Appendix A

The phase diagram was estimated using the data given by Druz et. al.\textsuperscript{[114]} in Table 2 of their publication about the miscibility of PE/EVA blends.

\[ M_{\text{PE}} = 100\,000 \text{ g/mol}, \quad M_{\text{0PE}} = 28, \quad N_1 \sim 3571 \]

\[ M_{\text{EVA}} = 15\,500 \text{ g/mol}, \quad M_{\text{0EVA}} = 42, \quad N_2 \sim 370 \]

\( N_1 \) and \( N_2 \) are the degree of polymerization.

The dependence of the existent phases in the PE/EVA blends on molar mass of PE (\( M_{\text{PE}} \)) is given by the following equations\textsuperscript{[114]}:

\[ \Phi_1' = \Phi_{1\infty} + \frac{k'}{M} \quad \text{and} \quad \Phi_1'' = \Phi_{1\infty} + \frac{k''}{M} \] (A-1)

where \( \Phi_1' \) and \( \Phi_1'' \) are the volume fractions at equilibrium.

The extrapolation allows to determine the phases at the temperature of the experiment and the solubility of the components (PE and EVA) at \( M \to \infty \).

We assume that the interaction parameter \( \chi \) is:

\[ \chi = A + \frac{B}{T} \]

Therefore, (in rough approximation) according to Ref. 114:

\[ \chi = -0.03 + \frac{14.25}{T} \] (A-2)

\( \chi_{393} = 6.3 \times 10^{-3} \) and \( \chi_{433} = 2.9 \times 10^{-3} \) are the binary interaction parameters at temperatures of 393 and 433 K respectively.

After Flory-Huggins approximation (see Appendix B): \( \Phi_{1\infty} \propto \frac{1}{\chi} \) and \( \Phi_{1\infty}' \propto \frac{1}{\chi} \). Hence, we assume temperature dependence of these quantities and the coefficients \( k' \) and \( k'' \):

\[ \Phi_{1\infty}' = \frac{\alpha' + \beta'T}{\chi} \quad \text{and} \quad k' = a' + \frac{b'}{T} \]

which results in:

\[ \Phi_{1\infty}' = \frac{-3.55 + 0.02T}{10^4 \chi} \quad \Phi_{1\infty}'' = \frac{-6.14 + 0.0166T}{10^4 \chi} \] (A-3)

\[ k' = 0.0288 + \frac{212.7}{T} \quad k'' = -0.805 + \frac{251.0}{T} \]
where $\chi$ is given by Equation (A-2). The phase compositions can be calculated by using Equations (A-1) – (A-3).

**Appendix B**

Flory-Huggins (FH) approximation:

\[
\frac{\Delta \mu_1}{RT} = \ln \frac{\phi}{N_1} + \left(1 - \phi \right) \left( \frac{1}{N_1} - \frac{1}{N_2} \right) + \chi (1 - \phi)^2 \tag{B-1}
\]

\[
\frac{\Delta \mu_2}{RT} = \ln \left(1 - \phi \right) - \phi \left( \frac{1}{N_1} - \frac{1}{N_2} \right) + \chi \phi^2 \tag{B-2}
\]

where $\Delta \mu_1$ and $\Delta \mu_2$ are the chemical potential of PE and EVA.

There is phase equilibrium when:

$\Delta \mu_1 = \Delta \mu_1^*$ and $\Delta \mu_2 = \Delta \mu_2^*$ \hspace{1cm} (B-3)

Using (B-1) and (B-2), the first equation of (B-3) refers to $N_1 \to \infty$

\[-\left(1 - \phi^* \right) + N_2 \chi (1 - \phi^*)^2 = -\left(1 - \phi^* \right) + N_2 \chi (1 - \phi^*)^2 \] and the solution of this equation is:

\[1 - \phi^* = \frac{1}{N_2 \chi}(1 - \phi^*) \tag{B-4}\]

By using (B-4) we obtain the second equation of (B-3):

\[
\ln \left[ \frac{1 - \chi N_2 (1 - \phi^*)}{\chi N_2 (1 - \phi^*)} \right] = 2 \left[1 - 2 \chi N_2 (1 - \phi^*) \right] \tag{B-5}
\]

which leads to: $\chi N_2 (1 - \phi^*) = 0.5$

$N_1$ and $N_2$ are degree of polymerization of PE and EVA respectively.