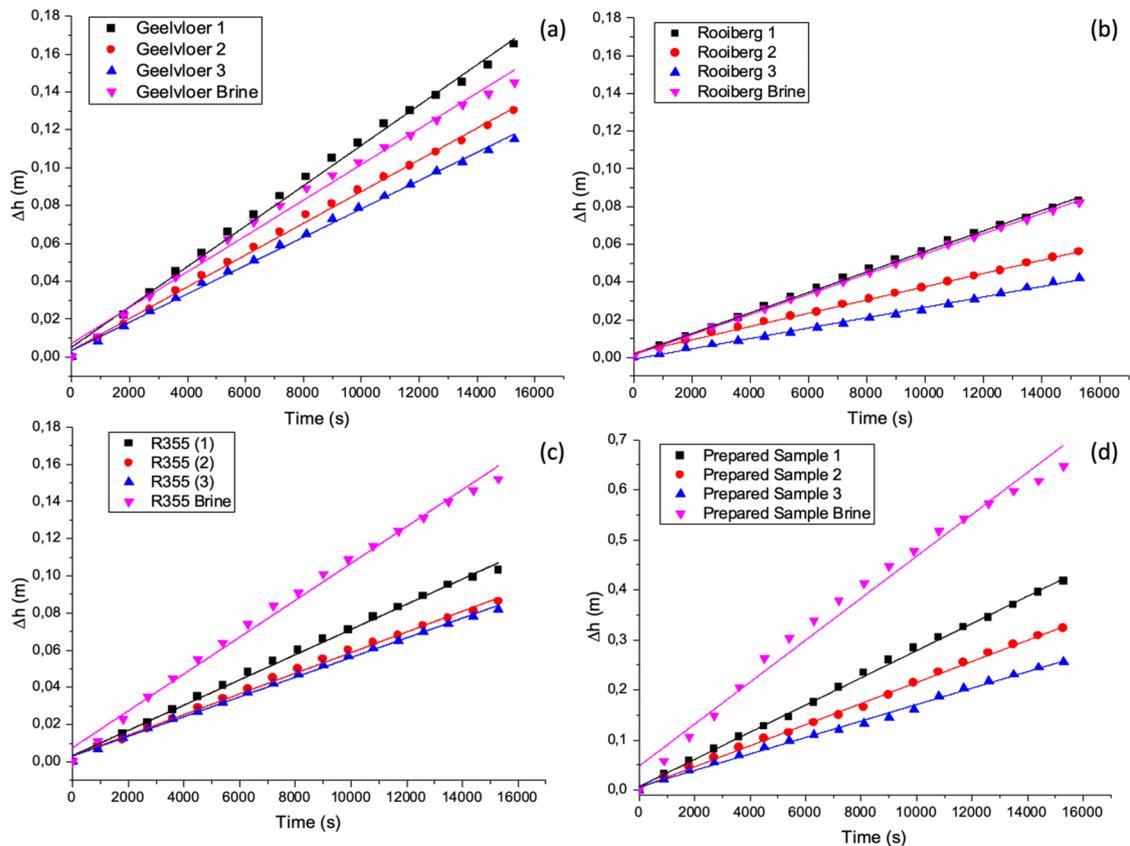


Figure 5. (a) Change in head per time interval for Geelvloer; (b) Change in head per time interval for Rooiberg; (c) Change in head per time for R355 and (d) Change in head per time for the prepared sample.

Discussion

Particle size distribution and XRD

The XRD analysis results and particle size distribution of the samples are shown in Figure 4, where evidently samples collected from Rooiberg are predominantly clay-sized particles, with 75% passing the 0.075 mm sieve, having 83.26% gypsum content. Geelvloer, with 91.95% gypsum, and R355 samples, with 75.35% gypsum, have 45% and 50% fine particles, respectively, and contain large portions of sand-sized particles, perhaps due to a degree of cementation between particles. The prepared sample has only 19% fines. The presence of albite and muscovite is deemed to be ineffective due to the minor quantities present in the samples.

Falling-head tests and electrical conductivity

The initial stage of the permeability tests for all the samples was marked by the first five minutes where the first water flowing through created flow channels that lead to faster drainage through the soil, a rapid drop in head and soil mass loss as some fine particles were expelled with the water, leaving it murky. Soil mass loss was greater for Rooiberg samples, as seen by a greater decrease in the length of the sample within the column. After this, clogging occurred as fine particles situated further up made their way through the sample, blocking the channels, reducing the void space and slowing flow conditions to reach steady-state conditions, where the remaining water flowing through was clear and consistent. After all the gravitational water from this stage had drained through the falling-head tests were started.

The first cycle of tested soil from Geelvloer (Figure 5(a)) shows a linear slope with a constant, saturated k value of 1.06×10^{-5} m/s. The second and third cycle also produced linear

slopes with a slightly lower hydraulic conductivity, showing constant, saturated k values of 8.38×10^{-6} m/s and 7.48×10^{-6} m/s, respectively. The sample is largely classified as sand, which may make these values seem slightly low, however with 45% fines, blocking of small void spaces, thus slowing the flow of water would have contributed to the values. Some of the gypsum went into solution, as is clear from the EC values (Table 1), but the particles still remained large enough to clog the sample. This is important as partial dissolution leads to smaller particles which would then be able to fit through inter-connected void spaces that were previously too small, and with time may increase the hydraulic conductivity. The EC of the water, that had drained through the soil after cycle one, was 2.91 mS/cm. The EC values remain relatively uniform throughout all three cycles, with cycle two (2.73 mS/cm) and cycle three (2.52 mS/cm) being only slightly less than cycle 1. This could be expected as the solubility of gypsum is dependent on the size of the particles and the concentration of the fluid; hence gypsum will continue to go into solution.

Samples from Rooiberg (Figure 5(b)) and R355 (Figure 5(c)), as well as the prepared sample (Figure 5(d)), show linear results similar to those of Geelvloer samples with the graphs for cycle one of each location having slightly higher hydraulic conductivities than cycles two and three. R355 samples show a k value of 6.78×10^{-6} m/s for cycle one, 5.55×10^{-6} m/s for cycle two and 5.29×10^{-6} m/s for cycle three. These results, from samples with 50% fines, support the results of samples from Geelvloer, where similar particle size distributions resulted in similar hydraulic conductivities. This is expected, and the low values are also attributed to the clogging of any open spaces between the larger particles as the fines move with the water through the sample. R355 samples had an EC of 2.62 mS/cm for cycle one, 2.31 mS/cm for cycle two and 2.24 mS/cm for cycle three. This reiterates the continual dissolution of gypsum as water flows through the samples.

Rooiberg and the prepared samples represent greater differences in particle size distribution, and in the case of the prepared samples, appreciably lower gypsum content as well. Cycle one of Rooiberg has a constant, saturated k value of 5.43×10^{-6} m/s, where cycles two and three show k values of 3.52×10^{-6} m/s and 2.75×10^{-6} m/s. Considering that 75% of the Rooiberg samples consist of fine particles, the permeability values may seem slightly higher than expected, and are similar to those of Geelvloer and R355. The greater surface area, from the higher fines content, allows for more particles to start going into solution (with some possibly dissolving fully) resulting in even smaller particles able to flow through the sample with greater ease. This increases the amount of particles being mobilised, leading to slightly more inter-connected void spaces, facilitating the passage of water flow. After cycle 1, Rooiberg had an EC of 2.79 mS/cm, and after cycles two and three an EC of 4.09 mS/cm and 3.19 mS/cm, respectively, was measured. The slightly higher EC values support the reasoning of additional dissolution due to increased surface area.

The prepared sample shows a k value of 2.72×10^{-5} m/s for cycle one, 2.10×10^{-5} m/s for cycle two and 1.64×10^{-5} m/s for cycle three. Again, despite the differences in gypsum content and

Table 1. Data interpreted applying Darcy's Law based on falling-head tests.

Sample	Test run	Length (mm)	k (m/s)	EC (mS/cm)
Geelvloer	1	150	1.06×10^{-5}	2.91
	2	140	8.38×10^{-6}	2.73
	3	140	7.48×10^{-6}	2.52
Rooiberg	Brine	150	9.43×10^{-6}	-
	1	150	5.43×10^{-6}	2.79
	2	130	3.52×10^{-6}	4.09
	3	130	2.75×10^{-6}	3.19
R355	Brine	150	5.63×10^{-6}	-
	1	150	6.78×10^{-6}	2.62
	2	140	5.55×10^{-6}	2.31
	3	135	5.29×10^{-6}	2.24
Prepared sample	Brine	150	9.90×10^{-6}	-
	1	150	2.72×10^{-5}	0.83
	2	130	2.10×10^{-5}	0.75
	3	120	1.64×10^{-5}	0.71
Prepared sample	Brine	150	4.19×10^{-5}	-

particle size distribution, with only 19% fines, the hydraulic conductivity is similar to those of Geelvloer, R355 and Rooiberg. This is also attributed to clogging being the overriding mechanism. Even though there is less gypsum in the sample, thus less dissolution, the fine particles which are present are able to move between the larger particles, eventually getting trapped and obstructing flow. This highlights that even minor gypsum contents, with minimal fines, would control the hydraulic conductivity of similar soils. The EC values are expectedly lower due to lower gypsum content in the sample and also remain consistent throughout the testing.

The accompanying test run on all samples using brine instead of water had the intent of evaluating the effect of NaCl on the gypsum particles. Although it is understood that brine will increase the solubility of gypsum, increasing the effects of leaching, there is no appreciable difference in the hydraulic conductivity between the tests where water was used in comparison to the brine. Geelvloer, Rooiberg, R355 and the prepared sample had constant, saturated k values of 9.43×10^{-6} , 5.63×10^{-6} , 9.90×10^{-6} and 4.19×10^{-5} , respectively. The similarities in results are also attributed to the aforementioned mechanisms, where dissolution (partial or full) and clogging are active simultaneously, the extent of which depends on the particle size distribution and presence of other salts. As more particles are able to be mobilised, due to partial or full dissolution, more particles are able to block flow; the two mechanisms resulting in opposing effects.

Limitations

Despite the similarities in hydraulic conductivity between the samples and cycles, extensive, long term tests may show the continued leaching of gypsum leading to larger, inter-connected void spaces and an increased hydraulic conductivity where clogging may still occur, but is largely ineffective in comparison. The areas where these soils are most prominent would also need to be taken into consideration in this regard, as low rainfall would help hinder the leaching process.

Although interesting, these results represent a small scale experiment performed on disturbed samples. The remoulding of material will have an effect on the hydraulic conductivity in comparison to undisturbed samples, which are representative of field conditions. It is also known that the pH and temperature of the permeant will influence the hydraulic conductivity, as too will a greater difference in particle size distribution, where less fines or more cementing could alter the results. The limited data set hinders efforts to fully understand all the conditions affecting the hydraulic conductivity of these soils, highlighting the need for additional research.

Conclusion

Gypseous soils are highly complex, which leads to misconceptions and oversights when dealing with them. This is compounded by limited research done on these soils with regards to the geotechnical and hydrogeological fields, especially in South Africa. The complexity of these soils is

predominantly as a result of the intricate behaviour of gypsum. Gypsum deposits are distributed across large areas of South Africa as a result of multiple formation processes, leading to different morphological forms throughout the country. It is not uncommon to find different morphological forms in one location either, as gypsum readily changes in nature. It is understood that the major influences on the hydraulic conductivity of gypseous soils are the solubility and origin of the gypsum, the particle size distribution and the presence of other salts. Hydraulic conductivity tests, using both water and brine, were run on samples from three locations along with a made sample. For all four test sets, there are minor differences in the hydraulic conductivities; however these are largely negligible despite differences in the abovementioned factors. The similar results are due to two processes occurring simultaneously, namely partial dissolution and clogging, where mobilised particles are able to block flow further down the sample.

The effect of solubility was most evident in the electrical conductivity of the water which had drained through the samples. Consistently high EC values for the natural samples suggest gypsum continually went into solution throughout testing. This would be the biggest precaution with these soils, as it could eventually lead to large scale leaching and the formation of cavities.

Gypseous soils have many advantageous qualities when understood and dealt with correctly and are becoming more appealing as development is expanding. Research into the geotechnical properties for the use in roads along the western part of South Africa is urged as it would be convenient due to locality, minimising haulage costs, and excess, as lower grade gypseous soils remain unused by mines and manufacturers. In depth research into more hydrogeological properties of gypseous soils from more locations around the country is also highly encouraged.

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