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GRANITE DUST DERIVED SILICA EMBEDDED MOLYBDENUM DISULFIDE-POLYETHERSULFONE MIXED MATRIX MEMBRANES FOR OIL-WATER SEPARATION

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ABSTRACT

Granite dust contains very fine particles that can cause respiratory health issues and pollute air and water sources. The amount of granite dust (GD) waste generated is in millions of tonnes annually. This amount is increasing rapidly, leading to numerous disposals, governance, and environmental issues. Therefore, finding a feasible application of GD-derived materials could significantly resolve the waste problem. In this work, a simple, low-cost sol-gel method was used to synthesise silica nanoparticles from GD. In order to improve the sol-gel process efficiency, an acid leaching pre-treatment process was added prior to the extraction of silica in the form of sodium silicate and the formation of silica gel through neutralisation processes. Various types of acids such as citric acid, acetic acid and hydrochloric acid were used to remove metallic impurities in the GD. Besides acid types, the acid concentrations (0.5 M, 1 M, 2 M, and 3 M) and reaction temperatures (25 °C, 60 °C, 80 °C, and 90 °C) were also varied to investigate their role during the acid leaching pre-treatment. The results revealed that the use of acid leaching pre-treatment can effectively eliminate most contaminants such as Al_2O_3, K_2O, Fe_2O_3 and Ag_2O in the GD. By using 1 M citric acid at 80°C, the purity of GD was significantly improved, and it was used as a precursor to synthesis silica. It was observed that if the acid concentration was less than 1 M, the impurities could not be completely removed as the acid strength was insufficient to break the metal oxide bonds in the GD. In addition, if the pre-treatment temperature was too high, the acid might be vaporise. The XRD pattern of the silica confirms the amorphous nature with BET specific surface area and particle size of 90.39 m^2/g and 228.4 nm, respectively. The silica derived from GD was then hydrothermally synthesised with molybdenum disulfide (MoS_2) and used as a filler in the fabrication of polyether sulfone mixed matrix membranes (PES MMMs) for oil-water separation. The amount of MoS_2-silica added used was varied from 0.25 wt% to 2.0 wt%, and their effects on mechanical, chemical, and surface morphology of the PES MMMs were evaluated. The addition of silica into MoS_2 leads to significant enhancement in porosity, hydrophilicity and water flux as compared to bare PES and MoS_2-PES membranes. The performances of MoS_2-silica PES membranes were then investigated for oil-water separation using different oil concentrations (500 ppm, 1000 ppm, and 1500 ppm) at 1 bar and room temperature. The membrane was first compacted for 2 hours at 2 bar before the oil-water separation test to maintain a stable flux. It was found that the oil-water flux of the composite membranes increased significantly from 5.85 LMH to 75.79 LMH when silica was added. This was due to the enhancement in membrane properties and morphology structure. The highest permeation flux of 75.79 LMH was obtained using 2M-S-PES. Generally, the oil rejection of the MoS_2-silica PES MMMS fabricated in this work was maintained above 95%. This study indicates that the environmentally acid leaching pre-treatment used in this work can improve the quality and yield of silica produced. It also confirms the potential of using GD-derived silica as one of filler for MMMs fabrication.

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CHAPTER ONE
INTRODUCTION

1.1 Research Background

Mining and quarrying industries are among the most important contributors to the economic development in Malaysia particularly for the construction industry [1, 2]. Figure 1.1 shows the flow process of granite quarrying processing. Granite will first be exploited from a stone mine and cut into primary products such as granite slabs and man-made slabs. These products will then be processed into floor or other products. Scrap materials are usually processed into fine powder such as calcium carbonate and nano calcium carbonate in order to meet the market demand. During the process, a huge amount of stone dust and waste water will be produced [3]. It is estimated that over 30% of the total volume of stone waste and granite dust (GD) were produced during the quarrying process. The vast amount of this GD waste has leads to severe problems from the handling to the disposal stage, which can be very harmful to human health and damage to the environment [4]. The disposal of this by-products also often involves high cost of transportation [5]. To date, countries such as Lebanon [6], Europe [7], etc, have a massive stone waste problem [7, 8]. Thus, sustainable recycling and reusing of GD is a great importance, which not only solve the problem of huge quantities of GD to be disposed, but also lead to a cleaner environment that society desires. Up to 15% of GD can be used as a sand replacement [9]. GD was also used by some researchers to prepare a hot asphalt mixture, ceramics, and bricks [10]. It also has a various application in art industry where artists used this dust in the production of stoneware clay bodies in ceramic artwork or as a filler/hardener in pastels drawings [11].