



Production of silica gel from Tunisian sands and its adsorptive properties



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ABSTRACT

Thanks to its highly absorbent character, silica gel is used in several applications, such as air moisture removal, as a treatment agent for effluents. In this study, silica gels were synthesised from Tunisian sands, collected from the Fortuna and Sidi Aich Formations in northern and central Tunisia. The collected quartz sand raw materials, as well as the prepared silica gels, were characterised by different techniques, such as X-ray diffraction (XRD), X-ray fluorescence (XRF) and scanning electron microscopy (SEM). XRD patterns of quartz sands showed quartz as main phase (86.1–98%), with lower contents of potassic feldspars, along with kaolinite and calcite. These quartz sands presented relatively small quantities of Fe₂O₃ (0.3%–0.5%) and TiO₂ (0.1%–0.6%). The synthesised silica gels exhibited pore diameters exceeding 20 Å and surface areas up to 194 m²/g, comparable with those described in the literature and commercial silica gel. N₂ adsorption isotherms showed that the silica gels prepared from Tunisian sands are mesoporous materials with high adsorption capacities. To understand better their adsorbent properties and applicability on an industrial scale, these gels were tested for methylene blue (MB) adsorption. Maximum decolourisation rates (up to 96% after a contact time of 180 min) occurred with products synthesised at pH 3. The adsorption mechanism fitted better with a Langmuir model, revealing a monolayer coverage process of MB molecules over the gel surface, and the adsorption kinetics of the dye on these materials is well described by the second order model.

The corresponding equilibrium adsorption capacities obtained from experimental data ($Q_{exp} = 292$ –214 mg/g) were close to the estimated maximum adsorption capacities ($Q_e = 333$ –250 mg/g), and to that of an industrial silica gel (250 mg/g).

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1. Introduction

Current global silica sand world production for industrial purposes is around 140 Mt/year (Mansour, 2015), with the USA accounting for 24%, the Netherlands for 20% and France for 5%. The price of quartz sand varies as a function of its characteristics, such as particle size and purity. In the case of Tunisia, this raw material is essential to the economy, and various Tunisian manufacturers and industries (glass, foundry sand ...) are obliged to import huge quantities from countries like Belgium, France, Germany, Italy, Spain, the USA and Mexico, despite the obvious natural resources

that Tunisia possesses of this material. This operation is costly (1.8 Million USD in 2000 to 5.2 Million USD in 2015, INS 2016) for the state and the economy, while there exist abundant unstudied and unexploited silica sand deposits from the Early Cretaceous to Quaternary periods in the country (Jamoussi, 1991; Essid, 1997; Aloui, 1999; Harrabi, 1999; Sdiri et al., 2010; Hajjaji et al., 2009; Hajjaji, 2011). In addition, it has been shown that Tunisian sand is a high purity material that can be used in many different industrial areas, such as glass making (Hajjaji et al., 2009), cements (Aloui, 2010; Mechti et al., 2012), electro-metallurgy (Khalifa et al., 2014), chemicals (Mansour, 2015), and especially as air and water waste purifiers (Farrukh et al., 2014).

In this context, silica gel is an interesting amorphous material (annual world production 25,000 t/year; Meljac, 2004) with a high adsorption capacity (Gómez et al., 2014), and is used as an

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adsorbent of moisture, or for the purification of effluents with its hydrophilicity (the high moisture content trapped within the silica structure creating the ideal environment to attract polar impurities) (Armarego and Chai, 2013). This material has several advantages: for example, it is easy to regenerate, as when it has become saturated with water the silica gel can be regenerated by heating at 100–120 °C until it returns to its original state (indicated by a change in colour), the heating literally drives off the adsorbed moisture. Silica gel is also chemically inert and non-toxic, with a very high melting point, it is very much like quartz sand and thus can safely be sent by any means of transport, has a long life, and has low abrasion (the wear rates decreasing with increasing of the porosity) (Trabelsi et al., 2009). In recent years, new studies are underway into the use of this versatile amorphous material as fillers (Lin et al., 2014), coatings (Liu et al., 2013), and hygroscopic packaging for optical (Château, 2013), electronics (Xu et al., 2010) and pharmaceutical (Brown et al., 2014) products. Silica gel exhibits superior adsorption and high surface area properties which paves its way for use in water filtration, as growing environmental concerns, coupled with the need to comply with stringent government regulations concerning waste-water treatment, are driving the industry. The global water filtration market was valued over 17 million USD in 2012, and the transition towards the physical treatment of water from conventional water-treatment methods is expected to have an impact on the need for more silica gel (Grand View Research Report). The Global Silica Gel market is forecast to grow at a rate of 4% annually over the period 2014–2019 (Tech Navio Report, 2015).

Dyes are widely used by textile industries to colour their products, and one of the major problems concerning textile wastewaters is the highly coloured effluent (Moursli and Bengueddach, 2010), which can be difficult to decolourise. Over 15% of the textile dyes are lost in the wastewater stream during dyeing operations, and so dye pollutants from the textile industry are a significant source of environmental contamination. About half of the global production of synthetic textile dyes (700,000 tons per year) is classified as azo compounds that have the –N=N– chromophore unit in their molecular structure, such as methylene blue (C₁₆H₁₈ClN₃S). These azo dyes are known to be largely non-biodegradable in aerobic conditions, and they reduced to form more hazardous intermediates in anaerobic conditions. These releases pose a real danger to humans and the environment because of their stability and poor biodegradability (Sakr et al., 2015). Several treatments have been used to reduce the harmful effect of discharged effluents. Traditional methods such as biological processes give unsatisfactory results, due to the composition of these discharges toxic materials and dyes that are difficult biodegradable (Sakr et al., 2015). Adsorption remains among the most used techniques for dye removal, and is easy to implement. Removal of dyes in aqueous solutions by adsorption on various solid materials, especially on silica gel, has been the subject of many studies (Kushwaha et al., 2010; Samiey and Toosi, 2010).

Methylene blue (MB) (3,7-bis(dimethylamino)-phenothiazin-5-ium chloride trihydrate) is a cationic dye that is used in the dyeing of cotton, wool, coating for paper stock and silk. The classification provided by ECHA European Chemical Agency (2016) in CLP notifications (EC/List no.: 615-731-6) showed that this substance is harmful if swallowed, causes serious eye irritation, may cause damage to organs through prolonged or repeated exposure, causes skin irritation and may cause respiratory irritation (Karim et al., 2010). It can cause chest pain, shortness of breath; an anxiety, tremors, hypertension, and even skin coloration if the dose is high (Kushwaha et al., 2014). Despite this, the toxicological data on the use of MB in humans for many years have not yet indicated a specific hazard in the use of this product, (Yi and Zhang, 2008), and

although MB is not classed as highly dangerous, it clearly has a harmful effect on living organisms (Gobi et al., 2011) and waters (Rafatullaha et al., 2010). Many synthetic dyes and pigments generate harmful by-products, particularly when dumped directly into the environment without any specific treatment of toxic constituents (Saidi, 2013).

This work is aimed to promote local georesources (silica sands) for the preparation of silica gel, and to investigate its decontamination capacity for the treatment of synthetic dyes in solution.

2. Experimental details

2.1. Geological setting

In this study, three representative samples were collected from different sites:

- Jebel Menchar (M): Fortuna formation (Burdigalian), Tunisian Dorsal structural domain,
- Jebel SidiAïch (S) and Jebel Attaf (A): Sidi Aich formation (Barremian), meridional Atlas.

The deposits of the Fortuna formation are located about 2 km north east of the village of El Hammem Jedidi and 15 km south of the city of Hammamet. It is an ENE/WSW anticlinal structure with an Eocene level extending over 3.5 km long and 1.5 km wide. These quartz sands appear in the channels interspersed with clayey materials, silt and rarely consolidated sandstone (Mejri, 2012). The Sidi Aich formation (Central Tunisia) was deposited in a regressive context in a field of passive margin. Of volcanic origin, the pre-existing mother rock was eroded and transported from the area of Dahar, south of the Saharian platform (M'Rabet, 1981; Gallala et al., 2009). Central Tunisia is characterised by E–W and NW–SE oriented faults, and ranges of NE–SW trending anticlines, separated by large synclinal basins.

In this Central and north Tunisia, previous studies have been carried out in sedimentology (Buroillet, 1956; Buroillet et al., 1983; Ben Ferjani et al., 1990; Chekhma et al., 1990; Chaabani et al., 1992; Ben Youssef, 1999; Chaabani and Razgallah, 2006; Gallala, 2010), tectonic studies (Bouaziz et al., 2002; Mejri, 2012; Said, 2011; Gharbi, 2013), paleogeography (Soussi, 2000; Abbès, 2004) and hydrocarbons prospection (Negra et al., 2009).

2.2. Methods

Mineralogical and major-minor chemical compositions were determined for whole-rock samples. The mineralogy was determined in raw and gel materials using a Panalytical X'Pert PRO X-ray diffractometer (CuK α , $\lambda = 0.154056$ nm). The collected data (10–80° 2 θ range, scan rate of 0.02° (2 θ)) were processed by Panalytical X'Pert Highscore software. The existing phases were identified, and their relative amounts were estimated by measuring and comparing the peak areas of the principal reflexion (Ali et al., 2016). The chemical composition of these quartz sands was estimated, on powdered samples, by X-ray fluorescence using a Panalytical Axios Dispersive X-ray Fluorescence Spectrometer. The results are expressed as percent concentrations of oxides of each element. The obtained gels were characterised by scanning electronic microscopy (SEM - Hitachi, SU 70) to observe the microstructural details. The specific surface area and porosity parameters were determined by B.E.T method based on the adsorption and desorption of nitrogen at 77 K. The sample with grain size <150 μ m is degassed before hand at 70 °C under high vacuum for 72 h and then the cell was plunged in a liquid nitrogen balloon (Brunauer et al., 1938). The grain-size distribution was obtained via dry sieving in an AFNOR

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