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Cosmetic and pharmaceutical qualifications of Egyptian bentonite and its suitability as drug carrier for Praziquantel drug

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ABSTRACT

The aim of this paper is to characterize and evaluate newly discovered bentonite deposits in Egypt for pharmaceutical and cosmetic applications as well as its suitability as carrier for Praziquantel drug. The study was performed for the raw bentonite sample, purified bentonite sample and alkali activated purified bentonite sample. The raw bentonite sample composed mainly of montmorillonite contaminated by little amounts of quartz and calcite, while the purified sample composed of montmorillonite without detected mineral impurities and matches the mineralogical properties of Wyoming bentonite as an international standard. Geochemically, the studied raw and purified samples appear to high purity with a chemical composition close to those of Wyoming bentonite and match the pharmacopeia specifications. The chemical properties in addition to the textural properties of the surface area, porosity, particle size distribution qualify the bentonite products to use as a function in powder, emulsion and creams. Investigation of pharmacopeia properties of pH, sedimentation volume and swelling capacity revealed the suitability of the raw and purified samples for pharmaceutical and cosmetic applications. Moreover, the microbiological tests indicated that the samples free from harmful microbial pathogens. At the optimum conditions of time (240 min), bentonite dose (250 mg) and reaction temperature (60 °C), the obtained encapsulation percentages of Praziquantel drug are 62%, 78.4% and 93.2% for raw bentonite, purified and alkali activated bentonite, respectively. The releasing percentage of the drug using an intestinal buffer at pH 7.4 is more efficient and the maximum obtained values were obtained after 420 min. The obtained releasing values are 71%, 79.2% and 87.4% for raw bentonite, purified bentonite and alkali activated bentonite, respectively

1. Introduction

The world demand of natural clays for cosmetic and pharmaceutical applications increased greatly in the later periods which associated with the continuous success of natural therapies (Abdel-Motelib et al., 2011). This prompted several researchers to search for and assess new clay mineral resources. Clay minerals are a class of layered silicate minerals that framed as secondary minerals during change or chemical weathering of silicate minerals (Zhang et al., 2010). The clay minerals were characterized into seven groups: (a) kaolinite-serpentinite, (b) pyrophyllite-talc, (c) smectite, (d) vermiculite, (e) mica, (f) chlorite, (g) interstratified clay minerals (Martin and Griswold, 2009). Clay minerals exhibit amazing physicochemical properties of the high surface area, high cation exchange capacity, chemically inert materials, non-toxic and non-irritant properties at the accepted limits for cosmetic and

pharmaceutical applications (Gamoudi and Srasra, 2017).

Natural clay was used widely in the production of therapies for arthritis, rheumatism, seborrhea, acne and traumatic damages of bone muscles (Veniale et al., 2007; Abdel-Motelib et al., 2011). It was reported that the cosmetic applications of clays restricted to external using as a skin treatment and facial (Pusch, 2014; Ngomo et al., 2014). The medical applications of natural clays related to their roles as active principals and/or excipients (Chen et al., 2010). The common applications of natural clays as active principals includes dermatological protections, antidiarrheal gastrointestinal, anti-inflammatory and antacid (Gamoudi and Srasra, 2017). As active excipients, natural clays can be used as disintegrants, anticaking agents, desiccants, lubricants, and as emulsifying materials (Park et al., 2008; Gamoudi and Srasra, 2017). Additionally clay minerals were used widely as drug carriers and vehicles for controlled release of therapies (Patel et al., 2007).

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Among the used clays for environmental, industrial and medical applications, bentonite attracted the attention for its availability and low prices in addition to its unique properties of high surface rea, very high cation exchange capacity, small particle size, high adsorption capacity and non-toxic properties (Abukhadra et al., 2015). Bentonite clays are geological term refers to mud rock composed mainly of smectite clay minerals admixed with other mineral phases as impurities (Shaban et al., 2017a). Pharmaceutically bentonite was defined as natural, crystalline and hydrous aluminum silicate minerals that present in nature as fine powder of pale buff, creamy or gravish color and free from grit (Raymond et al., 2003). Natural bentonite was described by US Pharmacopeia (2007) as colloidal and native hydrated aluminum silicate materials and commercial purified natural bentonite term refers to beneficiated and colloidal bentonite that purified from the associated impurities (Abdel-Motelib et al., 2011). The applicability of natural bentonite sample for pharmaceutical and cosmetic applications controlled mainly by its cation exchange capacity, specific surface area, mineralogical composition, swelling index, colloid properties and its absorption and adsorption capacity (Gamoudi and Srasra, 2017).

However the total estimated geological reserve of bentonite deposits in Egypt is about 63 million tons of bentonite (Kandeel, 1989), most of the performed studies focused only on bentonite deposits of Western desert and little on low quality bentonite deposits in Sinai and El Fayoum area. The only pharmaceutical study for Egyptian bentonite was introduced by Abdel-Motelib et al. (2011) for the bentonite deposits in the Western Desert. Up to our knowledge there are no studies or recording for bentonite deposits in Quseir area, Central Eastern Desert, Egypt. Thus the aim of this work is to characterize newly discovered bentonite deposits in Central eastern Desert of Egypt as raw materials and to evaluate the suitability of such deposits for cosmetic and pharmaceutical applications as well as their suitability as a drug carrier for Praziquantel drug. Effect of alkali activation on enhancing the pharmaceutical qualifications of Central Eastern Desert bentonite also was addressed in this paper.

2. Material and Methods

2.1. Sampling and Geological Setting

Several technological samples were collected from Zug El Bohar locality, Quseir area, Eastern Desert, Egypt representative to the total reserve of bentonite deposits in this area. The sedimentary package exposed at the Quseir area (Fig. 1A) is represented by siliciclastics (clays and sandstones), phosphorites and carbonates. Such sediments were accumulated under different environmental settings. Three main environmental regimes can be recognized. These sediments start with fluvial – dominated style, followed by tide – dominated one and finally open marine – dominated regime. The studied succession start with Nubia Sandstone followed upward by Quseir Shale, Duwi Formation, Dakhla Shale, Esna Shale and finally capped with Thebes formation (Said, 1962). Bentonite – bearing sediments of the Quseir area (Fig. 1B) are built up of varicolored siltstones and claystones enclosing thin bands of marl (~30 cm). This succession represents a part of the Quseir Shale which passes vertically into the overlying Duwi Formation.

2.2. Purification and Activation of Bentonite

The physical purification of bentonite was performed according to Hassan and Abdel-Khalek (1998). Raw bentonite was crushed by primary jaw crusher to 5 ml and treated using attrition scrubber at impeller speed 2500 rpm and solid concentration 50% for 1 h. The treated product was transferred into Mozley-2-hydrocyclone with adjusting the parameters at 10% solid and 30 psi operating pressure. The underflow fractions which are enriched in quartz and calcite (> 44 µm) were removed and the overflow fractions (< 44 µm) were collected and dried at 50 °C as pre-concentrate for the further purification processes.

The resulted bentonite pre-concentrate was washed with distilled water several times followed by acid leaching by diluted HCl acid (0.1 M) at room temperature for 2 h to remove iron and carbonate impurities. This was followed by washing with hydrogen peroxide to remove the associated organic matters. The obtained product was purified utilizing sedimentation technique as described by Patel et al. (2007). 150 g of bentonite was dispersed in 10 l of distilled water and the purified bentonite was collected after 10 h as the supernatant disperse particles with size < 2 μ m. Then the produced purified bentonite was ground for 10 h to reduce the whole sample to $-20 \,\mu$ m using agate ball mill free from contaminations.

Alkali activation of bentonite was carried out through dispersion of 300 g of purified bentonite in 100 ml of 0.1 M NaCl solution under stirring for 12 h at room temperature for three runs. The clay fractions were then separated from the slurry using a centrifuge for 15 min at 4000 RPM and washed several times with distilled water to be free from chloride ions.

2.3. Characterization

2.3.1. Mineralogical and Geochemical Characterization

X-ray powder diffraction (XRD) patterns of bentonite and the modified products were measured using a Philips APD-3720 diffractometer with Cu K α radiation operated at 20 mA and 40 kV. The patterns were measured in the 2 θ range of 5–70 at a scanning speed of 5°/min. This was done for raw bentonite sample, purified sample and alkali activated sample. The chemical composition of bentonite was obtained through XRF analysis, using utilizing Panalytical Axios Advanced XRF technique at The Central Metallurgical Research and Development, Egypt. Morphologic properties of raw bentonite samples were studied using scanning electron microscopy (JSM-6510, JEOL, and Tokyo, Japan). Transmission Electron Microscope images were taken by JEOL-JEM2100 (Japan) with an acceleration voltage of 200 kV. The Fourier Transform Infrared spectrometer (FTIR - 8400 S Shimadzu, Japan) was used to determine the chemical structural groups.

2.3.2. Physicochemical Properties

2.3.2.1. Surface Area and Particle Size Distribution. The specific surface area was determined by using Brunauer–Emmett–Teller (BET) method. The pore-size distribution was measured from the adsorption branches of the isotherms using the Barrett–Joyner–Halenda (BJH) method. The particle size distribution of ground bentonite sample was measured using BT-2001(Wet) Laser particle size analyzer device, at The Central Metallurgical Research and Development, Egypt.

2.3.2.2. Cation Exchange Capacity (CEC). Cation exchange capacity of newly discovered bentonite was investigated using $BaCl_2$ method (Abdel-Motelib et al., 2011). The raw bentonite sample was washed with deionized water to achieve efficient removal of extraneous cations and 1 g from the washed bentonite dried and dispersed within hydrated barium chloride. Then the solid was separated using filter paper and washed by 300 ml of 1N ammonium acetate solution (pH 7) to liberate the exchangeable barium ions. The final concentration of barium was estimated using inductively coupled plasma mass spectrometry (ICP-MS) (Perkin Elmer).

2.3.2.3. Thermal Stability. Thermal stability of the raw bentonite sample was investigated through thermogravimetric (TGA) analysis and different thermal analysis (DTA) utilizing TGA/DTA Setsys Evolution (Model; 1740). The operating temperature was adjusted to be from ambient to 1000 °C and the heating rate increased by 10 °C per minute.

2.3.3. Pharmaceutical Properties

2.3.3.1. Swelling Capacity. Swelling capacity of raw bentonite was

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