## Author's Accepted Manuscript

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 PII:
 S0376-7388(16)30498-7

 DOI:
 http://dx.doi.org/10.1016/j.memsci.2016.05.055

 Reference:
 MEMSCI14530

To appear in: Journal of Membrane Science

Received date: 16 September 2015 Revised date: 11 April 2016 Accepted date: 29 May 2016

Cite this article as: Nils Müller, Ulrich A. Handge and Volker Abetz, Physical ageing and lifetime prediction of polymer membranes for gas separation processes, *Journal of Membrane Science* http://dx.doi.org/10.1016/j.memsci.2016.05.055

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## **ACCEPTED MANUSCRIPT**

Physical ageing and lifetime prediction of polymer membranes for gas separation

processes

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## Abstract

The free volume in glassy polymers for gas separation processes is affected decidedly by physical ageing. In this work, we analyse the long-term volume relaxation of glassy polymer membranes using statistical data analysis in order to predict the lifetime of these membranes with minimal experimental effort. Several lifetime definitions are proposed and discussed in their use for the prediction quality. A principal objective of this work is the statistical analysis of the phenomenological ageing model developed by Struik using the bootstrapping method. We analyse the stepwise reduction of the number of data points used in model fitting. Based on these results we propose an optimal modelling and experimental strategy directed at the prediction of thickness dependent volume relaxation. In particular, the influence of the duration of pointwise measurements is discussed in detail. Our investigation is concluded by the dimensional analysis of the Struik equation, resulting in a master curve concept. The analysis of the master curve reveals a weak dependence of the lifetime of a membrane on its initial condition. The lifetime is proportional to the relaxation time for a criterion based on the absolute permeability and inverse proportional to the relaxation time for a criterion based on the derivative of the permeability for the chosen threshold parameters.

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