

Effect of substrate pre-coating on adhesion of sintered nickel plaques for electrode application in rechargeable batteries

Valery I. Chani^{a,b}, Quan Yang^a, David S. Wilkinson^{b,*}, George C. Weatherly^{b,1}

^a INCO Ltd., 2060 Flavelle Boulevard, Mississauga, Ont., Canada L5K 1Z9

^b McMaster University, 1280 Main Street West, Hamilton, Ont., Canada L8S 4L7

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Abstract

The battery industry sees a need to improve the adhesion strength between porous nickel plaques and substrates in sintered nickel electrodes in order to reduce the use of nickel and to achieve higher battery capacity. In this paper we report on the use of a thin buffer layer to enhance plaque/substrate adhesion. In particular, we have attempted to improve adhesion performance of conventional sintered plaques produced from filamentary Ni powders (INCO-255, particle size 2.2–2.8 μm) through the introduction of a thin, intermediate layer made of finer Ni powders (INCO Type 110 and INCO Type 210, particle size 0.8–1.5 μm and 0.5–1.0 μm , respectively). The process consists of two steps. First, a buffer layer was applied from a slurry containing the finer Ni powders. After drying, the substrate was coated again with a slurry containing standard Type 255 powder. A vertical coating apparatus with periodically contacting blades was designed to produce thin, uniform buffer layers with low surface density (1–10 mg cm^{-2}). For adhesion testing, a tensile strength test specimen holder was modified to ensure planar loading of the interface. Significantly enhanced bonding between the plaque and substrate was observed due to the introduction of the buffer layer. © 2004 Elsevier B.V. All rights reserved.

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1. Introduction

Porous nickel structures produced by sintering INCO Type 255 filamentary nickel powder on nickel-plated perforated steel sheets are called nickel plaques and are widely used in the fabrication of electrodes for nickel-based rechargeable batteries [1–4]. Although pasted or foam electrode technology has been employed more recently by many nickel based battery manufacturers due to their shorter production cycle and lower capital requirement, sintered nickel electrodes are still the choice of electrode technology for battery applications demanding high power and reliability, along with long cycle and calendar life, such as batteries for aerospace and railway applications, power tools, and some portable elec-

tronics. In addition to its higher cost, the high nickel content and the difficulty in completely filling the fine small pores with active material, sintered nickel electrodes usually exhibit lower specific battery capacity than foamed or pasted ones. The battery industry is therefore seeking to reduce the cost of sinter electrodes while retaining their positive merits. One possible approach is increasing the porosity of the sintered nickel plaque into which $\text{Ni}(\text{OH})_2$ is chemically impregnated to form the final electrode. In a recent paper we have reported an improved method for processing sintered plaques that leads to increased porosity at a given strength level [11]. However, an additional issue is the weakened bonding of the porous nickel structure to the perforated substrate, which could potentially cause problems in the electrode fabrication process and also jeopardize electrode service life. The objective of the present work is to explore methods of enhancing plaque adhesive strength as a means to manufacture high porosity sintered nickel plaques.

* Corresponding author. Tel.: +1 905 525 9140; fax: +1 905 528 9295.
E-mail address: wilkinso@mcmaster.ca (D.S. Wilkinson).

¹ Deceased.

The mechanical strength of the nickel plaque and its bonding strength to the substrate are both critical to the reliability of batteries in service as well as to the reduction of rejection rate during the electrode manufacturing process. First, the Ni plaque has to survive severe chemical, thermal, and mechanical cycles during the repeated chemical impregnation process for filling of its pores with active mass. Any breakage of the plaque or de-bonding from the substrate at this stage leads to failure and rejection of the electrode. Battery electrodes are then subjected to a demanding chemical environment (e.g. KOH), cyclic distortion associated with charge/discharge processes and thermal fluctuations. Currently the porous plaques are pasted or coated onto nickel-plated perforated steel sheets in order to enhance the lateral strength. Thus the overall mechanical performance of the electrodes depends mostly on adhesion between the plaque and the substrate.

A conventional slurry-based process is widely used for the fabrication of tape-like porous nickel compacts (as is well described elsewhere [1,5–11]). Briefly, a slurry is prepared by mixing filamentary Ni powder in an aqueous solution containing a binder such as hydroxypropyl methyl-cellulose (HPMC) (Fig. 1). The substrate is then coated on both sides with the slurry and dried producing a green part. Finally the green part is fired at 700 to 1050 °C for up to one hour in a reducing atmosphere to produce the final sintered plaque ready for chemical impregnation and electrode fabrication.

Generally the as-sintered plaques have a volume about one third that of the starting slurry as illustrated schematically in Fig. 1. Such a large compaction during drying and sintering builds in mechanical stresses thought to be tri-axial in nature. The structural gradients and stresses perpendicular to the surface of the substrate are negligible because it is a thin (<1 mm) layer. However the stresses acting in the plane of the substrate can result in bending, mechanical degradation and cracking of the coatings even during drying and sintering, as well as in service.

Many technological parameters need to be under precise control to achieve reasonable reproducibility of the structural, mechanical and electrical properties of the plaques. Therefore, it is evident that the formation of a high quality interface between the porous nickel and the substrate, resulting in sufficient adhesion, is a rather complicated task. Moreover, the stochastic nature of the contacts within the plaque and between plaque and substrate make this difficult to reproduce consistently.

In spite of the worldwide production and application of rechargeable batteries there are very few reports [1–11] offering details of the fabrication process or characterization of the adhesion properties of these materials. This is due in large part to the proprietary nature of processing research and the highly competitive market for batteries. This project therefore attempts to contribute to the development of the coating technology by offering a systematic approach to the measurement of plaque-substrate adhesion and its application in developing strongly adhered coatings. In particular

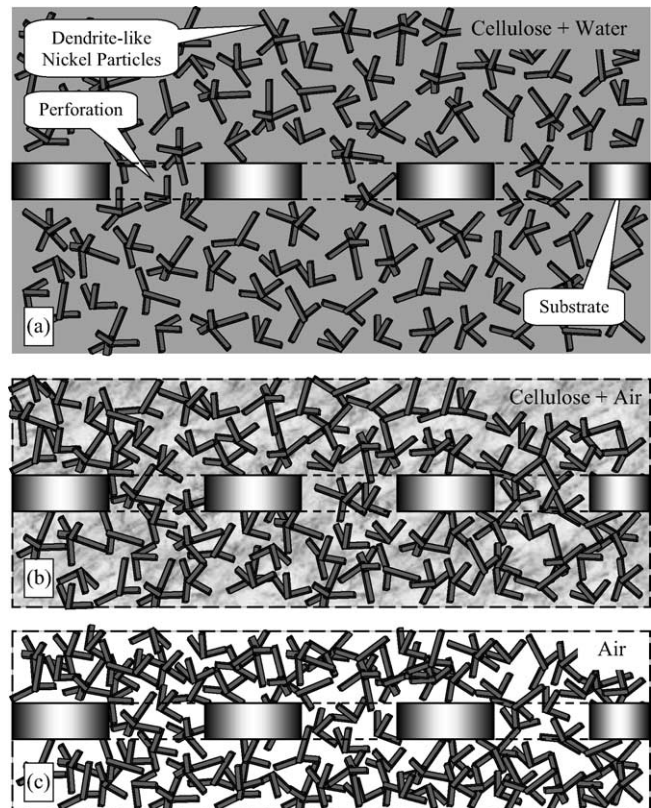


Fig. 1. Three phases in the processing of porous nickel plaques: (a) a perforated substrate is coated with a uniform (constant thickness) layer of fluid slurry (high-viscosity paste) containing individual nickel particles in a solution of cellulose in water, (b) dried plaque (nickel particles are still not chemically bonded to each other or to the substrate, instead temporary attachment is achieved through bonding to methyl cellulose molecules, and (c) sintered porous nickel plaque (nickel particles are bonded chemically and form a continuous porous network bonded to the substrate).

we have attempted to improve plaque adhesion by means of pre-coating the substrate with a slurry containing a finer Ni powder (following the suggestion of Ettel [1997]). The hypothesis is that the finer Ni powder is easier to sinter and may lead to greater contact density with the substrate, resulting in strengthened bonding between the substrate and the plaque. We also hypothesize that the bulk powder slurry that is then deposited over the pre-coated layer penetrates into it and offers a strong bond at the interface between the pre-coating layer and functional porous plaque as well.

2. Tape casting

An ordinary slurry based procedure widely used for the fabrication of the tape-like porous nickel compacts in industry was selected for the study of the coating process and adhesive properties of the resulting coupled (plaque/substrate) structures. The slurries were produced from filamentary nickel powders (Table 1) supplied by INCO Ltd. (types 255, 110, and 210) and partially described in Refs. [4–8,11].

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