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Sensing Exposure Time to Oxygen by Applying a Percolation-Induced Principle

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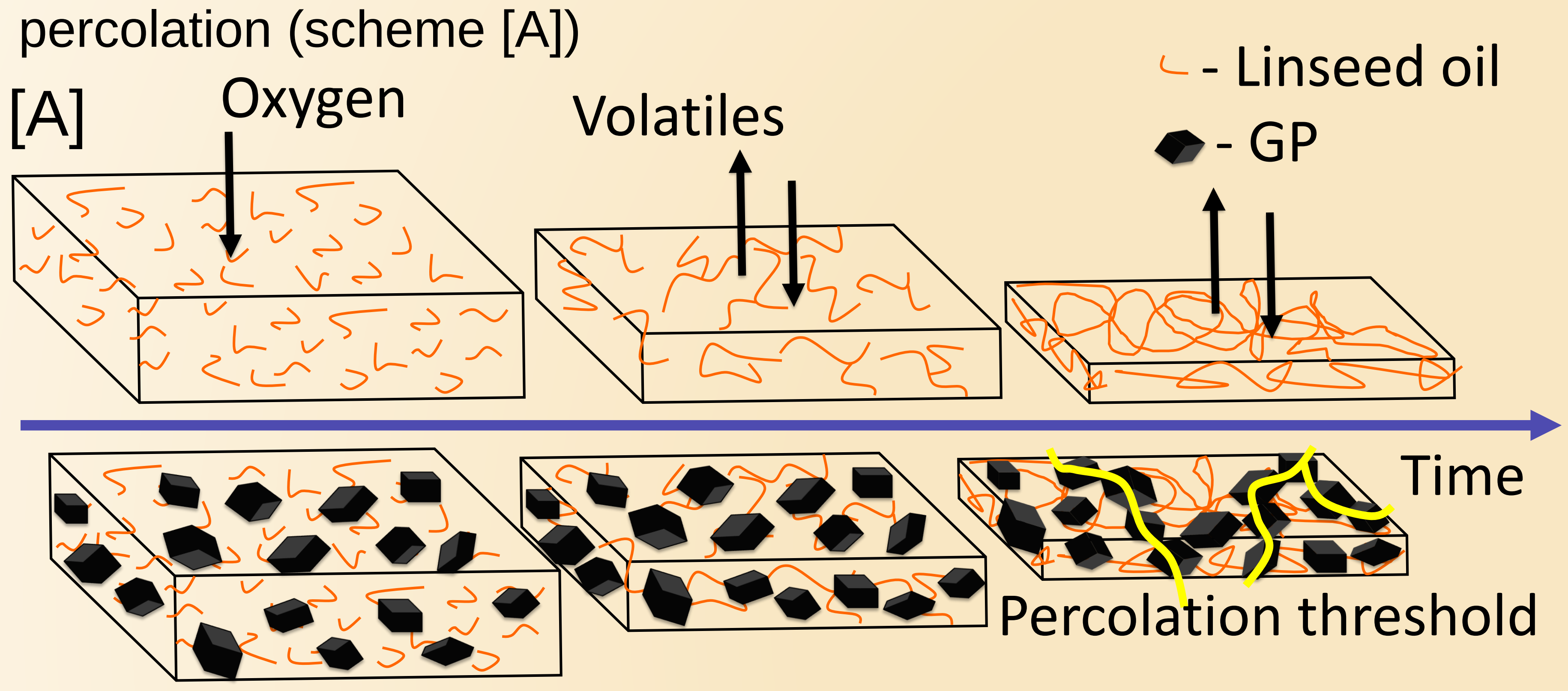
Abstract

The determination of food freshness along manufacturer-to-consumer transportation lines is a challenging problem that calls for cheap, simple, and reliable sensors inside food packaging. We present a novel approach for oxygen sensing in which the exposure time to oxygen—rather than the oxygen concentration per-se is monitored. We developed a nontoxic hybrid composite-based sensor consisting of graphite powder (conductive filler), clay (viscosity control filler) and linseed oil (the matrix). Upon exposure to oxygen, the insulating linseed oil is oxidized, leading to polymerization and shrinkage of the matrix and hence to an increase in the concentration of the electrically conductive graphite powder up to percolation, which serves as an indicator of food spoilage. In the developed sensor, the exposure time to oxygen is obtained by measuring the electrical conductivity though the sensor. The sensor functionality could be tuned by changing the oil viscosity, the conductive filler aspect ratio, and/or the clay concentration.

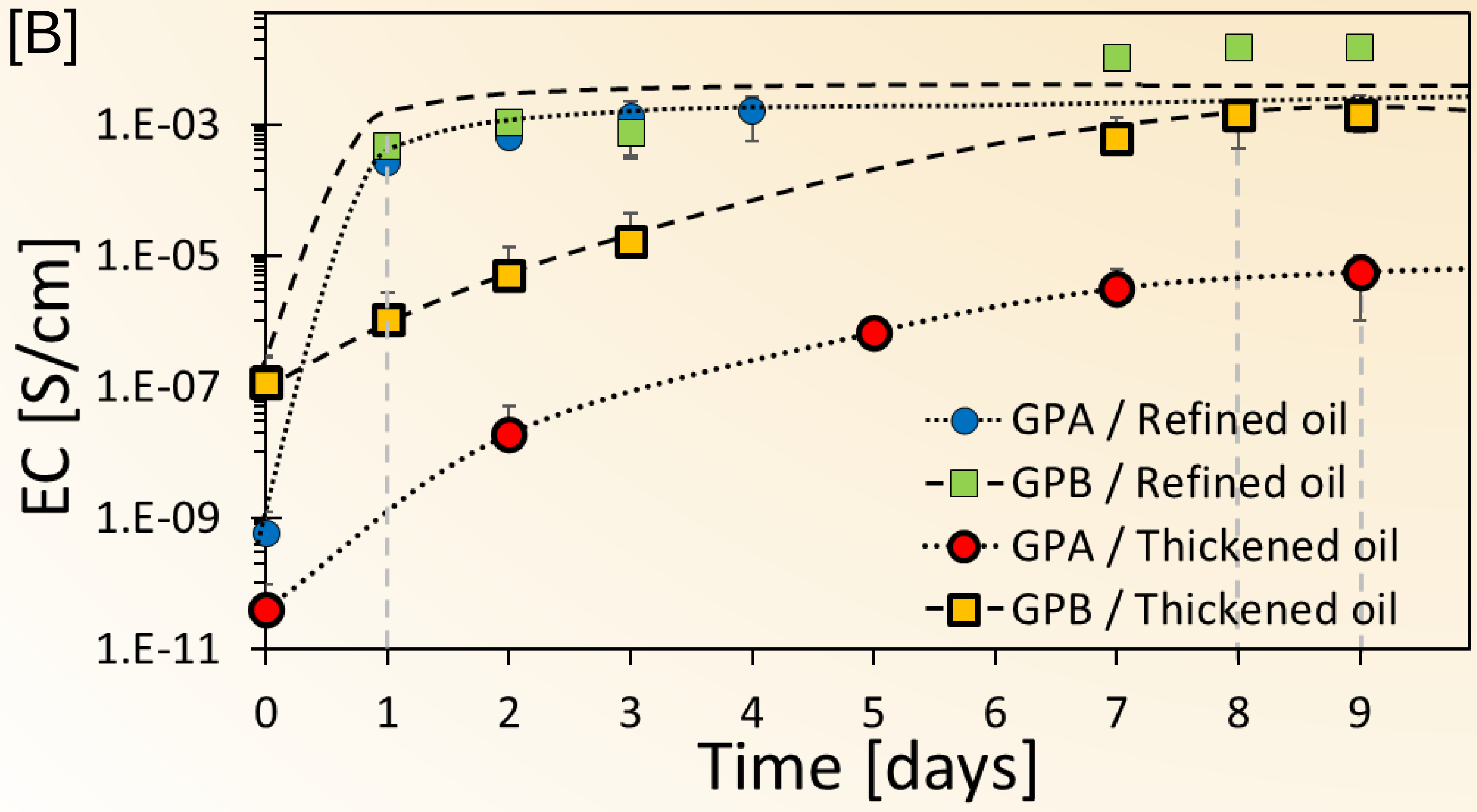
Oil Oxidation - Percolation Threshold

The autoxidation process of linseed oil induces shrinking. When conductive fillers are added (graphite powder (GP)), shrinking results in network formation, and sharp increase in the EC due to percolation (scheme [A])



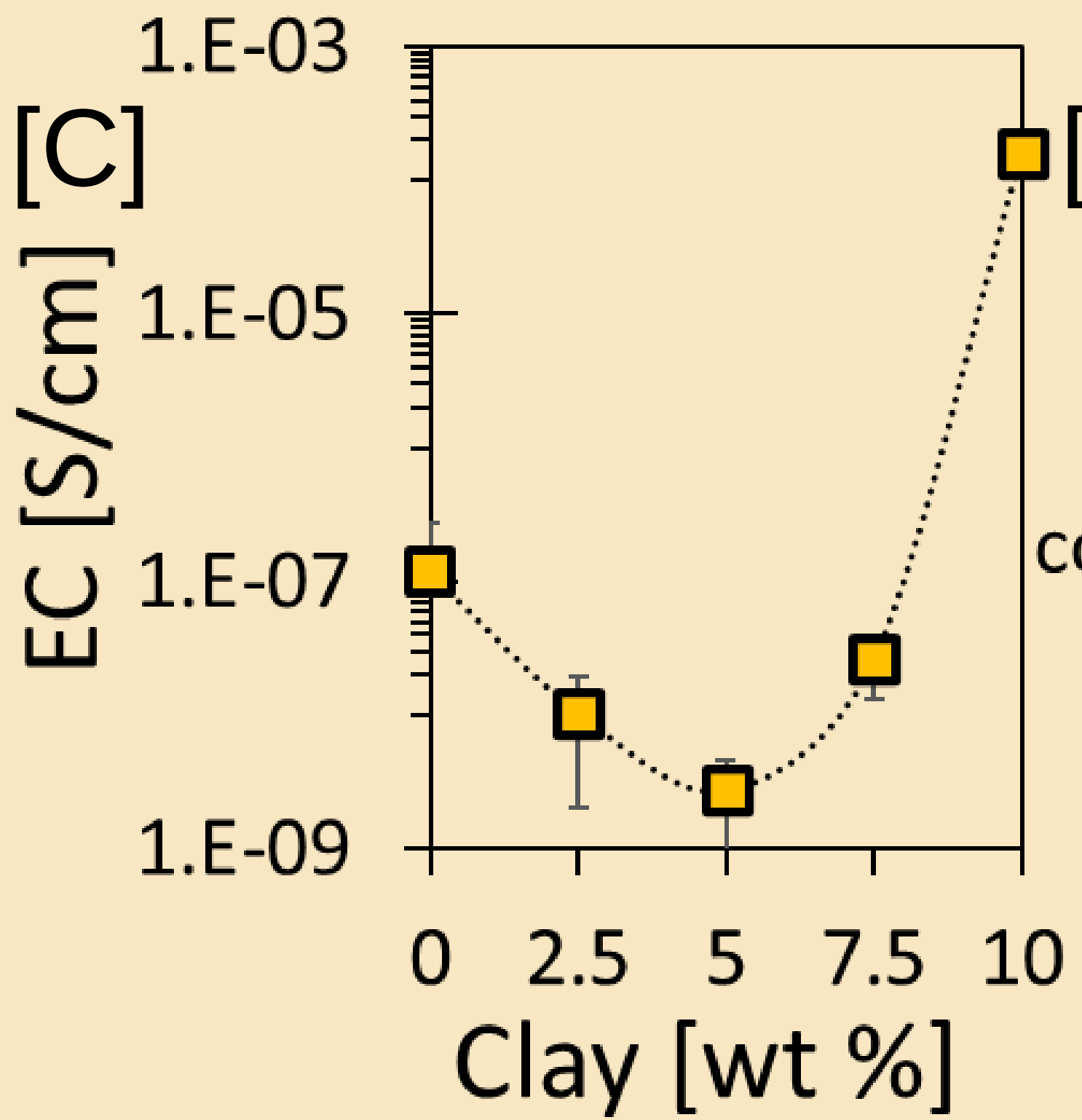
Oil Viscosity and Filler Aspect Ratio Effects

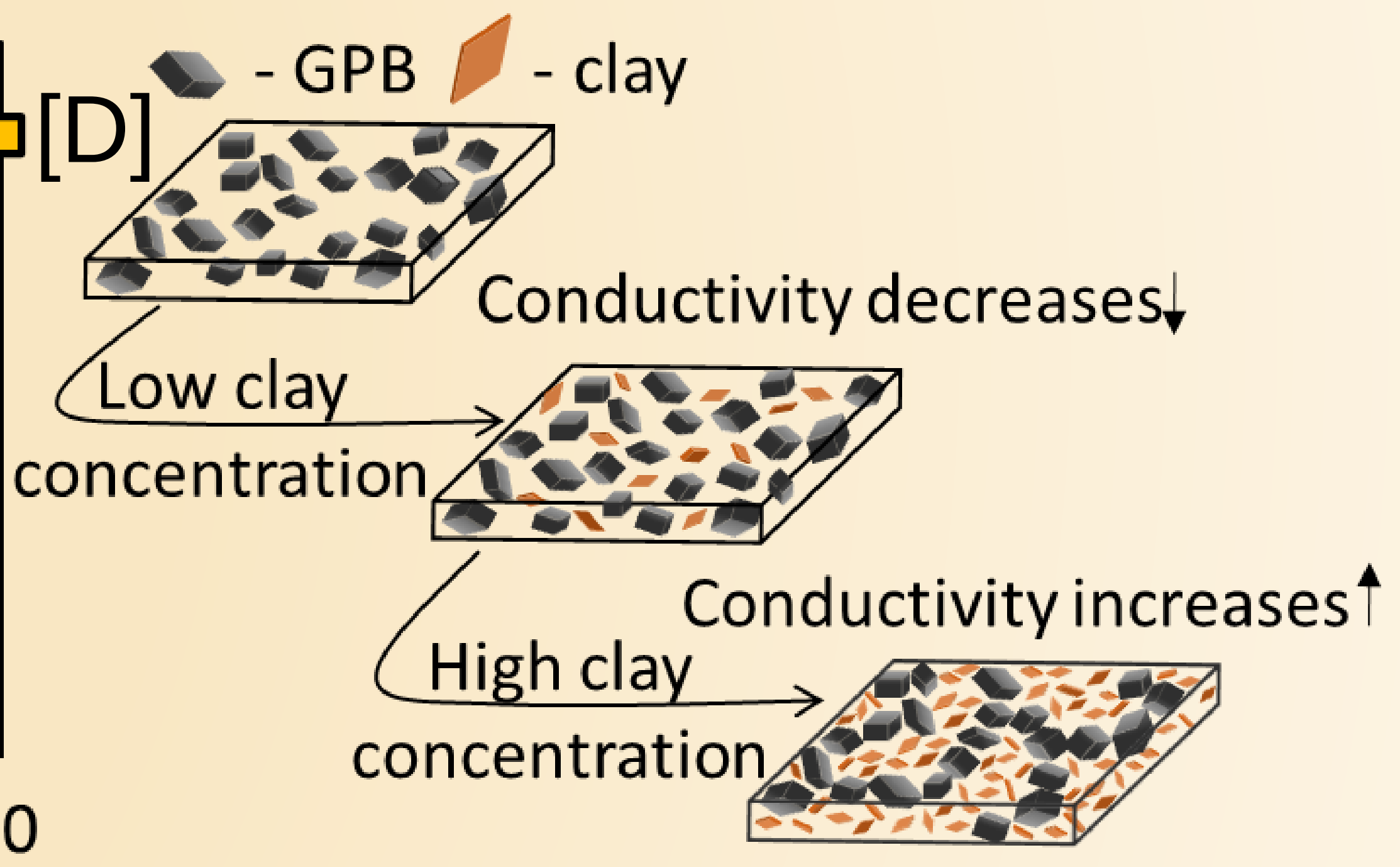
Temporal EC of refined and thickened linseed oils loaded with 25 wt% GPA or GPB upon exposure to air is shown [C]. The dashed lines denote the indication of air-exposure time (IAET) during which the composite ends the shrink-percolation process. The thickened linseed oil systems have lower EC values due to their higher viscosity (by 2 orders of magnitude, than the refined), which restricts the mobility of the fillers. Given the higher aspect ratio of GPB (39±6 of GPB vs. 1.5±0.4 of GPA), the EC value of GPB is higher, due to the promotion of the EC conducting network formation. However, despite the aspect ratios difference, their percolation time is similar.



Clay-Induced Excluded Volume Effect

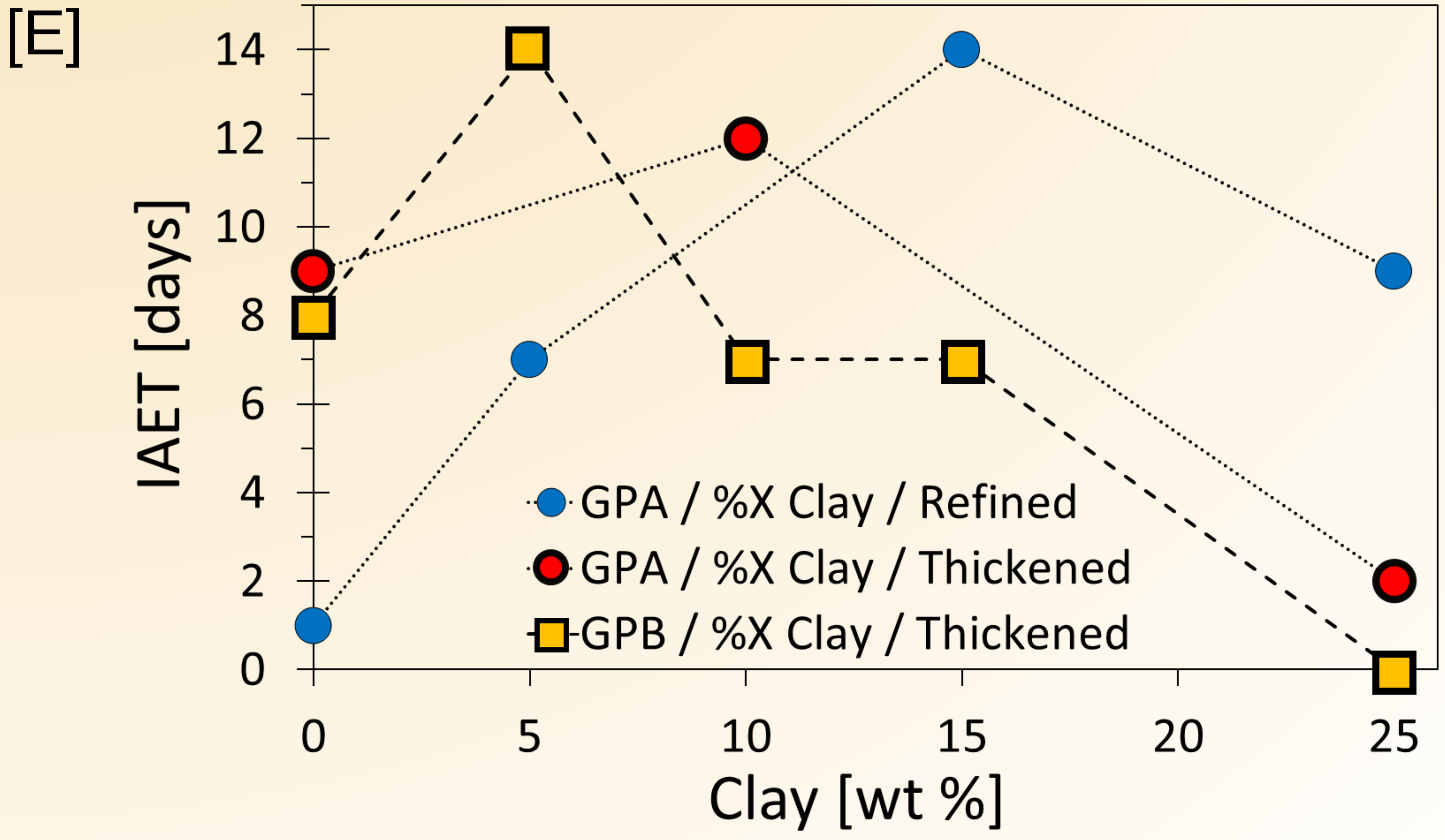
To maintain the integrity of the sensor (i.e., keeping a constant film thickness and avoiding dripping), we added to the GP-loaded oil a second non-conductive filler material - clay (Cloisite 20A) to increase its viscosity. Upon addition of a low concentration of clay, the EC decreased [C], since the clay particles impeded the formation of the GP network [D]. However, at a higher clay concentration, there was an increase in the EC [C], most probably due to the excluded volume effect by which the added volume of clay particles forced the conductive GPB fillers to percolate [D].





Manipulation of The IAET

The evaluated IAET was determined for air-exposed linseed oil loaded with a 25 wt% fixed GPA or GPB and different clay concentration [E]. At a low clay concentration, there is an increase in the IAET, due to decreased oxygen permeability. However, above a maximal clay concentration, there is a decrease in IAET, due to the excluded volume effect, resulting in GP percolation.



Conclusions

- ☐ The IAET values for various compositions of the proposed sensor could serve as a road map for sensor design, by adjusted oil viscosity, aspect ratio of the conductive filler and the clay loading.
- ☐ Excluded volume effect is observed at high clay loadings.

References

(1) Sensors 2020, 20(16), 4465 **(2)** Polymer Degradation and Stability 1999, 65(2), 303-313 **(3)** ACS Appl. Mater. Interfaces 2017, 9(8), 7556–7564