Response letter to reviewer # 1

**Title:** “Stearic acid/layered double hydroxides composite thin films deposited by combined laser techniques”

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We thank the reviewers for their valuable evaluation of the manuscript, as well as for their comments, which helped us strengthen the presentation of our results.

We would like to emphasize that the aim of this paper is to study stearic acid-layered double hydroxide (LDH) composite films, with controlled wettability capabilities, deposited by a combined pulsed laser deposition (PLD) - matrix assisted pulsed laser evaporation (MAPLE) system. We believe that these results are very promising for a variety of applications, in particular for using them as protective coating.

We have modified the text to respond to all the issues and have elaborated the responses below. We hope that the revised manuscript fulfils all requirements for being published in Molecules, section Physical Chemistry, issue Surfaces, Interfaces, Coatings and Nanostructures of Molecular and Soft Materials.

Please find enclosed the list with the required modifications.

The reviewer’s comments are marked with the letter Q.

The answers of the authors are marked with the letter A.

In the manuscript text we have added in red the modifications.

We have also checked the manuscript to eliminate grammatical and typing errors.

Q1: The abstract part needs to be rewritten with a special focus on the aim of the work (explain why you obtain this type of thin films - short information).

A: We have modified the abstract.

*We report on the investigation of stearic acid-layered double hydroxide (LDH) composite films, with controlled wettability capabilities, deposited by a combined pulsed laser deposition (PLD) - matrix assisted pulsed laser evaporation (MAPLE) system. Two pulsed lasers working in IR or UV were used for experiments, allowing the use of proper deposition parameters (wavelength, laser fluence, repetition rate) for each organic and inorganic component material. We have studied the time stability and wettability properties of the films and we have seen that the morphology of the surface has a low effect on the wettability of the surfaces.*
obtained composite films consist in stearic acid aggregates in LDH structure, exhibiting a shift to hydrophobicity after 36 months of storage.

Q2: Could you correct: line 86 should be 40°C and line 110, should be 2 J/cm².
A: We have made the corrections mentioned above.

Q3: The structure of thin film was described in introduction but will be more clear when you put a new figure with the scheme of this structure.
A: We have modified figure 1, to evidence the deposited composite thin films.

Q4: The resolution of figure 4 is very low, so please adjust (could you remove “4 um” from the thirds scheme of figure 4).
A: The figure has been modified and corrected.

Q5: The Conclusion part needs to be rewritten.
A: We have entirely changed the conclusions.

Composite organic-inorganic (stearic acid/Mg,Al-LDH) films were obtained by using a set-up combining the standard PLD and MAPLE. A MAPLE deposition system was adapted to provide the possibility of irradiating two concentric targets, with two different laser beams. The composite films exhibit long-term behavior different from the inorganic LDH films deposited by PLD and the stearic acid film deposited by MAPLE. A shift to hydrophobicity after 36 months of storage was observed for both composite films although they display different morphologies and roughness. The result was explained, based on FT-IR measurements, on the reorientation in time of the alkyl chains of the highly dispersed stearic-acid aggregates embedded between LDH oriented layers. This particular structure is due to the simultaneous deposition of both the inorganic and the organic component. Such films exhibiting long-term stability and enhancement of their hydrophobicity are good candidates to be used as protective coatings. The combined PLD-MAPLE setup displays a large versatility in terms of ability of producing composite complex films or organic-inorganic heterostructures and sandwiches without opening the deposition chamber and by this, reducing the contamination.