Separation and Purification Technology 204 (2018) 108–115

Contents lists available at ScienceDirect

Separation and Purification Technology

journal homepage: www.elsevier.com/locate/seppur

Electrodialysis for fluoride and nitrate removal from synthesized photovoltaic industry wastewater

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ARTICLE INFO

Keywords:
PV factory
Wastewater
Fluoride
Nitrate
Electrodialysis

ABSTRACT

Fluoride and nitrate ions are the main pollutants in the photovoltaic cells manufacturing effluent. Their presence in the effluent is due to the extensive use of HF and HNO_3 acids during the silicon wafers production process. This work deals with the electrodialysis application to remove these ions. Synthetic solutions were used for the investigation of the main operational factors affecting the treatment performance; such as current intensity, initial pollutants concentration and pH. Significant fluoride removal was obtained in an acidic or neutral medium after a relatively short treatment time of 6 min. Nitrate ions were also removed showing an efficiency of 98% after 18 min of treatment using a current intensity of 0.1 A and for an initial concentration of 1000 ppm. The study of competition between these two ions during their eliminations showed that the fluoride ions presence does not affect the nitrate removal, while; the nitrate ions presence in the solution delays the fluoride removal from 7 to 20 min, and makes the removal less effective. The results obtained can be used to optimize the pollutant removal recovery at large scale.

1. Introduction

The present regime of industrial activities still depend largely fossil fuels, until new energy technologies are developed with comparable cost and energy production. One of these technologies could be the photovoltaic (PV) energy conversion systems. Over the last decade, the PV energy conversion has become an emerging technology, while its demand is increasing rapidly, and is being considered as an alternative technology which may contribute to the world energy supply [1].

Photovoltaic is the direct conversion of sunlight into electricity; the process generates environmental-friendly renewable electricity; to appropriate market conditions, research and development –R&D- is crucial to the further PV technology development. However, to ensure that PV energy can indeed achieve this expectation, a careful consideration of PV energy conversion and environmental potential risks is necessary [2]. In this industry, large water, ultrapure water (UPW) volumes and considerable chemicals amounts are required [3,4]. The wastewaters from the production process must be treated in such way to recycle as much water as possible. The treated wastewaters must comply with the norms in order to protect the environment and the public health. As the water is becoming increasingly valuable as a raw material, an efficient water management of this resource is necessary. Notably, in this industry, the water resistivity levels required were reported (> 17.5 Mohm) and lower sensitivities to dissolved organics were indicated (< 20 ppb). The Etch and Diffusion steps use approximately 50% of the UPW during the manufacturing process. Chemical usage in the “etch step” is limited to a variety of acids (HF/HNO_3) which are fairly easy to treat making it an excellent opportunity for reclamation.

Several researchers have suggested different treatment such as adsorption, electrocoagulation, flotation, advanced oxidation processes (AOPs), and the membranes techniques which include the ultrafiltration, the reverse osmosis, electrodialysis, and nanofiltration for the treatment of both pollutants [2,5–16]. The Thereby using membrane processes which is increasingly popular for wastewater reuse applications, and membrane technology could be considered as a reliable option for wastewater reclamation. Indeed, the membrane technology advantage over conventional separation methods are high removal capacity, operation flexibility and cost effectiveness [16]. From the different membrane processes available for the ions separation from solutions, only two, reverse osmosis and electrodialysis, have got the practical application stage for the inorganic contaminants removal from drinking water and wastewater [17–23]. The most present pollutants in these effluents are the Fluoride and the Nitrate ions. Indeed, wastewaters from PV industries have high fluoride concentrations, typically

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https://doi.org/10.1016/j.seppur.2018.04.068
Received 14 October 2017; Received in revised form 24 April 2018; Accepted 24 April 2018
Available online 25 April 2018
1383-5866/

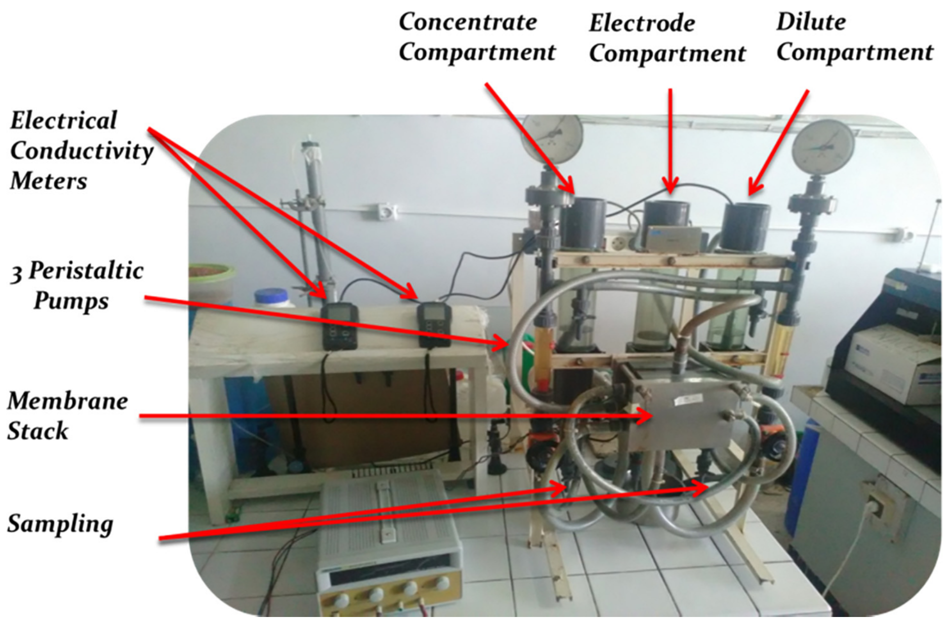

Fig. 1. A Corning P1 electrodialysis pilot system.

in a range of 500–2000 mg/L and a nitrate ions important amount [6,7]. The direct discharge of such wastewaters indeed represents a massive hazard for the environment.

Prolonged exposure to high fluoride amount can result in several neurological damages. In severe cases, teeth mottling, bones softening, tendons and ligaments ossification occurs [8,23]. Due to its high toxicity, industrial wastewater containing fluoride is rigorously controlled. The current discharge standards for wastewater containing fluoride vary according to different countries; Algeria and Taiwan recommend 15 mg L^{-1}. The second pollutant in our effluents is the nitrate ion which also has harmful effect on the human health (Methaemoglobinaemia and other diseases) [18], and the ecosystem (eutrophication phenomena); besides infant mortality there are several reports indicating that the use of drinking water with NO_3^- high level could result in certain cancers including stomach cancer, central nervous system, birth defects and hypertension [18,20]. In Algeria, the nitrate concentration limit in drinking water is fixed at 50 mg L^{-1}; in some countries in Europe the same limit is retained; the World Health Organization has set the maximum limit to 10 mg L^{-1} in drinking water. One treatment step in the PV industry is the formation of calcium fluoride (CaF_2) by adding excess lime, which is insufficient to meet environmental standards. In the evaluation, the precipitation can only decrease the fluoride concentration to 20–100 mg/L [8]; definitely, additional decontamination process must be accomplished.

In this work, investigations were made on the fluoride and nitrate ions elimination by electrodialysis process and on these two ions competition in their elimination from the synthesized PV industry wastewater. The present work also aims to treat the water suitably, in order to recycle the water and use it, thereby attain the zero discharge concept is targeted, since the UPW is very expensive and its consumption in this industry is very important.

2. Materials and methods

2.1. Materials

Electrodialysis in which the ionic components removal from aqueous solution through ion exchange membranes is carried out by an electrical field driving force was selected in this work. Investigations were made on the fluoride and nitrate ions elimination by electro-dialysis process and on competition of these two ions in their eliminations from the simulated PV industry wastewater. Synthetic solutions simulating the PV cells rinsing wastewaters were used for the study of the main operational factors affecting the electrodialysis performance; such as current intensity, initial pollutants concentration, and pH.

A Corning P1 electrodialysis pilot equipment was employed for experimental studies (Fig. 1). The P1 stack contained 10 unit cells consisting of 19 Anionic Exchange Membrane (AEM) and 20 Cationic Exchange Membrane (CEM). The effective area of each membrane is 69 cm^2. These membranes were manufactured by Asahi Glass (Chiyoda-ku, Tokyo). Three peristaltic pumps Seibec 38600 models with a 50 L/h flow capacity were used to move out continuously the solutions circulation. Three solution tanks were used for holding the diluted, the concentrated and the electrode rinse solutions. The electrodialysis tests were performed at ambient temperature. When a direct current potential was applied, the pollutant (fluoride or nitrate ions) migrates towards the anode. The ions leave the dilute compartment and move through the anion exchange membrane and were retained by the cation exchange membrane in the concentrate compartment.

2.2. Methods

The batch tests were carried out using synthetic solutions prepared by dissolving NaF and/or $NaNO_3$ in deionized water at different concentrations and at ambient temperature (that means that none heating system was used, and according to the experiment day's temperature, a fluctuation of ± 2 °C might have occurred and it was estimated that doesn't make a significant change to the obtained results). In the case of fluoride removal, NaF solutions at various concentrations ranging from 120 to 180 ppm were introduced in the dilute compartment. Besides, a NaF solution at a lower concentration was introduced into the concentrate compartment. In the nitrate removal case, $NaNO_3$ solutions at different concentrations varying from 750 to 2000 ppm were introduced in the dilute compartment. Moreover, a $NaNO_3$ solution at a lower concentration was introduced in the concentrate compartment. For all experiments, a Na_2SO_4 solution was introduced into the electrode compartment. The solution concentrations details have been presented in the following Table 1:

In addition to the initial pollutant concentrations, the investigations of others operational factors affecting the ions removal process performance; such as current intensity, and pH were also done. After selecting the optimum experimental parameters for each pollutant