## **Supplementary Material**

Herein, we presented a series of graphics that are not included in the paper. This material supports the experimental work of our investigation.

## 1. Determination of ionisation degree

The results of the percentage of DMAE groups, the ionisation degree and the zeta potential values for polymers Eudragit<sup>®</sup> E 100 and Eudragit<sup>®</sup> E PO, as well as their processed forms EuCl-E-100 and EuCl-E-PO are presented as potentiometric titration profiles.



**Figure S1.** Potentiometric curves obtained for the polymeric materials derived from Eudragit E-100.

The formation of the ionised species of the EuCl polymers were observed by a qualitative variation in the solubility of the Eudragit<sup>®</sup> E polymers, which changed from a heterogeneous mixture into a homogeneous dispersion, due to the protonation of the DMAE groups. In the potentiometric titrations study of the Eudragit E 100 polymers a single inflection point corresponding to the presence of the DMAE groups in the polymeric backbone was observed.

## 2. Determination of zeta potential.

For the Eudragit® salt derivatives.

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Figure S2. Zeta potential curves for EuCl– E-100.



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Figure S3. Zeta potential curves for EuCl-E-PO.

3. Structural characterisation of polymer materials: NMR spectra for the Eudragit® E derived materials.

 $^{1}$ H and  $^{13}$ C NMR spectra are presented; the experiments that were carried out were  $^{1}$ H– $^{1}$ H COSY,  $^{1}$ H– $^{13}$ C HMQC and HMBC.



Figure S4. <sup>1</sup>H NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S5. <sup>13</sup>C NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S6. COSY NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S7. ZOOM COSY NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S8. HSQC NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S9. HMBC NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S10. DEPT-135 NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit® E 100.



Figure S11. J-MOD NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E 100.



Figure S12. <sup>1</sup>H NMR spectra (600 MHz, CDCl<sub>3</sub>) of Eudragit<sup>®</sup> E PO.



Figure S13. <sup>1</sup>H NMR spectra (600 MHz, D<sub>2</sub>O) of EuCl-E-100.



Figure S14. <sup>13</sup>C NMR spectra (600 MHz, D<sub>2</sub>O) of EuCl-E-100.







Figure S16. <sup>1</sup>H NMR spectra (600 MHz, D<sub>2</sub>O) of EuCl-E-PO



4. Heating cycles (Cycle 3) in the DSC thermal analysis.

Figure S17. Cycle 3 (heating) for Eudragit® E100



Figure S18. Cycle 3 (heating) for Eudragit® E PO



Figure S18. Cycle 3 (heating) for EuCl-E - 100



Figure S19. Cycle 3 (heating) for EuCl-E-PO



**Figure S20.** Representative scheme of the fusion / degradation process for the Eudragit E® polymeric materials and their processed forms. (Performed in capillary melting point apparatus).