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**Fenton coupled with nanofiltration for elimination of tartrazine**I. Escalona<sup>\*1</sup>, F. Stüber<sup>1</sup>, A. Fortuny<sup>2</sup>, C. Bengoa<sup>1</sup>, A. Fabregat<sup>1</sup>, J. Font<sup>1</sup><sup>1</sup>Universitat Rovira i Virgili, Spain, <sup>2</sup>Universitat Politècnica de Catalunya, Spain

Dyes is one of the most important group of water pollutants occurring in the wastewaters from textile, leather, paper-making, plastics, food, rubber, pharmaceuticals and cosmetics industries [1]. Approximately 10,000 different types of dyes are produced worldwide annually and it is estimated that about 10% of dyes used are lost in waste streams during the processing operations [2]. Discharge of such effluents into the ecosystem is a source of aesthetic pollution, eutrophication and perturbations to aquatic life. Dyes exhibit considerable structural diversity and the two classes of dyes mostly used are azo and anthraquinone dyes. Azo dyes are considered an objectionable type of pollutant because they are toxic generally due to oral ingestion and inhalation, skin and eye irritation, skin sensitization and also due to potential carcinogenicity [3]. The treatment of dye effluents is difficult and ineffective with conventional biological processes because many dyes are very stable to light, temperature and are non-biodegradable. Colour removal, especially from textile wastewaters, has been a big challenge over the last decades, and up to now there is no single and economically attractive treatment that can effectively decolourise dye polluted water. Therefore a number of techniques aimed at preferential removing dyes from wastewater have been developed, including adsorption, photo degradation, chemical oxidation, membrane processes, coagulation, flocculation, etc [4].

Fenton's reagent is a suitable chemical for treating wastewater resistant to biological treatment or poisonous to live biomass. Fenton's reagent is particularly attractive because of the low costs, the lack of toxicity of the reagents (i.e.,  $\text{Fe}^{2+}$  and  $\text{H}_2\text{O}_2$ ), the absence of mass transfer limitation due to its homogeneous catalytic nature and the simplicity of the technology required. However, since Fenton process requires ferrous salt for the oxidation reaction to take place, the iron sludge formed after the reaction has to be removed before discharging.

Commonly applied treatment methods for colour removal from dye contaminated effluents consist of integrated processes involving various combinations of methods [5]. The application of nanofiltration (NF) is a promising membrane technology that can be seen as an alternative method for removing low molecular weight organic micropollutants. There have been numerous attempts to enhance oxidation with additional process steps. In this case, membrane separation is becoming a more attractive alternative because of the purely physical nature of its separation principle as well as the modular design of membrane processes. Separation without phase change, low energy consumption and operability at ambient temperature have given an edge to membrane processes over other conventional processes. Therefore, NF has a potential applicability in Fenton process as catalyst remover, in order to reduce the iron concentration in wastewater before discharge.

Due to the extensive use and particular refractoriness of azo dyes, Tartrazine has been selected as main model pollutant in this research. Tartrazine (E 102, FD & C Yellow N°5) is an azo dye used as a foodstuff additive and in various human drugs. The enduring nature of tartrazine, together with the difficult decontamination problems associated with dye wastewaters; make it a suitable model target for exploring new approaches to water purification. Taking this into consideration, the main objective of this study was to investigate the degradation of Tartrazine by means of Fenton's process under different operational conditions

and, additionally, to combine these processes with retention of low concentrations of Tartrazine, intermediates from oxidation, and iron sludge in a Membrane Reactor. In this sense, the investigation aimed at evaluating the membrane removal efficiency of tartrazine, intermediates product, and iron sludge and investigating the influence of trans-membrane pressure and membrane pore size.

The Fenton reaction was done in a water-jacketed lab scale glass reactor. The  $\text{Fe}^{2+}/\text{H}_2\text{O}_2$  and  $\text{H}_2\text{O}_2/\text{Tartrazine}$  molar ratio was tested in the range from 0.00 to 0.024 and from 8 to 63, respectively. The effects of different system variables namely initial concentration of  $\text{Fe}^{2+}$ , initial concentration of  $\text{H}_2\text{O}_2$ , initial concentration and reaction time were studied. Following the tartrazine degradation and selection the optimal oxidation variables, the final solutions were filtrated in a nanofiltration system. This equipment was a home-made cross flow laboratory pilot plant. The coupling of Fenton process and NF proved to be viable to the decolourisation and consequent degradation of the Tartrazine, obtaining effluents free of sulphanilic acid. The removal efficiencies for colour, TOC and COD were improved by increasing the  $\text{Fe}^{2+}$  and  $\text{H}_2\text{O}_2$  concentrations.  $\text{H}_2\text{O}_2/\text{Tartrazine}$  molarconcentration ratio of 16 appeared as optimum dosage for the Fenton step. The decolourisation of dye increased by increasing the  $\text{Fe}^{2+}$  dosage from 0 to 6 mg/L. Increase in dye concentration decreased the removal rate. The polymeric NF membranes used showed superior retention of iron species. Only a slight difference on the iron retention was observed for the five commercial membranes tested. Thus, NF demonstrated to be adequate for iron recovery. The ESNA membrane achieved the highest colour and sulphanilic acid retention, above 97%; which made it a more attractive alternative.

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