

Recycling the Lanthanides from Permanent Magnets by Electrochemistry in Ionic Liquid

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Abstract : Thanks to their high magnetization and low mass, permanent magnets (NdFeB and SmCo) have quickly become essential for new energies (wind turbines, electrical vehicles...). They contain large quantities of neodymium, samarium and dysprosium, that have been recently classified as critical elements and that therefore need to be recycled. Electrochemical processes including electrodisolution followed by electrodeposition are an elegant and environmentally friendly solution for the recycling of such lanthanides contained in permanent magnets. However, electrochemistry of the lanthanides is a real challenge as their standard potentials are highly negative (around -2.5V vs ENH). Consequently, non-aqueous solvents are required. Ionic liquids (IL) are novel electrolytes exhibiting physico-chemical properties that fulfill many requirements of the sustainable chemistry principles, such as extremely low volatility and non-flammability. Furthermore, their chemical and electrochemical properties (solvation of metallic ions, large electrochemical windows, etc.) render them very attractive media to implement alternative and sustainable processes in view of integrated processes. All experiments that will be presented were carried out using butyl-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide. Linear sweep, cyclic voltammetry and potentiostatic electrochemical techniques were used. The reliability of electrochemical experiments, performed without glove box, for the classic three electrodes cell used in this study has been assessed. Deposits were obtained by chronoamperometry and were characterized by scanning electron microscopy and energy-dispersive X-ray spectroscopy. The IL cathodic behavior under different constraints (argon, nitrogen, oxygen atmosphere or water content) and using several electrode materials (Pt, Au, GC) shows that with argon gas flow and gold as a working electrode, the cathodic potential can reach the maximum value of -3V vs Fc+/Fc; thus allowing a possible reduction of lanthanides. On a gold working electrode, the reduction potential of samarium and neodymium was found to be -1.8V vs Fc+/Fc while that of dysprosium was -2.1V vs Fc+/Fc. The individual deposits obtained were found to be porous and presented some significant amounts of C, N, F, S and O atoms. Selective deposition of neodymium in presence of dysprosium was also studied and will be discussed. Next, metallic Sm, Nd and Dy electrodes were used in replacement of Au, which induced changes in the reduction potential values and the deposit structures of lanthanides. The individual corrosion potentials were also measured in order to determine the parameters influencing the electrodisolution of these metals. Finally, a full recycling process was investigated. Electrodisolution of a real permanent magnet sample was monitored kinetically. Then, the sequential electrodeposition of all lanthanides contained in the IL was investigated. Yields, quality of the deposits and consumption of chemicals will be discussed in depth, in view of the industrial feasibility of this process for real permanent magnets recycling.

Keywords : electrodeposition, electrodisolution, ionic liquids, lanthanides, recycling

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