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Journal of Civil Engineering and Construction Technology

Review

Towards enhancing sustainable reuse of pre-treated drill cuttings for construction purposes by near-infrared analysis: A review

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Sustainable reuse of pre-treated drill cuttings (a hazardous waste) as part substitute for fine aggregate in concrete for construction purposes is becoming increasingly attractive; however, issues remain. With recent studies focusing on the use of near-infrared spectroscopic technique for non-invasive determination of chloride concentration in concrete structures, this review examines the applicability of this new technique in the rapid determination of other equally important physicochemical characteristics of concrete produced with this hazardous waste. The nature, source, composition, ecological effects of, and management options for drill cuttings are reviewed. Furthermore, the principles of near-infrared diffuse reflectance spectroscopy are highlighted and lessons from its practical applications in soil science and petrochemical, environmental, and civil engineering are discussed. A framework for a rapid near-infrared analysis of concrete produced with pre-treated drill cuttings for enhanced sustainability as a construction material is also proposed.

Key words: Characterization, concrete, drill cuttings, diffuse reflectance spectroscopy.

INTRODUCTION

The predictable generation of drill cuttings during crude oil drilling activities has in recent times become one of the major global environmental concerns because of the hazardous nature of the drilling waste. Until recently, untreated drill cuttings were dumped into the seabed and on land during off-shore and land-based crude oil drilling operations respectively. From innumerable sources, records show staggering negative impacts of sea and land disposal of untreated drill cuttings on benthic and edaphic fauna and flora. Currently, various treatment technologies are available for drill cuttings; a majority of

which is driven largely by the intention to either recover the solid components for possible land application for agricultural purposes (Kogbara et al., 2017) or reuse them for civil engineering works (International Association of Oil and Gas Producers (IOGP), 2016).

One of the tenets of sustainable reuse of hazardous wastes like drill cuttings in civil engineering works like concrete production is to ensure that the resulting concrete complies with environmental guidelines. These guidelines are intended to protect ecological health well into the future. Available records show that drill cuttings

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are heavily laden with heavy metals and hydrocarbons (Ayotamuno et al., 2007; Okparanma et al., 2007). Even when drill cuttings have been thermally pre-treated, some of these contaminants remain (IOGP, 2016). Reports also show that pre-treated drill cuttings could be reused as part substitute for cement or fine aggregate in concrete (IOGP, 2016). In keeping with the tenets of sustainable reuse of hazardous wastes like drill cuttings, the resulting concrete is first analyzed for residual contaminants before it is put to the intended use.

However, issues remain particularly with the conventional concrete characterization techniques, which include crushing, acid digestion, solvent extraction, and instrumental analysis. These issues include costly delays due to lengthy sample preparation protocols, occupational health concerns associated with the use of noxious extraction solvents, and high analytical costs. These conundrums may have prompted the quest for possible alternative analytical approaches for concrete. There are indications to suggest that near-infrared (NIR) spectroscopy has the potential to be used for the analysis of certain chemical properties of concrete such as chloride concentration (Abbas, 2015). NIR spectroscopy is well known for its short analytical time, cost-effectiveness, non-invasiveness, portability, and ecological friendliness (Okparanma and Mouazen, 2013a). Besides, NIR spectroscopy has been extensively used in proximal soil sensing (Mouazen et al., 2010) and, on occasion, for the determination of chloride concentration in concrete (Abbas, 2015). However, NIR spectroscopy has never been used to determine the fundamental physical properties and other mechanical properties of concrete (such as compressive strength, permeability, bulk density, etc.). Similarly, NIR spectroscopy has not been used to assess the physicochemical characteristics of pre-treated drill cuttings such as bulk density, moisture content, as well as hydrocarbon and heavy metal contents etc.

The objectives of the present report were: (1) to review the source, composition, ecological effects of, and management options for drill cuttings, as well as the practical applications of NIR spectroscopy in soil science and petrochemical, environmental, and civil engineering; and (2) to highlight the possibility of using NIR spectroscopy for rapid characterization of pre-treated drill cuttings and concrete produced with pre-treated drill cuttings as part substitute for fine aggregate to enhance the sustainable reuse of pre-treated drill cuttings as a construction material.

SOURCE AND COMPOSITION OF OILFIELD DRILL CUTTINGS

During oil and gas exploration and production, generation of hazardous wastes from crude-oil drilling operations is inevitable. Managing such wastes has become one of the major environmental concerns for petroleum industries. This is because of the volume and chemistry of the resulting waste stream. Drill cuttings constitute the solid phase of the waste stream and consist of ground rocks, sands, and clays released from the well drilled through geological formations; whereas produced water, crude-oil, spent drilling muds, and additives make up the liquid phase (Lyons, 1996; Department of Petroleum Resources (DPR), 2002; Norwegian Oil Industry Association (OLF), 2003; Alberta Environment, 2005). Discussion on the liquid phase is outside the scope of the current review; however, interested readers are referred to other literatures such as Lyon (1996) for more information.

Drill cuttings are known to contain both inorganic and organic pollutants, including heavy metals and petroleum hydrocarbons, respectively (Okparanma and Ayotamuno, 2008; Okparanma et al., 2010). This is because of the influence of insitu chemical formulation of the proprietary drilling mud and mud additives used in drilling on the chemistry of the resulting drill cuttings (Wills, 2000). There are three most commonly used drilling muds, namely; oil-based mud (OBM), water-based mud (WBM), and synthetic-based mud (SBM). Figure 1 shows the approximate compositions, in weight percent, of the common drilling muds (IOGP, 2016). As shown in Figure 1a, WBMs mostly consist of brine/water mixed with bentonite (clay)/polymer and barite; and are said to be generally less harmful to the environment than other mud systems. OBMs and SBMs most times are collectively known as non-aqueous drilling fluids (NADFs) and largely composed of non-aqueous or synthetic base fluid mixed with brine and barite (Figure 1b).

Synthetic base fluids in SBMs are synthesized to behave like petroleum-derived oils during drilling. They are said to biodegrade readily and are generally less harmful to the environment than the non-aqueous base fluids in OBMs (Lima, 1996). The non-aqueous base fluids are polycyclic aromatic hydrocarbon (PAH)-carrying petroleum fractions such as crude oil, diesel invert, and conventional mineral oils (Lyons, 1996). They are responsible for the toxicity of OBMs, which explains why OBMs are more harmful to the environment than other mud systems (Lima, 1996). As a result, the use of OBMs has been outlawed in some countries including Nigeria (DPR, 2002), Norway (OLF, 2003), Indonesia (Permata and McBride, 2010) etc. However, because of their peculiar drilling properties, including superior temperature stability, lubricity, and hole stabilization qualities (Caenn and Chillingar, 1996), OBMs are often preferred to the other mud systems especially during deep drilling. As it turns out, OBMs are still being used (albeit in limited quantity) in some oil-producing countries at the detriment of human and environmental health (Okparanma et al., 2010). Apart from non-aqueous base fluids, NADFs also contain thinners, emulsifiers, and weighting agents such as barite and other additives (Figure 1b), which account
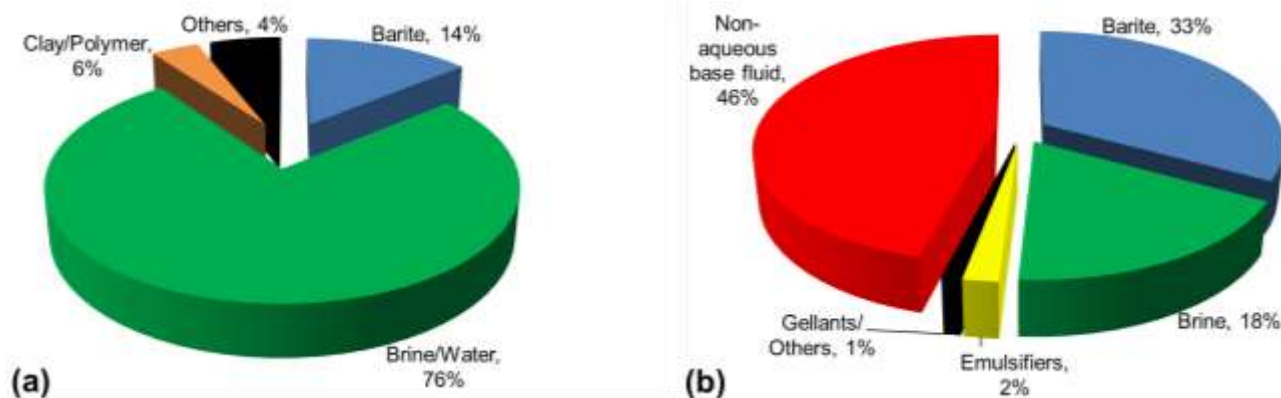
Figure 1. Approximate percentage weight composition of typical: (a) Water-based mud; and (b) Non-aqueous drilling fluids. Source: IOGP (2016).

for the heavy metal content of the drilling muds. Table 1 shows the reported physicochemical characteristics of untreated and thermally treated oil-based drill cuttings and the common mud systems. As can be seen in Table 1, the drill cuttings still contain traces of heavy metals and hydrocarbons even after thermal treatment. This underpins the need for proper physicochemical characterization of concretes produced with thermally treated (also known as pre-treated) drill cuttings before putting them to the intended use.

ECOLOGICAL EFFECTS OF DRILL CUTTINGS

The ecological effects of drill cuttings when disposed of on land or sea without prior treatment have been widely documented in the open access literature (Ellis et al., 2012; Zamora-Ledezma and García, 2013; Larsson et al., 2013; Durgut et al., 2015; Nilssen et al., 2015). In a recent review, Ellis et al. (2012) revealed a loss of benthic biodiversity and suspension-feeding communities due to discharge of drilling waste from oil and gas platforms. Zamora-Ledezma and García (2013) reported that drill cuttings are extremely toxic for some species of tropical grasses including *Urochloa decumbens* and *Urochloa maxima*. There have also been reported lower growth rates of cold-water coral *Lophelia pertusa* when exposed to water-based drill cuttings for 12 weeks (Larsson et al., 2013). Recently, while assessing the potential impact of water-based drill cuttings on two deep-water calcareous red algae, namely; *Mesophyllum engelhartii* and *Lithothamnion* sp., Nilssen et al. (2015) observed an occasional risk for severe impact on the calcareous algae. Therefore, effective management of drill cuttings becomes a consequent necessity, not least because some drill-cuttings contaminants such as PAHs are non-threshold carcinogens known to possess some genotoxic potential (Contaminated Land: Applications in Real Environments (CL:AIRE), 2010).

DRILL-CUTTINGS MANAGEMENT

Figure 2 shows a schematic of possible management options for drill cuttings, modified after International Association of Oil and Gas Producers (IOGP). As pointed out, one of the two key objectives that need to be addressed in the management of drilling waste is to comply with environmental regulations (IOGP, 2016), which are meant to protect ecological health.

Solids control, cuttings re-injection, and sea and land disposal

As can be seen in Figure 2, management of the waste stream from drilling operation starts from the shale shaker, which separates the solid (drill cuttings) from the liquid phase. The liquid part is recycled after pretreatment by de-sanding, desilting, or centrifuging. The drill cuttings may be re-injected into a re-injection well in suitable formations after slurrification, disposed of on land or sea. As stated, cuttings re-injection, sea and land disposal of drill cuttings without prior treatment have negative ecological impact associated with them. This explains why it is often recommended that drill cuttings are subjected to further secondary treatment before final fate. The current review focused on the reuse of pre-treated drill cuttings as construction material, particularly concrete. However, this could not be discussed in isolation of other treatment options for drill cuttings, especially secondary treatment technologies.

Secondary treatment technologies for drill cuttings

Secondary treatment technologies for drill cuttings may be broadly categorized into two, namely; the non-biological and biological treatment technologies (Figure 2).

Table 1. Physicochemical properties of untreated and treated oil-based drill cuttings and the two common mud systems.

Property		Oil-based drill cuttings		NADF[d, e]	WBM[d, e, f]
		Untreated[a, b, c]	Treated[b]		
pH		7.1	NR	NR	11.7
Sodium adsorption ratio (meq 100 g⁻¹)		5.3	NR	NR	NR
Exchangeable bases (meq 100 g⁻¹)		6.2	NR	NR	NR
Cation exchange capacity (meq 100 g⁻¹)		6.8	NR	NR	NR
Base saturation (%)		99	NR	NR	NR
Exchangeable aluminium (%)		1.2	NR	NR	NR
Electrical conductivity (mS cm⁻¹)		0.8	NR	NR	NR
Salinity (g l⁻¹)		0.9	NR	22.5 (%)	NR
Oil and grease (% weight)		13.7	NR	NR	NR
Moisture content (w/w %)		20.5	0.3	NR	NR
Dry matter (g/kg)		561 – 798	990 – 997	NR	NR
Volatile dry matter (g/kg)		51.8 – 90.2	40.5 – 91.9	NR	NR
Sand (%)		NR	5.4 – 29.1	NA	NA
Silt (%)		NR	19.4 – 75.4	NA	NA
Clay (%)		NR	19.2 – 64.7	NA	NA
	Aluminium (Al)	40,400	NR	52,000	10,800
	Arsenic (As)	2.9 – 8.5	4.0 – 8.4	NR	NR
	Barium (Ba)	270 – 8100	160 – 6900	487,000	720 – 449,000
	Iron (Fe)	21,900	NR	76,300	0.002 – 27,000
	Lead (Pb)	12 – 27	21 – 30	NR	NR
HM (mg/kg):	Cadmium (Cd)	0.06 – 0.36	0.08 – 0.22	0.39 – 12	0.16 – 54.4
	Copper (Cu)	15 – 52	25 – 180	NR	NR
	Chromium (Cr)	19 – 26	22 – 28	190 – 1,350	0.1 – 5,960
	Mercury (Hg)	0.017 – 0.073	0.025 – 0.087	NR	0.017 – 10.4
	Nickel (Ni)	16 – 25	17 – 31	NR	NR
	Zinc (Zn)	44 – 160	55 – 200	160 – 2,100	0.06 – 12,270
	Naphthalene	<0.0005 – 0.6	<0.0005 – 0.25	NR	NR
	Acenaphthylene	<0.0005 – 0.76	<0.0005 – 0.011	NR	NR
	Acenaphthene	<0.0005 – 3.8	<0.0005 – 0.18	NR	NR
	Fluorene	<0.0005 – 0.17	<0.0005 – 0.024	NR	NR
	Phenanthrene	<0.0005 – 0.26	<0.0005 – 0.17	NR	NR
	Anthracene	<0.0005 – 0.023	<0.0005 – 0.023	NR	NR
	Fluoranthene	<0.0005 – 0.056	<0.0005 – 0.045	NR	NR
PAH (mg/kg):	Pyrene	<0.0005 – 0.093	<0.0005 – 0.091	NR	NR
	Benzo[a]anthracene	0.003 – 0.042	0.006 – 0.035	NR	NR
	Chrysene	<0.0005 – 0.041	<0.0005 – 0.057	NR	NR
	Benzo[k]fluoranthene	<0.005 – 0.025	0.004 – 0.045	NR	NR
	Benzo[a]pyrene	<0.005 – 0.013	0.002 – 0.029	NR	NR
	Indeno[1,2,3-cd]pyrene	0.003 – 0.07	0.004 – 0.07	NR	NR
	Dibenzo[a,h]anthracene	<0.005 – 0.01	0.003 – 0.02	NR	NR
	Benzo[ghi]perylene	<0.0005 – 0.01	0.004 – 0.1	NR	NR

HM, Heavy metals; NADF, Non-aqueous drilling fluid; PAH, Polycyclic aromatic hydrocarbons; NA, Not applicable; NR, Not reported; WBM, Water-based mud. [a] Data from Zamora-Ledezma and García (2013); [b] Data from Vik et al. (2014); [c] Data from Deeley (1990); [d] Data from Health and Safety Executive (2000); [e] Data from Neff et al. (2000); [f] Data from Safi et al. (2016).

Non-biological treatment technologies

Drill cuttings are treated by non-biological methods such as drying (Schlumberger, 2016), solidification and stabilization (Akinlade et al., 1996; Shaffer et al., 1998; Fleming, 2000; Leonard and Stegemann, 2010; Opete et al.,
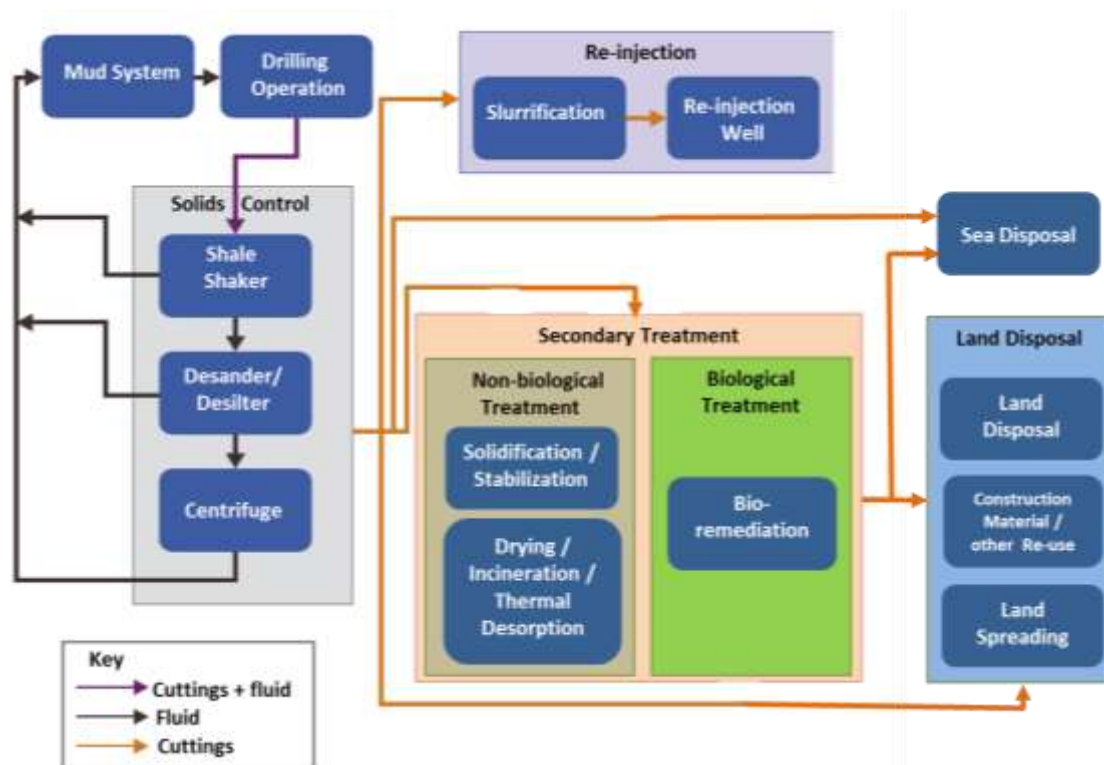
Figure 2. Schematic of possible management options for drill cuttings. (Source: IOGP (2016); reproduced after the permission of IOGP but with slight modification).

2010; Ghasemi et al., 2017), incineration (Dugat, 1988), and thermal desorption (Zupan and Kapila, 2000; Onwukwe, 2014). In recent years, more hot technologies for treatment of drill cuttings have emerged, including microwave-assisted heating process (Robinson et al., 2009).

Drying: This involves the physical removal of the liquid phase from the solid phase of the wet cuttings. This process is often aimed at recovering valuable drilling fluids for possible recycling and collecting dried drill cuttings for further treatment before final disposal. Schlumberger Limited (Schlumberger, 2016) reported a drill cuttings dryer, namely; VERTI-G® with a high-speed vertical centrifuge and maintained by programmable logic control. Drill cuttings are conveyed continuously from the shakers to the dryer's charge hopper through a range of site-engineered conveyance systems. Through independently adjustable flights, the drill cuttings are continuously directed to the screen surface. Under high G-forces, the flights create a rolling action that promotes liquid/solids separation instantly as drill cuttings make contact with the fine-mesh, high-capacity centrifuge screen. The result is dry solids discharge and clean fluid that can be recycled. However, the percentage of oil on cuttings or base fluid retained on cuttings after drying the drill cuttings was not reported in the literature. The

literature notes that retention of fluid on cuttings represents a financial loss (IOGP, 2016) and may be indicative of the drying efficiency of the dryer.

Solidification and Stabilization (S&S): S&S involves a chemical interaction between the drill cuttings and the solidifying and stabilizing agents as well as the mechanical binding of the drill cuttings to form a solid block of high structural strength (Drilling Waste Management Information Systems (DWMIS), 2000). The S&S approach is to ensure that base fluid retained on cuttings, anions, and heavy metals in the pre-treated drill cuttings are prevented from leaching to enhance land disposal or reuse as construction material (Figure 2). Akinlade et al. (1996) used Portland cement and Geosta-A® (as the fixing agents) on drill cuttings taken from depths of 1,828.8 and 3,368.3 m of the sandy Nun River Well-16 in the Niger Delta region of Nigeria. The Geosta-A® fixed the heavy metals through ion-exchange process and enhanced the binding of the cement/cuttings matrix through dehydration for a quick setting of the material. Using cement to cuttings ratio of 1:12 and 0.1 to 0.3% Geosta-A®, the slurry was moulded into blocks and allowed to cure for 28 days. They reported that a post-fixation leachability test indicated that except chloride ion, all leachable ions like arsenic, barium, cadmium, chromium, and lead were totally encapsulated. Moreover,
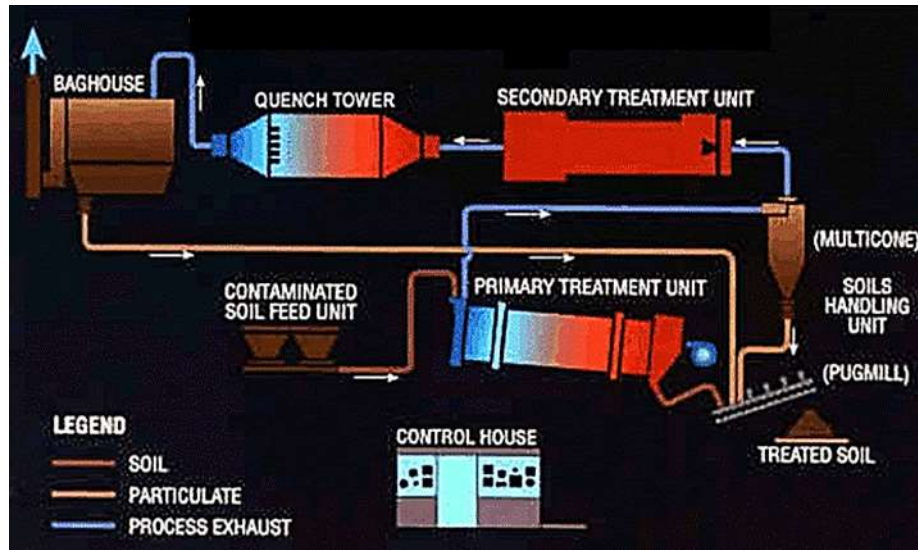
Figure 3. The thermal desorption process at Fort Edward, NY, USA (Source: http://employees.oneonta.edu/vislovt/mgp/excavation/Waste disposal.htm).

a compressive strength of 1,377.14 kN/m^2 was reported. However, in a similar exercise conducted by ChevronTexaco, Fleming (2000) reported that eight different commercial additives tested failed leachability tests due to sudden breakdown of the matrix or failure of the fixing agents to keep the drill-cuttings constituents from leaching to the environment over a long period of time. Studies carried out in Southern Louisiana University indicated that drill cuttings stabilized in a silica matrix had a pH greater than 11 and did not support plant growth when land applied (Shaffer et al., 1998). Again, Leonard and Stegemann (2010) used Portland cement mixed with high carbon-power plant fly ash (as a sorbent for organic contaminants) to stabilize drill cuttings. They reported that the drill cuttings and high carbon-power plant fly ash addition both reduced the unconfined compressive strength of the S&S products. Moreover, in the S&S treatment of synthetic drill cuttings with ordinary Portland cement (OPC), Opete et al. (2010) reported that concentrations of arsenic, mercury, and silver in the resulting concrete were reduced below local regulatory values. However, they observed that cadmium, chromium, and zinc contents were higher than the local permissible limits. Similarly, Ghasemi et al. (2017) used OPC (Type I 42, 5) and modified clay (Claytone$^®$ 40) mixes for the S&S treatment of oil-based drill cuttings. They observed that at a given clay content, an increase in cement content lowers the leaching rate; but, at a given amount of cement, increasing the clay content does not necessarily reduce the rate of leaching.

Thermal treatment: Thermal treatment involves high temperature heating of drill cuttings in order to reclaim or completely destroy them, which results in immobilization of inorganic metals and salts and overall reduction in drill cuttings volume (Bansal and Sugiarto, 1999). There are two main categories of thermal treatment, namely; incineration and thermal desorption.

Incineration: Incineration is the combustion of wastes at high temperatures (1,200 to 1,500°C), which is aimed at converting them into ash that is less bulky, non-hazardous, or less hazardous than they were before incineration (Morillon et al., 2002). An incineration method and apparatus for treatment of oilfield wastes was patented by Dugat (1988). The pre-treated wastes are dried and incinerated to a char solids condition. The char solids and the heating gases are cooled by heat exchange to provide hot air for combustion in the dryer-incinerator, thereby saving heating fuel cost. The gases are then scrubbed to remove contaminants and the condensed water-hydrocarbon phases are separated for reuse or for further treatment for recovery (Dugat, 1988). Since the main objective of thermal treatment of drill cuttings is not to char them but to reclaim the drill cuttings for possible land application or other forms of reuse, incineration method is less frequently used in the treatment of drill cuttings. Nonetheless, incineration has greater application in the treatment of medical wastes (Adeniran, 2003), which is outside the scope of the current review.

Thermal desorption: Thermal desorption refers to the indirect heating of drill cuttings to vapourize volatile and semi-volatile compounds without incinerating the drill cuttings (Figure 3). It is important to state that unlike incineration, thermal desorption does not produce ash as its end product since the aim is not to completely destroy

Figure 4. Heap of pre-treated drill cuttings from the anaerobic thermal desorption unit (ATDU) of a commercial treatment facility in the Niger Delta, Nigeria.
Source: Corresponding Author.

the cuttings by converting them to ash but to recover the oil-free cuttings. This distinction is important because thermally desorbed drill cuttings apparently have often been referred to as drill-cuttings ash in some literatures.

Low-temperature thermal desorption units (250 to 350°C) are used to remove light aliphatics and aromatics (e.g., benzene, toluene, ethylbenzene, and xylenes); while high-temperature thermal desorption units (up to 520°C) are used to remove heavier aromatics such as PAHs (Owens et al., 1993). Thermal desorption includes (among others) thermal phase separation (DWMIS, 2000), which consists basically of five subsystems, namely; the pretreatment unit, the anaerobic thermal desorption unit (ATDU), the solids handling unit, the vapour recovery unit (VRU), and the water treatment unit (WTU) (Zupan and Kapila, 2000). At the pretreatment unit, the drill cuttings are screened to remove foreign and oversized materials (>50 mm in diameter) before delivery to the ATDU. The drill cuttings are heated indirectly in the ATDU under an oxygen deficient condition to a temperature equal to the boiling point of the hydrocarbons, usually about 220°C but sometimes up to 520°C. At this temperature, the hydrocarbons are volatilized and separated from the drill cuttings. The

processed drill cuttings exiting the ATDU are received in the solids handling unit where they are cooled and kept in a storage area (Figure 4).

The gases (mixture of water vapour and gaseous hydrocarbons) exiting the ATDU enter the VRU housing the scrubbers. The condensed liquids and recirculated quenched water are then sent to the WTU where oil is separated from water. However, because of the indirect heating system of the ATDU, large heating surfaces that consume large amounts of energy and space are needed (Zupan and Kapila, 2000). Moreover, the operating temperature of 520°C for high temperature TDUs could lead to thermal degradation and decomposition of the residual drill cuttings (DWMIS, 2000). It has also been reported that thermal desorption is associated with high cost implications (Bansal and Sugiarto, 1999), environmentally threatening consequences such as high-level occupational hazard resulting from heat and fugitive dust emissions from the ATDU (DWMIS, 2000), and costly delays due to the length of time spent in the separating units (Ball et al., 2012). Furthermore, using low temperature thermal desorption to treat oil-based drill cuttings containing up to 21.8% of oil; Onwukwe (2014) reported that only 0.33% of the oil were left in the drill

cuttings after 45 min of treatment. They suggested that thermal desorption can remove a reasonable amount of oil from oil-based drill cuttings.

Microwave-assisted heating: In microwave-assisted heating of materials, energy is transferred directly to substances. This causes every single element in the material to be heated instantly through molecular or ionic vibrations under the effect of an oscillating magnetic or electric field (Mutyala et al., 2010; Robinson et al., 2009). Although, microwave-assisted heating technology is not applicable to every material, it is becoming increasingly attractive in the treatment of oilfield drill cuttings because of the presence of microwave-absorbing materials like carbon and water in drill cuttings (Robinson et al., 2008). In one study, Robinson et al. (2009) used a pilot-scale microwave heating system to remediate oil-based drill cuttings, reducing the residual oil contents to less than 1% of its initial amount. However, during the microwave remediation of drill cuttings, Petri et al. (2015) reported that a high initial drilling fluid content in drill cuttings reduced the oil removal efficiency of the microwave heating method. Generally, microwave heating method has not been commercialized on a large scale as yet due to the huge financial implications for initial setup; as only a few small-scale commercial applications of the technology have been reported in the literature (Mutyala et al., 2010).

The enumerated challenges associated with the non-biological methods prompted the shift of emphasis to biological treatment technologies like bioremediation, which according to several literatures has high potential for restoring polluted media with least negative impact on the environment at relatively low cost.

Biological method

Bioremediation: This is the use of microorganisms such as bacteria and fungi to enhance the natural decomposition of hydrocarbon-contaminated wastes into nontoxic residues (Atlas and Bartha, 1972). Unaided bioremediation can take a long time (up to 2 years) to achieve complete decontamination of hydrocarbon-contaminated matrixes (KMC Oiltools, 2005); thus, different strategies have been evolved to enhance the bioremediation process. These strategies are broadly categorized into ex-situ and in-situ techniques including (but not limited to) composting with decomposing organic materials in biopiles and/or windrows (Coulon et al., 2010), biostimulation using organic or inorganic fertilizers (Ayotamuno et al., 2009), bioaugmentation using bacteria isolates (Okparanma et al., 2009), mycoremediation using fungi species (Okparanma et al., 2011), remediation by enhanced natural attenuation (RENA) (Ebuehi et al., 2005), and onsite exsitu RENA (Okparanma et al., 2017). At the laboratory level,

composting with farmyard manure and poultry droppings, bioaugmentation using two strains of bacteria (*Pseudomonas aeruginosa* and *Bacillus subtilis*), and mycoremediation using white-rot fungus (*Pleurotus ostreatus*) have been applied in the bioremediation of drill cuttings (Okparanma et al., 2009, 2011; Ayotamuno et al., 2009, 2010). However, it has been suggested that field-scale treatment of drill cuttings by RENA whether in biopiles or windrows has the potential to pollute local groundwater sources and faces limited land availability challenges because of large-space requirement (Ayotamuno et al., 2010; Maduekwe et al., 2016). Similarly, bioaugmentation has been reported to have huge field-scale adaptability challenges (KMC Oiltools, 2005), which include: (1) overarching issue of the ability of the laboratory-prepared microbes to adapt to field conditions; (2) requirement of large volume of bio-preparation of microbial consortium for use in the field; and (3) associated huge financial burden. For mycoremediation, there is no information as yet in the literatures on the field-scale application of the strategy in the bioremediation of drill cuttings. Meanwhile, current researches are focusing on the possibility of beneficially and/or sustainably reusing pre-treated drill cuttings.

REUSE OF PRE-TREATED DRILL-CUTTINGS AS CONSTRUCTION MATERIAL

The world over, the costs of cement as well as fine and coarse aggregates is quite high due to rising inflation. This has prompted the need to look at other potential substitutes for these construction materials that are relatively cheap to obtain. As stated, pre-treated drill cuttings is increasingly becoming recognised as a potential substitute for cement in concrete (Chen, 2007), as aggregate in cold-mix asphalt for road pavement construction (Allen et al., 2007), and as fill material for road construction and fine aggregate in concrete production (IOGP, 2016). Chen et al. (2007) used drill cuttings as partial substitute for cement in the manufacture of bricks and concrete for building construction. They reported that the compressive strength and permeability of the resulting bricks were adequate for building purposes. However, the resulting concrete showed variable compressive strength and permeability and was sensitive to the mix design (Chen et al., 2007). Allen et al. (2007) reported the use of drill cuttings as aggregate in cold-mix asphalt for road pavement construction. Initial constraints, which according to Allen et al. (2007) were later overcome, were the high moisture and hydrocarbon contents of the drill cuttings (Table 1). It should be pointed out at this point that detailed comparison of the possible management technologies for drill cuttings in terms of portability, relative cost, commercialization, environmental impacts, advantages, and constraints is well documented in the literatures.

Interested readers are referred to IOGP (2016) for further information.

Concrete: Mix ratios and concrete grades/strength classes

Concrete is generally used for the production of structural elements used for the construction of buildings and/or trafficked pavements (Igwe et al., 2009). Concrete usually is produced by mixing Portland-limestone cement with fine and coarse aggregates and water; and allowing the mix to cure for a period of 28 days (Adewole et al., 2015). It is important to state that the curing period of 28 days is only limited to some laboratory-scale studies (Anonymous Reviewer). The type of loading to which the concrete is subjected when in normal use is determined by its grade/strength class, which is dependent on the mix ratio. While the most common mix ratio is the 1:2:4 (cement–fine aggregate–large aggregate); other mix ratios such as the 1:1.5:3 and 1:1:2 have also been tested (Adewole et al., 2015). Concrete grade/strength class indicates the minimum compressive strength of concrete, which is taken as the 28-days crushing strength of concrete cubes (150 × 150 × 150 mm) or cylinders (150 dia. × 300 mm) (BSI, 2004). According to BSI (2004), the concrete grade/strength class of C20/25, for example, should have at least a cylinder and cube crushing strengths of 20 and 25 MPa, respectively. This concrete grade/strength class is largely recommended in the design of reinforced concrete columns, beams, and slabs for use under mild exposure condition (Mosley et al., 2007). To achieve the minimum compressive strength of concrete recommended for use under a given loading condition, it is also important that the appropriate grade of cement is used in the mix design to avoid structural failure (Adewole et al., 2015).

Nonetheless, before putting concrete produced with pre-treated drill cuttings as partial substitute either for cement or fine aggregate to the intended use, it is first analyzed to check if it is fit for purpose. This is in relation to compliance with environmental guidelines and recommended minimum compressive strength of concrete. More often than not, conventional methods for the determination of the physicochemical properties of the resulting concrete have a variety of challenges, which call for alternative approaches.

Conventional methods of concrete characterization and their challenges

Determination of the organic and inorganic contents of concrete such as hydrocarbons, chloride, and heavy metals are based on laboratory wet-chemistry analyses. The protocols generally adopted include slicing and crushing of the concrete, acid digestion of the crushed concrete, solvent extraction and clean-up of the extract,

and analysis of the extract either by gas chromatography or chemical titration. For hydrocarbon analysis, a gas chromatograph coupled to either a flame ionization detector or mass selective detector is used according to USEPA method 8270 (USEPA, 1994). For heavy metal analysis, an atomic absorption spectrophotometer is used according to APHA method 3111C (APHA, 1998). Both the water extractable and the acid soluble chlorides are analyzed using potentiometric titration with silver nitrate according to AASHTO method T26097 (AASHTO, 2005). According to a recent review (Okparanma and Mouazen, 2013a), solvent extraction is both time-consuming and associated with ecological health risks because extraction solvents and stock solutions often used in the extraction process and *a priori* instrument calibration respectively are carcinogenic. In the same vein, analytical cost per sample for hydrocarbons using gas chromatography-mass spectrometry (GC-MS) is very high and the process is slow in addition to the need for an experienced operator to run the analysis and later process the data. This implies that the GC-MS method is a major economic burden to researchers when dealing with a large sample size. Moreover, the total dependence of the Cube Crushing Machine on electricity during the determination of compressive strength of concrete cubes in the laboratory is a huge limitation especially in areas where regular power supply is unreliable. Again, since the Cube Crushing Machine is not portable, it will require lots of labour and man-hour to determine the compressive strength of a large number of concrete cubes. Therefore, there is a need to evolve a rapid, cheap, portable, and ecologically sound approach for the determination of the physicochemical characteristics of concrete produced with pre-treated drill cuttings as fine aggregate.

NEAR-INFRARED (NIR) SPECTROSCOPY

As stated, because of the challenges associated with wet chemistry methods of concrete analysis, alternative methods are now emerging. One of these alternatives is the optical method based on visible (vis), NIR, and mid-infrared (MIR) spectroscopy. Compared to MIR, NIR is more robust for in-situ measurement and less sensitive to moisture. However, as subsequently discussed, the current application of vis-NIR spectroscopy in concrete analysis has been largely focused on the determination of chloride concentrations in concrete structures. This is despite the fact that it is well known that chloride is not the only contaminant usually found in concrete (IOGP, 2016). Meanwhile, to drive home the aim of this review, it would be worthwhile to briefly describe the fundamental principles of NIR spectroscopy.

Principles of NIR spectroscopy

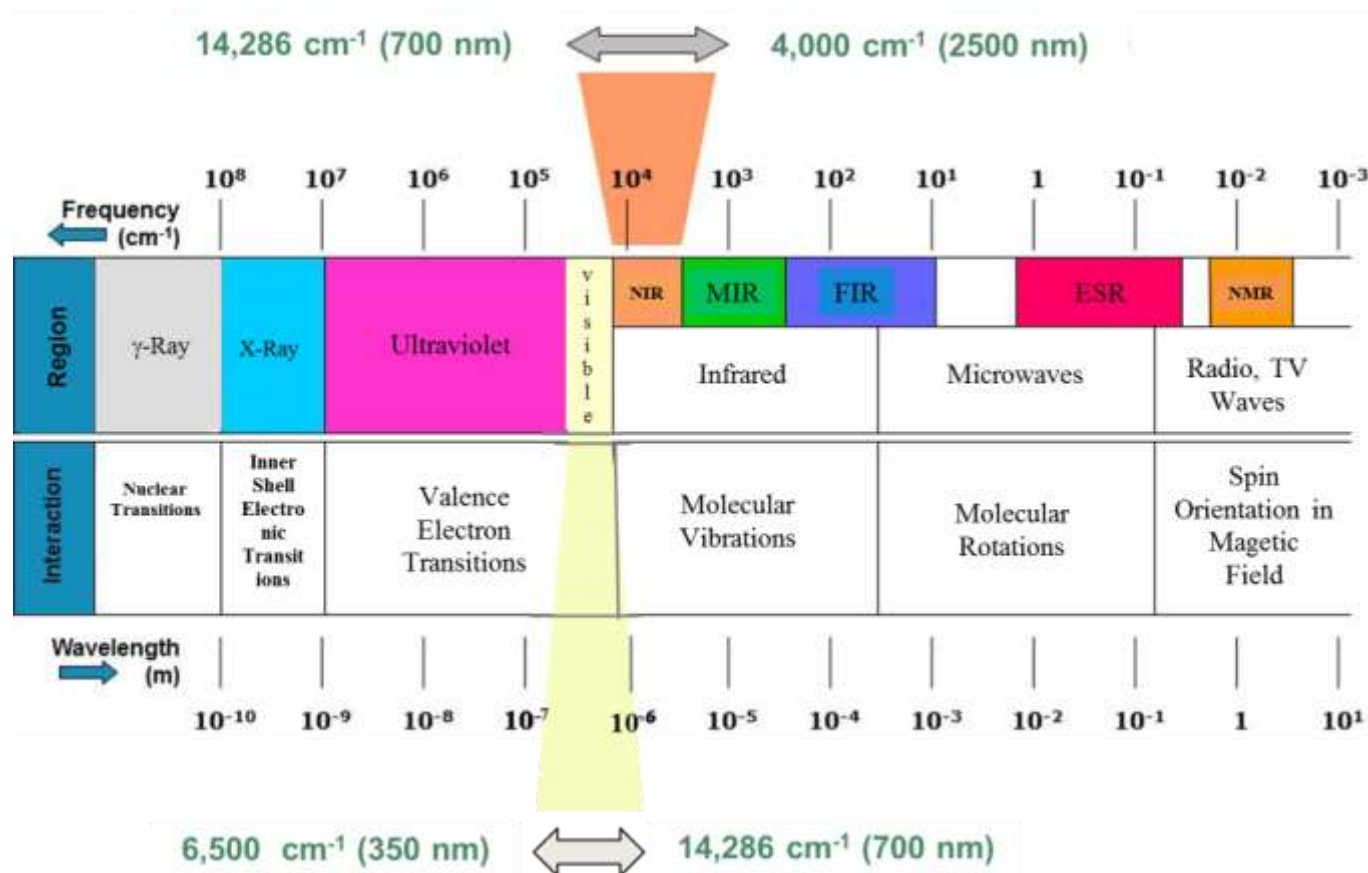Figure 5 shows the different regions in the electromagnetic

Figure 5. The electromagnetic spectrum. (Source: Bruker Optik GMBH https://www.bruker.com/products/infrared-near-infrared-and-raman-spectroscopy.html; reproduced with permission of Bruker Optik GMBH but with slight modification) NIR = Near-Infrared, MIR = Mid-Infrared, FIR = Far-Infrared, ESR = Electron Spin Resonance, NMR = Nuclear Magnetic Resonance.

spectrum. In the current review, only the vis and NIR regions are of interest because of their versatility in the analysis of a wide range of samples including both solids and liquids such as cereals, soils, powders, oils, etc.; all in one single stand-alone device. In the electromagnetic spectrum, the vis region is around 350–700 nm while the NIR region is around 700–2500 nm (Figure 5). Thus, vis-NIR spectroscopy covers the region around 350–2500 nm. The use of vis-NIR spectroscopy in soil physicochemical analysis both spatially and temporally is now an all too common practice.

Vis-NIR spectroscopy is based on the interaction of electromagnetic waves (radiation) with sample constituents (matter). The principles of vis-NIR diffuse reflectance spectroscopy involving the interaction of electromagnetic waves with sample constituents is shown in Figure 6. Unlike specular reflected light (related to mirror-like surfaces), diffuse reflected light (related to powder-like material such as soils) interacts with the constituents of the samples; and its spectrum on a detector provides qualitative and/or quantitative information about the sample. Therefore, the spectrum of a sample is unique — like a fingerprint; and is used to

identify the sample. The detector is equipped with an array of Michelson interferometers and filters. There are different types of detectors, including diode arrays, monochromators, etc.

Origin of NIR spectroscopy

According to Davies (2005), NIR was discovered by William Herschel in 1800. With inputs from Ampere in 1835 and James Maxwell in 1864, the first NIR spectra were measured in 1881 by Abney and Festing. Following the successes recorded by Abney and Festing by accurately predicting that spectral absorptions were related to the chemical composition of liquids, Coblentz in 1905 published the spectra and corresponding wavelengths (1000–10,000 nm) of a large list of compounds. This paved the way for vibrational spectroscopy of molecules in the MIR (2500–50,000 nm) region. Then in 1960 Karl Norris of the United States Department of Agriculture showed the effectiveness of the NIR region in quantitative analysis of agricultural samples.
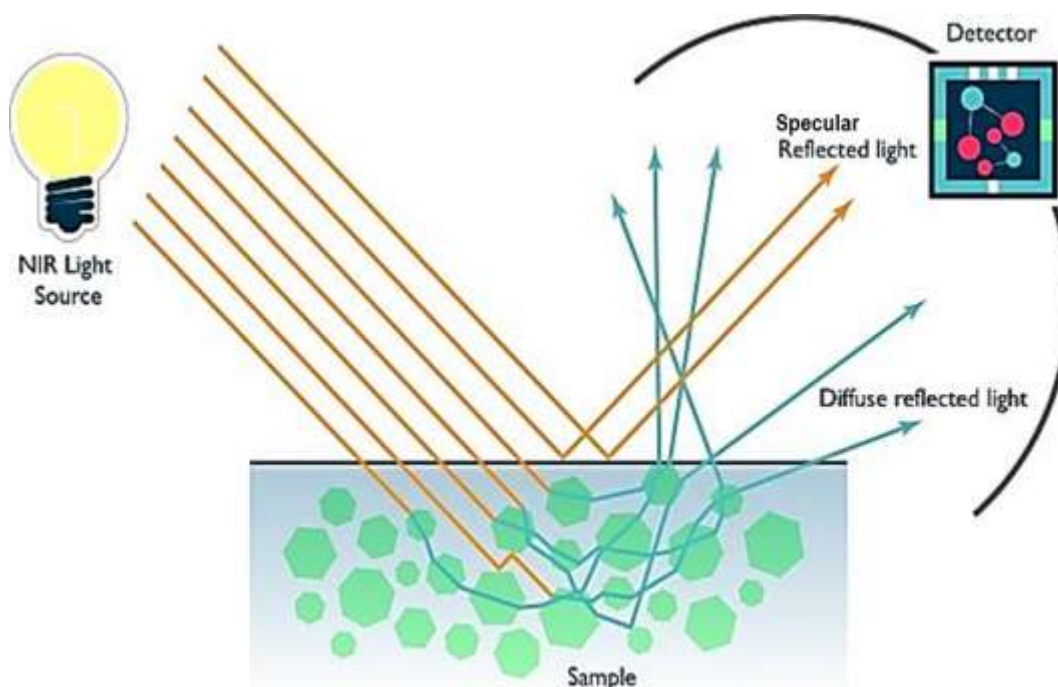
Figure 6. Principle of near-infrared diffuse reflectance spectroscopy (Source: Pulp & Paper, Canada http://www.pulpandpapercanada.com; reproduced with permission of Pulp & Paper, Canada).

Scattering and absorption of energy (light) by sample in the vis-NIR region

Scattering and absorption of light by solid samples in the vis-NIR region are two important phenomena that influence the measurement (in diffuse reflectance mode) of both physical and chemical properties of solid samples.

(a) Scattering of light by sample: Light scattering by solid samples (Figure 6) is associated with the physical characteristics of the sample (e.g., particle size), as well as the chemical characteristics such as hydrocarbon content (among others). When light is incident on a solid sample, the light is scattered in different directions depending on the geometry and size of the solid granule, as well as the wavelength and frequency of the incident light. If the light scattering is elastic, it is referred to as Rayleigh scattering (Kaminow and Li, 2002). Rayleigh scattering intensity is largely dependent on the particle size of the sample; it is proportional to the sixth power of the particle diameter and inversely proportional to the fourth power of the wavelength of light (Kaminow and Li, 2002). Therefore, Rayleigh scattering intensity is stronger around 450 nm (blue colour) than 680 nm (red colour) in the vis region; suggesting that the larger the particle size of the sample is, the stronger is the Rayleigh scattering intensity and vice versa. On the other hand, if the light scattering is inelastic, it is referred to as Brillouin or Raman scattering. Although central to Brillouin and

Raman scattering is the interaction of light with the constituents of the solid sample, both types of light scattering are different in the way the light interacts with the solid sample (Kaminow and Li, 2002). Raman scattering may be used to assess the vibration of Raman-active molecules such as the PAHs, as well as for identifying species. According to a recent review (Okparanma and Mouazen, 2013a), qualitative and quantitative applications of Raman spectroscopy is widespread.

(b) Absorption of light by sample: Absorption of light by solid samples is linked to both the physical characteristics (e.g., colour) and chemical characteristics (e.g., water, hydrocarbons, amines, clay minerals) of the sample. In the vis region of the electromagnetic spectrum (Figure 5), absorption of light by solid samples is linked to colour and electron excitation. Colour associated changes due to colour absorption bands around 450 nm (blue colour) and 680 nm (red colour) have been linked to changes in spectral geometry of solid samples (Mouazen et al., 2007; Okparanma and Mouazen, 2013b). For instance, soil diffuse reflectance is known to decrease as the soil gets darker in colour due to increased light absorption, and vice versa (Mouazen et al., 2007). Similarly, oil contamination in the presence of water is known to decrease soil darkness with increased soil diffuse reflectance (Okparanma and Mouazen, 2013b). Newer concrete has also been reported to have an overall higher reflectance than older concrete due to age
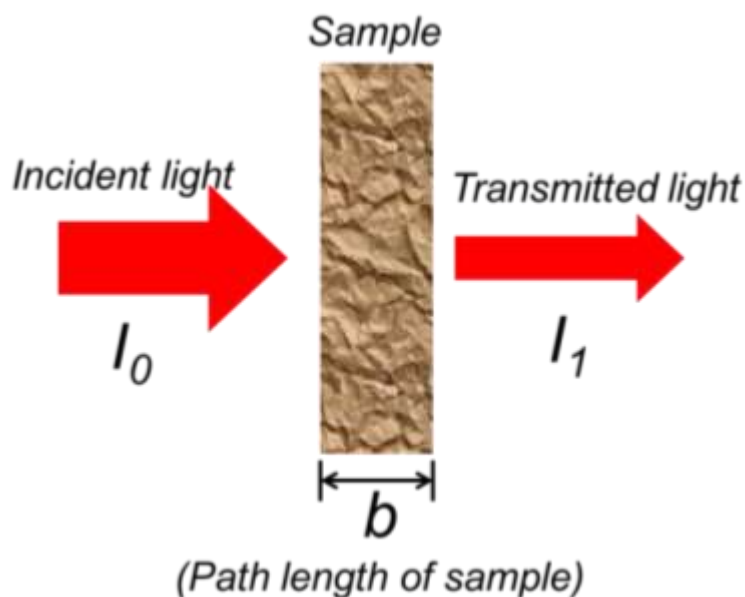
Figure 7. Process of light absorption by a sample.

and contamination with other materials such as soil and vegetation (Fairbarn, 2013). This may be explained by the change in colour of concrete with age; the newer concrete being lighter in colour than the darker older concrete. On transparency of solid materials, though, increased transparency of newer materials results in overall decrease in reflectance while older materials with rougher surfaces have higher reflectance (Nasarudin and Shafri, 2011). From classical physics, it is known that when light is incident on solid material, some of the light is absorbed and electrons within a molecule are excited as they transit from a lower energy level to a higher one. This occurs at certain wavelengths where the energy of the incident light corresponds to an electronic transition within the molecule. The resulting absorption spectrum, which is a function of absorbance and the wavelength at which the absorption occurs, is often used to identify the chemical constituents of the material.

In the IR region of the electromagnetic spectrum (Figure 5), molecular vibration is due to absorption of light by sample linked with the type of chemical bonds between atoms in a molecule. The process of light absorption by a sample is illustrated in Figure 7. With soil sample, light absorption by a sample occurs if $I_1 < I_0$ (Figure 7).

Absorptions in the NIR region require short path length of sample. Energy absorption by a sample is explained by simple harmonic (parabolic) approximation. Absorptions in the IR region are due to overtones and combinations of fundamental absorptions (occurring in the MIR spectral region) due to deviation in molecular vibration from the harmonic approximation (discussed shortly), which holds true only when almost all the

molecules are in their vibrational ground state (fundamental vibration) in which the dominant spectral transition is the fundamental transition (Atkins and de Paula, 2002). This is because most absorption in the NIR region requires higher vibrational energy (Davies, 2005). At high vibrational excitation, the parabolic approximation tends to break down at some regions in the potential energy curve and the concomitant anharmonicity results in overtones and combinations of fundamental vibrations (Atkins and de Paula, 2002; Osborne et al., 1993; Pasquini, 2003). With hydrogen atom dominating NIR spectroscopy (forming C–H, O–H, N–H, and S–H chemical bonds), these overtones and combinations are assigned bands as shown in Figure 8 involving different modes of hydrogen stretching and bending. Stretching refers to vibration involving axial change in the interatomic distance between two bonding atoms while bending refers to vibration involving change in bond angle (Osborne et al., 1993). Stretching could be symmetrical or asymmetrical while bending could be scissoring, rocking, wagging, or twisting as dictated by the direction of bond vibration (Osborne et al., 1993). Since overtones and combinations are of broad bands (several wavelengths), chemometrics or machine learning techniques are commonly used to extract useful quantitative and qualitative information from the vis-NIR spectra.

Chemometrics and NIR spectroscopy

The initial set back in the study of NIR spectra was partly attributed to the requirements of a knowledge of
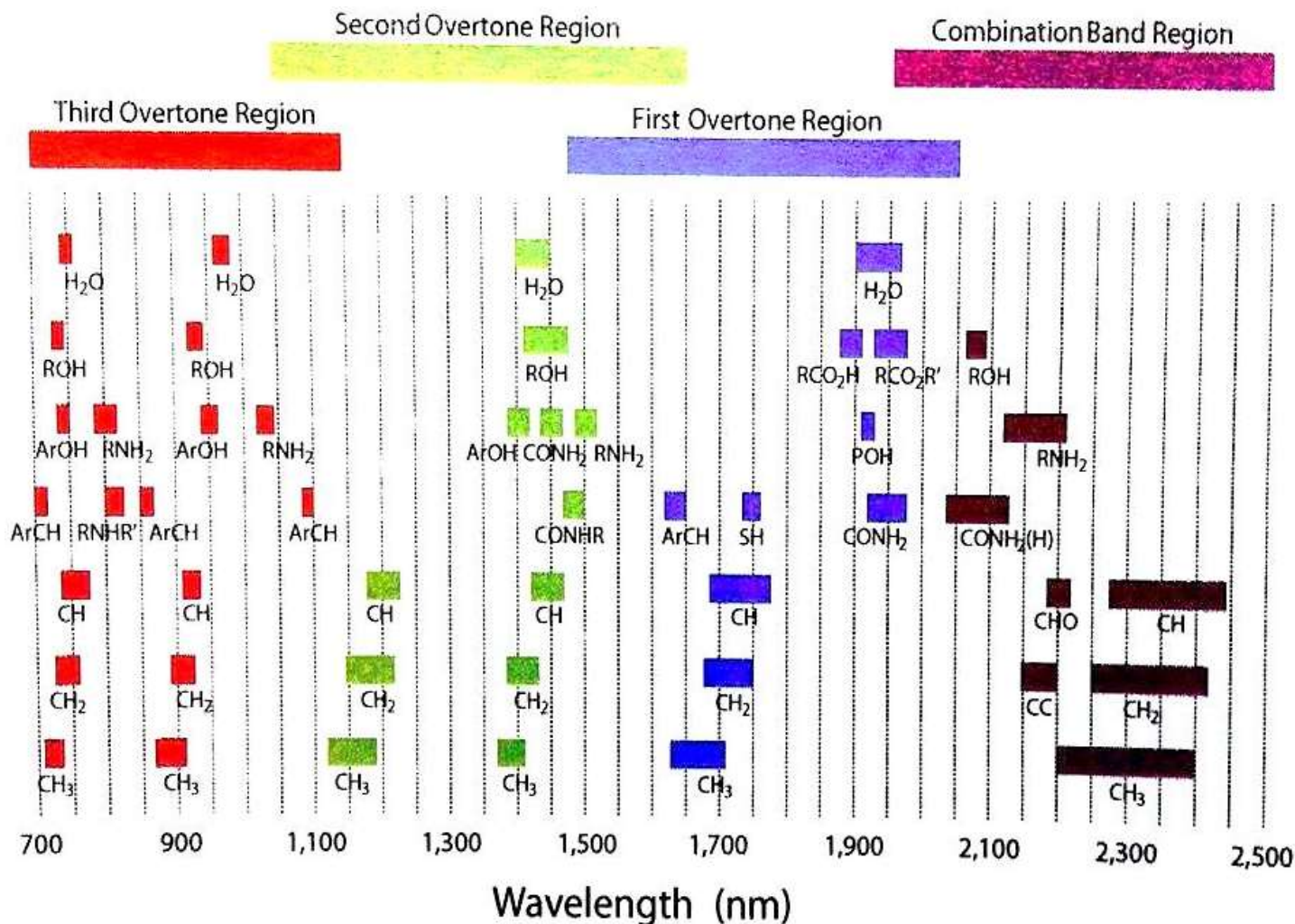
Figure 8 NIR band assignment table.
Source: Metrohm Monograph 8.108.5026EN – A guide to near-infrared spectroscopic analysis of industrial manufacturing processes. Metrohm AG, CH-9101 Herisau, Switzerland.

chemometrics, the computer, and use of very low-noise spectrometers; and the person who put all this together was Karl Norris (Davies, 2005). Chemometrics is the use of advanced statistics and mathematics in multivariate analysis (MVA) — calibration and classification. The apparent 'miscibility' between chemometrics and NIR spectroscopy stems from the dependence of diffuses reflectance measurement on the highly variable physical properties of a sample. This variability in physical properties causes changes in the resulting spectrum. So, to extract the relevant information necessary for a given analytical procedure, variables as high as between 20 and 1000, in most cases, need to be considered (Pasquini, 2003). This usually is implemented by employing both linear and non-linear MVA for qualitative and quantitative applications. Some linear MVA techniques that have been adopted include multiple linear regression (MLR), principal component regression (PCR),

partial least-squares (PLS) regression analysis and penalised spline while the non-linear models include artificial neural network (ANN), support vector machine (SVM), boosted regression trees (BRT), random forest (RF), and wavelet (Mouazen et al., 2010; Okparanma and Mouazen, 2013b; Gammoun et al., 2006, 2007; Pérez-Caballero et al., 2009; Chakraborty et al., 2010, 2012; Dos Santos et al., 2010; Viscarra Rossel et al., 2010; Okparanma et al., 2014a, b).

However, since solid samples are known for scattering radiation in diffuse reflection thereby making it difficult to clearly discriminate among the spectra due to background noise, it becomes imperative to have the spectra pre-treated to correct for the physical scatter effects in order to make them suitable for qualitative and/or quantitative applications (Viscarra-Rossel et al. (2006a) has a list of some applications). Spectral pre-processing techniques that have been applied include

(but not limited to) the following: (1) averaging replicate spectra, (2) maximum normalisation, (3) fitting a weighted (inverse measurement variance) smoothing spline to each spectra with direct extraction of smoothed reflectance, (4) use of first and second derivatives of the original spectra, (5) individually combining the resulting average reflectance with the first- and second-derivatives, (6) multiplicative and extended multiplicative scatter correction, (7) Savitzky-Golay, and (8) standard normal variate (Mouazen et al., 2010; Dematte et al., 2004; Viscarra-Rossel et al., 2006b; Westad et al., 2008; Yu et al., 2008; Sundaram et al., 2010; Chen et al., 2010; Kuang and Mouazen, 2011). Alternatively, variable-selection techniques, which minimize the number of simultaneous variables to be used in modelling, have also been applied in this regard. Some variable-selection techniques include principal component analysis (Mouazen et al., 2010), uninformative variable elimination (Chen et al., 2010) and jack-knife (Westad and Martens, 2000).

Usually, the calibration methods that provide the largest accuracy are optimized using cross-validation procedures (Westad et al., 2008) and validated independently with independent validation set not used in the cross-validation procedure. The model performance in terms of the root mean square error (RMSE) of validation or prediction, as well as residual prediction deviation (RPD) and the corresponding coefficient of determination (r^2) may be evaluated. But among the calibration sample set, it is possible for differences between the NIR-predicted and reference values to occur. So, the pre-processed spectra are normally checked for these differences by subjecting the dataset to a priori outlier detection techniques such as principal component analysis (PCA), influence plot etc. (Mouazen et al., 2010; Okparanma and Mouazen, 2013c). Detected outliers are then removed by cluster analysis (Pérez-Caballero et al., 2009). The pre-processing of different individual and combined spectra usually is implemented using software packages such as the Unscrambler-X (CAMO Software AS, Oslo, Norway), while multivariate calibrations are implemented with packages like the MatLab v. 7.1 (The Math Works, Natick, MA, USA), Unscrambler-X, Pirouette (Infometrix, Seattle, WA, USA), TQ Analyst (Thermo Nicolet Corp., Madison, Wisconsin), among others.

Theory of the practical applications of NIR spectroscopy

Beer-Lambert law defines a linear relationship between reflectance (R) and concentration (C) of a substance and assumes the substance to be homogenous. This implies that Beer-Lambert Law will not be valid for soil — a heterogeneous substance. Therefore, for quantitative analysis of soil, Kubelka and Munk modified the Beer-Lambert law. The Kubelka-Munk (K-M) remission function is given by Equation 1.

$$f(C) = \frac{(1-R)^2}{2R} \tag{1}$$

$$\text{And } R = \frac{I_R}{I_{RO}} \tag{2}$$

Where, I_R = Intensity of reflected radiation by sample and I_{RO} = Intensity of reflected radiation by a white-referencing Spectralon disc of approximately 100% reflectance. However, measurement of I_R and I_{RO} is complicated. Therefore, for practical purposes in diffuse reflectance mode, R is transformed to absorbance as given by the modified K-M function in Equation 3.

$$f(C) = \text{Log} \frac{1}{R} \tag{3}$$

APPLICATIONS OF NIR SPECTROSCOPY — PAST AND PRESENT

According to reports, initial applications of NIR spectroscopy were for qualitative purposes, especially in the foods and beverages industry (Osborne et al., 1993). However, NIR spectroscopy gained acceptance for both qualitative and quantitative applications in soil science in the 1980's (Adamchuk et al., 2004). In recent times, the application of NIR spectroscopy has transcended soil science. The reasons for this attraction may not be too far from its simplicity, portability, non-destructiveness, cost effectiveness, rapidness, and little or no associated ecological health risks.

Soil science

A plethora of literature can be found on the use of vis-NIR spectroscopy for the rapid assessment of a wide range of soil physicochemical properties including pH, soil colour, soil texture, lime requirement, organic carbon, total carbon, total nitrogen, plant-available P, K, Mg, exchangeable Al, Ca, K, electrical conductivity (Mouazen et al., 2010; Viscarra Rossel and Behrens, 2010; Viscarra Rossel et al., 2006a, b; Mouazen et al., 2006, 2007, 2009; Mouazen, 2006; Maleki et al., 2006; Gomez et al., 2008a; Canasveras et al., 2010; Wetterlind et al., 2010). Other soil physicochemical properties are soil inorganic carbon, total iron, mineral composition, cation exchange capacity (Brown et al., 2006; Gomez et al., 2008b), moisture content, soil roughness, surface crust (Whiting et al., 2004; Mouazen et al., 2005; Dilawari et al., 2006), nitrogen Kjeldahl (NKj) content (Guerrero et al., 2010). Additionally, biological properties such as respiration rate by CO_2 evolution (Chang et al., 2001) and biological and chemical changes occurring during the co-composting of green waste and sewage sludges (Albrecht et al., 2009) have been characterised with NIR spectroscopy. Certain variables relating to humification of sewage sludge (Dos Santos et al., 2010) and stability of sewage sludge and

Table 2. Application of vis-NIR spectroscopy in the determination of organic and inorganic contaminants in environmental matrixes.

Environmental matrix	Target contaminant	MVA employed	Wavelength range (nm)	References
Soil	TPH	SMLR	1100-2498	Malley et al. (1999)
Water	*n*-hexane, isooctane, and toluene	PLSR	909-2000	Gammoun et al. (2006)
Water	BTEX	NR	800-2500	Lima et al. (2007)
Water	Very light crude oil to heavy fuel oil	PCA, MPR	1000-2500	Pérez-Caballero et al. (2009)
Soil	TPH	PLSR, BRT	350-2500	Chakraborty et al. (2010)
Soil	TPH	PLSR	400-2500	Forrester et al. (2010)
Soil	PAH	OLR	350-2500	Bray et al. (2010)
Water	Organophosphorus pesticide (*Chlorpyrifos*)	PLSR	1000-2500	Chen et al. (2010)
Soil	TPH	PLSR	NR	Schwartz et al. (2011)
Soil	TPH	PLSR, ANN	NR	Schwartz et al. (2012)
Soil	TPH	PLSR, MLR, PS	350-2500	Chakraborthy et al. (2012)
Soil	TPH	PLSR	2222-2439	Forrester et al. (2013)
Soil	PAH	PLSR	350-2500	Okparanma and Mouazen (2013b)
Soil	Mn, Pb, and Zn	PLSR	400-2500	Gannouni et al. (2012)
Soil	Mn, Cu, Cd, Zn, and Pb	SVMR	350-2500	Gholizadeh et al. (2015)
Soil	Cd, Cr, Pb, Cu, Zn, Hg and As	UR	400-2500	Ji et al. (2010)
Soil and Plant	As	SMLR	350-2500	Shi et al. (2016)

ANN, Artificial neural network; BRT, Boosted regression tree; BTEX, Benzene-toluene-ethylbenzene-xylene; MLR, Multiple linear regression; MPR, Multivariate polynomial regression; NR, Not reported; OLR, Ordinal logistic regression; PAH, Polycyclic aromatic hydrocarbon; PCA, Principal component analysis; PLSR, Partial least-squares regression analysis; PS, Penalized Spline; SMLR, Stepwise multiple linear regression; SVMR, Support vector machine regression; TPH, Total petroleum hydrocarbon; UR, Univariate regression.

derived compost (Soriano-Disla et al., 2010) have been predicted with NIR spectroscopy with acceptable prediction errors.

Petrochemical and wastewater industries

As stated, the expanding scope of vis-NIR spectroscopy in the analysis of environmental media has transcended soil science. For instance, vis-NIR spectroscopy has found application in the petrochemical industries for gasoline classification (Balabin and Safieva, 2008) and identification of polymers and resins in painting materials (Vagnini et al., 2009). Vis-NIR diffuses reflectance spectroscopy (DRS) (350-2,500 nm) and vis-NIR spectroscopy (909-2000 nm) in transmittance mode has been respectively used to determine the amount of hydrocarbons (toluene, isooctane and n-hexane) sorbed on chrome shavings and the adsorption capacity of the chrome shavings during the remediation of hydrocarbon-polluted waters. Using partial least-squares (PLS) regression and BRT, Gammoun et al. (2006) reported an adsorption capacity of 6.30 g hydrocarbons per g solid. A similar study was carried out to separate motor oils, oily wastes, and hydrocarbons in water (Gammoun et al., 2007). Fourier transform (FT) NIR spectroscopy (14,000–3,500 cm^{-1}) in transmittance mode and PLS regression analysis were employed to estimate the amount of motor oil, oily waste, and CCl$_4$ leached-hydrocarbons (toluene, isooctane or *n*-hexane) removed by chrome shavings

in water. In the wastewater treatment industries, aromatic hydrocarbons such as benzene, toluene, ethylbenzene and xylene (BTEX) in water have been determined with NIR spectroscopy (Lima et al., 2007) as well as concentrations of pesticide in water (Chen et al., 2010).

Environmental pollution monitoring and control

Currently, vis-NIR spectroscopy is becoming increasingly recognized as a rapid, cost-effective, portable, and ecologically sound technique for determining organic and inorganic contaminants in various forms of environmental matrixes including soil and water (among others). Table 2 shows a number of applications of vis-NIR spectroscopy in the determination of organic and inorganic contaminants in environmental matrixes. As can be seen in Table 2, hydrocarbon-based contaminants such as TPH, PAH, BTEX, etc. have all been determined by vis-NIR spectroscopy. The vis-NIR spectra of petroleum derivatives is known to originate mostly from overtones or combinations of C-H stretch vibration modes of CH$_2$ and CH$_3$, or aromatic C-H (ArCH) functional groups (Aske et al., 2001). As shown in Figure 8, these functional groups have absorption features around 712, 750, 850, 900, 1097, 1137, 1162, 1227, 1417, 1675, and 1712 nm in the NIR region. Similarly, inorganic contaminants such as heavy metals, especially the transition elements like Cr, Mn, Co, Ni, and Cu have been predicted using vis-NIR spectroscopy (Table 2). It is important to point out that

the determination of hydrocarbons and heavy metals using vis-NIR spectroscopy have often been made possible by the joint application of vis-NIR spectroscopy and a variety of multivariate statistical analysis (Table 2). This is partly because some of these contaminants (e.g., PAH and heavy metals) do not have direct spectral responses in the vis-NIR regions (Okparanma and Mouazen, 2013c; Rathod et al., 2013); and are barely detectable directly, especially at very low concentrations.

Nevertheless, PAH and heavy metals in soils can be indirectly predicted with reasonable accuracies because of their co-variations with spectrally active soil properties like iron oxides, water, clay minerals, and organic matter (Choe et al., 2009; Okparanma and Mouazen, 2013c). In the vis region, iron oxides have spectral absorption features around 500 nm (Ben-Dor et al., 1999) due to bending modes of R-OH (where R is a metallic ion such as Al, Fe, Mn, or Si) (Viscarra Rossel et al., 2006b). In the NIR band, water absorption features are observed around 950, 1450 and 1950 nm due to O-H stretch vibration mode of O-H functional group (Whalley and Stafford, 1992). Organic matter have absorption features around 2150 and 2212 nm due to vibrational modes of long-chain C-H+C-H and C-H+C-C stretch combinations (Forrester et al., 2010). Similarly, spectral absorption features of clay minerals are observed around 2200 and 2300 nm due to vibrational modes of R-OH bend plus O-H stretch combinations (Clark, 1999; Viscarra Rossel et al., 2006b).

CHARACTERIZATION OF CONCRETE USING VIS-NIR SPECTROSCOPY — CURRENT STATUS

Currently, there are only a few studies in the literature on the use of vis-NIR spectroscopy for characterization of concrete (e.g., Song and Saraswathy, 2007; Brook and Ben-Dor, 2011; Toda et al., 2012; Lee et al., 2012). As stated, these studies mostly focused on the chemical characterization of concrete; particularly the determination of chloride concentration in concrete structures like bridges to monitor the rate of corrosion in the steel reinforcement. With vis-NIR spectroscopy, chloride content in concrete is usually predicted indirectly because chloride ion is said to have no direct spectral response in the vis-NIR region (Song and Saraswathy, 2007). Therefore, analysis of chloride ion has often been made possible by applying sodium chloride with absorption features around 1630-1650 nm in the NIR region on the target specimen and measuring the apparent diffusion coefficient of chloride ion, which has good correlation with the heat dissipation characteristics of concrete (Song and Saraswathy, 2007).

Meanwhile, from several literatures, it has been observed that chloride is not the only inorganic compound usually found in concrete. It is widely known that concrete contains large amounts of heavy metals predominantly from constituent metal oxides such as Al_2O_3, SiO_2, Fe_2O_3, MgO, CaO, etc. Moreover, due to residual oil content in pre-treated drill cuttings (IOGP, 2016), the presence of heavy metals and hydrocarbons in the concrete may be unavoidable when these cuttings are used as a partial substitute for fine aggregate in concrete. As stated, iron oxides, water, clay minerals, organic matter, and TPH are spectrally active in the vis-NIR regions. Thus, these compounds are amenable to vis-NIR analysis, suggesting that spectrally inactive heavy metals and PAH in concrete can be indirectly predicted with vis-NIR spectroscopy. However, since the advent of the use of pre-treated drill cuttings as fine aggregate in concrete, no studies can be found in literatures on the use of vis-NIR spectroscopy in the determination of these equally important chemical characteristics of concrete. This is despite the successful application of vis-NIR spectroscopy in the determination of TPH, BTEX, PAH, and heavy metals in both soil and water (Table 2). This situation is the same for the pre-treated drill cuttings itself.

Apart from the chemical characteristics of concrete, other equally impotent physiognomies of concrete include (but not limited to) compressive strength, bulk density, and permeability, which are not spectrally active in the vis-NIR regions. Since these physical properties of concrete do not have direct spectral response in the vis-NIR regions, their direct determination by vis-NIR spectroscopy will not be possible. Nonetheless, by proximate analysis using suitable spectrally active surrogates such as iron oxides, water, clay minerals, or organic matter; it might be possible to determine most of these physical properties of concrete by vis-NIR spectroscopy. This is because most of these physical characteristics (e.g., compressive strength and permeability) have good correlations with water-cement ratio of concrete (Alawode and Idowu, 2011). Figure 9 illustrates the suggested technical approaches for the application of vis-NIR spectroscopy in the characterization of concrete produced with pre-treated drill cuttings.

RESEARCH NEEDS/KNOWLEDGE GAPS

It is clear that vis-NIR diffused reflectance spectroscopy has been extensively used in Soil Science and Petrochemical, Environmental, and Civil Engineering. Even though the work of researchers, including Song and Saraswathy (2007), Brook and Ben-Dor (2011), Toda et al. (2012), Lee et al. (2012), and Abbas (2015), has taken important steps towards addressing key knowledge gaps in the determination of chloride concentration in concrete structures using vis-NIR diffuse reflectance spectroscopy, there is still work to be done.

Further investigation is still necessary to increase our knowledge, within NIR circle, on the determination of
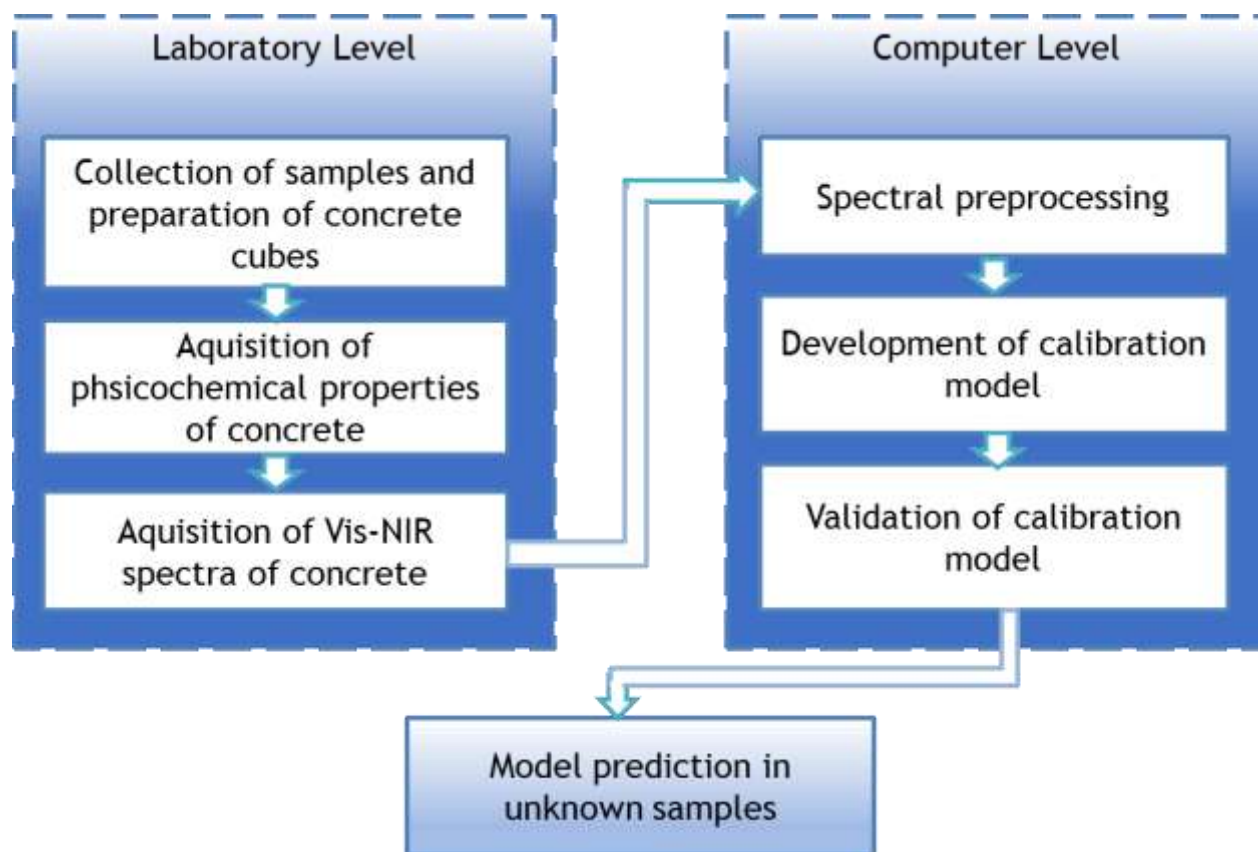
Figure 9. Proposed framework for application of vis-NIR spectroscopy in the physicochemical characterization of concrete produced with pre-treated drill cuttings.

other equally important physicochemical characteristics of concrete produced with pre-treated drill cuttings such as heavy metals, TPH, permeability, bulk density, compressive strength etc. This concrete characterization approach will significantly help to enhance the sustainable reuse of pre-treated drill cuttings as part substitute for fine aggregate in concrete by rapidly and economically characterizing resulting concrete to ensure that it complies with recommended minimum compressive strength and environmental guidelines for the protection of ecological health well into the future.

CONCLUSIONS

This review has shown that drill cuttings are the solid component of the waste stream from crude oil drilling operations. They are composed of organic and inorganic compounds such as hydrocarbons and heavy metals respectively; a consequence of the chemical formulation of the proprietary drilling mud used in the drilling process. Sea and land disposal of untreated dill cuttings have negative impacts on benthic and edaphic fauna and flora. There are currently several management options for drill

cuttings including (among others) their sustainable reuse for civil engineering works; and several factors are coalescing to making this option increasingly attractive. However, problems of lengthy sample preparation protocols, high analytical and labour costs, and occupational health risks associated with traditional concrete characterization techniques are now driving research towards the much faster, less expensive, portable, and less risky analytical technique based on vis-NIR spectroscopy.

The progress reported so far with vis-NIR spectroscopy in Soil Science and Petrochemical and Environmental Engineering, as well as prediction of chloride in concrete are indications that there could be opportunity for application of vis-NIR spectroscopy in the determination of other equally important physicochemical characteristics of drill cuttings as well as concrete produced with pre-treated drill cuttings like heavy metals, hydrocarbons, and compressive strength.

funding agencies in the public, commercial, or not-for-profit sectors.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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