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Compression-recovery model of absorptive glass mat (AGM) separator guided by X-ray micro-computed tomography analysis

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HIGHLIGHTS

- Analytical model for compression-recovery behavior of AGMs has been developed.
- 3D fiber orientation from X-ray micro-computed tomography was used as input data.
- A comparison was made between theoretical models and experimental results.

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ABSTRACT

Absorptive glass mat (AGM) separators play a key role in enhancing the cycle life of the valve regulated lead acid (VRLA) batteries by maintaining the elastic characteristics under a defined level of compression force with the plates of the electrodes. Inevitably, there are inherent challenges to maintain the required level of compression characteristics of AGM separators during the charge and discharge of the battery. Herein, we report a three-dimensional (3D) analytical model for predicting the compression-recovery behavior of AGM separators by formulating a direct relationship with the constituent fiber and structural parameters. The analytical model of compression-recovery behavior of AGM separators has successfully included the fiber slippage criterion and internal friction losses. The presented work uses, for the first time, 3D data of fiber orientation from X-ray micro-computed tomography, for predicting the compression-recovery behavior of AGM separators. A comparison has been made between the theoretical and experimental results of compression-recovery behavior of AGM samples with defined fiber orientation characteristics. In general, the theory agreed reasonably well with the experimental results of AGM samples in both dry and wet states. Through theoretical modeling, fiber volume fraction was established as one of the key structural parameters that modulates the compression hysteresis of an AGM separator.

1. Introduction

Valve regulated lead acid (VRLA) batteries, a common variant of traditional lead-acid batteries, have evolved as a major constituent serving the worldwide battery market share. Technological advances in terms of requirement of powerful and reliable power sources pose an enormous challenge for VRLA batteries to compete with the emerging dominance of lithium ion and nickel based batteries [1–3]. Some of these challenges can be realized by deconstructing the design aspects of key attributes of VRLA batteries. Absorptive glass mat (AGM) used as a separator plays a key role for the successful functioning of VRLA batteries throughout the lifetime [4]. One of the major functions of the AGM separator is to enhance the life cycle of the VRLA battery by maintaining the elastic characteristics under defined level of compression forces with the plates of the electrodes when it is soaked in an electrolyte solution [5]. This is a challenging task for an AGM separator as it shrinks in the thickness direction due to the fact that the constituent glass fibers have a greater affinity to each other in the wet state that inevitably results in a loss of compression force [6–9]. Further, there is a reduction in the thickness of an AGM separator

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after the load is being removed (also known as recovery) due to the losses in internal friction and the viscoelastic effects of glass fibers [8,10]. Consequently, these effects result in the loss of the contact between the separator and the electrode plates. Such a loss of contact triggers the expansion of the positive active material leading to the premature capacity loss (PCL2), which inevitably reduces the life cycle of the battery [10–16]. One of the ways to circumvent the issue of compression restraint and resilience is by either modifying the AGM separator using polymeric emulsions [17] or utilizing the gelled electrolyte resulting in 'acid jellying separator' (AJS) [15,18]. Although, AJS emerged as a promising separator but due to high cost, low porosity of the material and the issues pertaining to the commercial viability potentially encouraged to modify the existing AGM [8]. Alternatively, the improvement in the compression properties of AGM inside the cell can be attained by precompressing the separator before being assembled, which effectively enhances the density of the separator [10]. According to Zguris [1,9], higher density of separator causes lower loss of compression force from dry to wet state and overall enhances the battery cycle life. On the other hand, the presence of coarser glass fibers in a multi-layered separator can alter its compression characteristics [19]. Therefore, there is a need to develop an analytical model of compression-recovery characteristics of AGM separators that guides us to improve such a behavior by modulating the constituent fiber and structural parameters.

In general, an AGM separator is a voluminous, wet-laid nonwoven material having constituent glass fiber diameters in a range of micron or sub-micron scale [8]. Interestingly, the compression characteristics of AGM separators were found to be similar as that of organic or thermoplastic fiber based nonwoven separators that are used in alkaline batteries [9]. The compression characteristics of such thermoplastic based nonwoven materials have been investigated in detail via phenomenological and micromechanical models [20]. Phenomenological models are developed by formulating the empirical relationships that are incapable of elucidating the role of fiber or structural parameters [21]. On contrary, the micromechanical models require the use of continuum mechanics approach presuming the homogeneity in the fiber network of nonwovens [22]. For understanding the compression behavior of nonwoven materials, most of the micromechanical models developed in the literature assumed fiber bending as the main mode of deformation [23–29]. Ball et al. [30] have also corroborated the deformation of an AGM separator under compression loading in terms of fiber bending. Despite the fact that micromechanics approach can form the basis of analytical model of compression properties of AGM separators, certain pre-requisites are still desirable, as highlighted below. Firstly, the realistic three-dimensional (3D) fiber orientation data should be used as input data to predict the compression properties of an AGM separator. Secondly, the production method of wet laying process of an AGM separator can sometimes result in the preferential orientation of fibers where majority of the fibers are aligned in the machine (production) direction. Therefore, the analytical model should be capable of handling the preferential alignment of fibers. Lastly, designing an AGM separator with multi-fold characteristics requires the prediction of recovery characteristics in the wet state. Carnaby and Pan [24] have developed the model for recovery behavior of nonwoven materials but the internal friction losses have not been accounted, which is of paramount importance for AGM separators as glass fibers are held together by means of frictional forces [8,31]. Based on these gaps in the literature, the central aim of the research work was to develop a 3D analytical model of compression-recovery characteristics of AGM separators that formulates a direct relationship with the constituent fiber and structural parameters of the separators. X-ray micro-computed tomography (microCT) analysis has assisted in obtaining the 3D data of fiber orientation distributions in AGM separators, which were further used as input parameters to obtain the compression-recovery characteristics via theoretical modeling. Subsequently, a comparison has been made between the theory and experiments of compression-recovery characteristics of AGM separators with pre-defined fiber orientation characteristics. Finally, a roadmap to enhance the compression-recovery behavior of AGM separators has been proposed on the basis of parametric analysis.

2. Theoretical framework

AGM comprises of network of glass fibers that are distributed stochastically by means of wet-laying process. Accordingly, an in-depth understanding of structural analysis of fiber networks is essential in order to analyze the contact forces between the fibers during compression and recovery of AGM. Before discussing the theoretical framework, it is necessary to highlight some of the assumptions considered to simplify the analysis.

1. The fibers are considered to be distributed homogenously within an AGM in order to apply the continuum theory of elasticity to the system [22].
2. Each fiber segment is presumed to be straight between the two successive fiber-fiber contacts. Since, AGM is an ensemble of glass fibers that are rigid entities with linearly elastic characteristics [9,30].
3. Various deformation types (torsion, compression, tensile) of fiber segments are considered to be small such that they can be neglected [24].
4. New contacts between the fibers are not formed specifically for a small amount of loading or even for small incremental load [24].

2.1. Structural analysis of AGM

As aforementioned, AGM comprises of network of glass fibers, which have similar physical and geometrical characteristics of discrete media. Accordingly, the mechanics of discrete media has been formulated at three defined length scales [32,33]. Pan et al. [33] have described the "microelement" as a lowest length scale on which all the continuum concepts are valid. Next, an intermediate length scale known as "mesodomain" that comprises of statistical ensemble of microelements. Here, the physical and geometrical parameters of the mesodomain are predicted statistically based upon the properties of microelements and it is a representative element of the network, on which the continuum mechanics is also applicable. Lastly, the "macroscopic" length scale is developed by means of defined non-intersecting mesodomains. In our analysis, AGM separator is presumed to be macroscopic level entity consisting of fiber segments (microelements) between the two successive contacts in the mesodomain, as illustrated in Fig. 1. Accordingly, the alignment of each fiber segment in an AGM separator is defined by in-plane (ϕ) and out-of-plane (θ) orientation angles, also known as azimuthal and polar angles, respectively, in a typical spherical co-ordinate system. The probability that a direction of the fiber segment lies in the infinitesimal small range of angles, θ and $\theta + d\theta$, and ϕ and $\phi + d\phi$ is given by $\Omega(\theta, \phi) \sin\theta d\theta d\phi$, where $\Omega(\theta, \phi)$ is a probability density function. In such a case, the following normalization condition must be fulfilled.