

Cod proiect PN-III-P2-2.1-PED-2021-1870

Nr contractului 646PED/2022

**Titlul proiectului (în română) ECOTECHNOLOGIE DE OBȚINERE A UNOR
FITOINGREDIENTI INCAPSULATI IN HIDROGEL PE BAZA DE COMPLECSI
BIOACTIVI IMOBILIZATI IN MATRICE DE HIDROXIZI DUBLI LAMELARI**

Titlul proiectului (în engleză)

**ECOTECHNOLOGY FOR OBTAINING PHYTOINGREDIENTS ENCAPSULATED IN
HYDROGEL BASED ON BIOACTIVE COMPLEXES IMMOBILIZED IN A LAYER
DOUBLE HYDROXIDE MATRIX**

Acronimul proiectului DUACTIONMER

Valoare totală contract: 658.675,00 lei

Din care :

Sursa 1 de la bugetul de stat : 598.795,00 lei

**Sursa 2 - din alte surse atrase: 59.880,00 lei
(cofinanțare)**

Durata contractului: 24 luni

REPARTIZAREA SUMELOR PE ANII BUGETARI

ANUL	Sursa 1 (lei)	Sursa 2 (lei)	TOTAL(lei)
2022	146.765,00	13.430,00	160.195,00
2023	345.176,00	46.450,00	391.626,00
2024	106.854,00	0,00	106.854,00
TOTAL	598.795,00	59.880,00	658.675,00

ORGANIZAȚIILE PARTENERE ÎN PROIECT

Denumirea organizației participante în proiect	Acronim organizație	Tip organizație	Rolul organizației în proiect (Coordonator/Partener)
(CO) Universitatea din București	UB	UNI	Coordonator
(P1) Institutul Național de Cercetare-Dezvoltare pentru Chimie și Petrochimie ICECHIM	INCDCP ICECHIM	INCD	Partener P1
(P2) CENTRUL DE CERCETARE SI PRELUCRARE A PLANTELOR MEDICINALE PLANTAVOREL SA	PLV	Întreprindere Mică	Partener P2

Repartizarea fondurilor pe parteneri

	2022		2023		2024		TOTAL	
	Finanțare de la bugetul de stat	Finanțare din alte surse atrase(contribuție financiară proprie)	Finanțare de la bugetul de stat	Finanțare din alte surse atrase(contribuție financiară proprie)	Finanțare de la bugetul de stat	Finanțare din alte surse atrase(contribuție financiară proprie)	Finanțare de la bugetul de stat	Finanțare din alte surse atrase(contribuție financiară proprie)
CO	57972	0	121313	0	57239	0	236524	0
P1	49166	0	101815	0	49615	0	200596	0
P2	39627	13430	122048	46450	0	0	161675	59880
Total	146765	13430	345176	46450	106854	0	598795	59880

Abstract: Proiectul urmărește dezvoltarea unor eco-tehnologii integrate validate (TRL4) cu dublu scop: 1) de obținere a ingredientelor bioactive încapsulate în hidrogel pe baza fitocomplexșilor din 2 plante medicinale (Rhamnus frangula L (RfL) și Helianthus tuberosus L (HtL) cu biocompatibilitate crescută care sunt protejate contra degradării prin imobilizarea într-o

matrice de hidroxid dublu lamelar (LDH) si 2) de valorificare a resturile vegetale ramase dupa extractie, ca adsorbant ieftin al poluanților din ape reziduale. Conceptul de bază al tehnologiei (TRL2) pornește de la rezultatele preliminare obținute de partenerii din consorțiu în i) extracția fitocomplexilor din RfL și HtL, ii) imobilizarea anionilor organici în matrici Mg/Al-LDH și Zn/Al-LDH, iii) obținerea hidrogelurilor hibride cu fitoextracte bioactive din Rosa Canina și Hypericum Perforatum L imobilizate in zeoliți naturali și iv) tratarea apelor reziduale poluate cu metale grele și poluanți organici. Proiectul extinde aplicarea fitoextractelor dublu-protejate la noi sisteme bazate pe fitoextracte RfL și HtL incorporate în matrici Mg,Ca/Al,Fe-LDH și încapsulate apoi în hidrogeluri. Tehnologia integrată, cu producere minimă de deșeuri va permite utilizarea ingredientelor active obținute în suplimente alimentare cu efecte benefice în reglarea unui tranzit intestinal lent prin aportul constant de glucofranguline din RfL și de electroliți precum Mg/Ca si Fe, având totodată și efectul benefic de prebiotic, îmbunătățind absorbția mineralelor și scăderea colesterolului datorită HtL. Se vor obține si beneficii în tratarea apelor reziduale. Folosirea produselor obținute la suplimente alimentare este benefică pentru bunăstarea oamenilor, scăzând efectele negative ale stilului de viața haotic, cu prea multe ore de muncă, stres continuu și decizii greșite în ce privește alimentația, care toate conduc la repercusiuni grave asupra sănătății sistemului digestiv.

Abstract: The project aims the development of integrated validated eco-technologies (TRL4) with double action: 1) obtaining hydrogel encapsulated bioactive ingredients based on phytocomplexes from 2 medicinal plants (e.g. Rhamnus frangula L (RfL) and Helianthus tuberosus L (HtL)) with increased biocompatibility and protected from degradation by

immobilization in a layer double hydroxide (LDH) matrix, and 2) valorization of spent vegetal after extraction as cheap adsorbent of pollutants from wastewaters. The basic concept of the technology (TRL2) lays on preliminary results obtained by consortium partners in: i) extraction of RfL and HtL phytocomplexes; ii) immobilization of different organic anions in Mg/Al-LDH and Zn/Al-LDH matrices, iii) obtaining of hybrid hydrogels containing bioactive phytoextracts from *Rosa Canina* and *Hypericum Perforatum* L immobilized in natural zeolite and iv) treatment of wastewaters polluted with heavy metals and organic pollutants. This project extends the application of double-protected bioactive phytoextracts to new systems based on RfL and HtL phytoextracts incorporated in Mg,Ca/Al,Fe-LDH matrices and further encapsulated into hydrogels. The integrated technology ensuring minimum waste production, will allow the further utilization of obtained active ingredients in food supplements with benefic effects for improving a lazy intestinal transit due to a constant low apport of glucofrangulins brought by RfL along with electrolytes such as Mg/Ca and Fe, while acting also as prebiotic and promoter of mineral absorption and cholesterol-lowering due to HtL. A benefit for wastewater treatment would also be brought. The use of the obtained products in food supplements could be benefic to people well-being, by reducing the negative effects of their chaotic lifestyle, with too many working hours, continuous stress and most important, poor dietary decisions, all leading to tremendous repercussions on the health of the digestive system.

Obiectivul proiectului:

Obiectivele științifice ale proiectului sunt: 1) selectarea modalității de extracție adecvate a complexilor activi din *Rhamnus frangula* L (RfL) și *Helianthus tuberosus* L (HtL) pentru obținerea de extracte bogate compatibile pentru încapsularea în LDH; 2) selectarea parametrilor

operaționali necesari imobilizării cantității maxime de fitocomplecși în matrice de tip LDH (ce conțin Mg/Ca/Al/Fe); 3) selectarea tipului de hidrogel și a celei mai adecvate metode de sinteză pentru încapsularea eficientă a fitocomplecșilor imobilizați în matrice LDH (RfL-LDH; HtL-LDH) în scopul obținerii HYG-RfL-LDH și HYG- HtL-LDH; 4) determinarea capacității de adsorbție a deșeurilor vegetale rămase după extracția fitocomplecșilor bioactivi pentru reținerea poluanților metale grele (Fe, Ni, Cu, Cr) și coloranților organici (indigo carmin) din ape reziduale în scopul minimizării deșeurilor și dezvoltării unei tehnologii cu dublă acțiune; 5) evaluarea influenței pH-ului la eliberarea in vitro a ingredientelor active din RfL-LDH; HtL-LDH; 6) proiectarea celor două tehnologii de laborator pentru obținerea: a) HYG-RfL-LDH și b) HYG-HtL-LDH și valorificarea resturilor vegetale ca adsorbanți ai poluanților; 7) optimizarea parametrilor operaționali care guvernează extracția fitocomplecșilor activi din RfL și HtL; 8) punerea la punct a celor două tehnologii de laborator optimizate, cu dublă acțiune, aplicate la obținerea fitocomplecșilor în matrice LDH încapsulați în hidrogel; 9) evaluarea influenței pH-ului asupra capacității de eliberare a ingredientelor active din HYG-RfL-LDH and HYG-HtL-LDH in vitro, după optimizare, în vederea determinării cantităților adecvate de compozite care pot fi utilizate în formularea suplimentelor alimentare; 10) demonstrarea funcționalității celor două tehnologii; 11) validarea celor două tehnologii menționate anterior (TRL4). Obiectivele indirecte ale proiectului sunt: a) creșterea calității și diversității suplimentelor alimentare; b) scăderea efectelor secundare negative ale fitocomplecșilor concentrați; c) creșterea competitivității produselor românești pe piața internațională; d) îmbunătățirea vizibilității științifice pe plan național și internațional.

Obiectivul proiectului

(în engleză)

The scientific objectives of the project are: 1) the selection of the adequate extraction route of the active phytocomplexes from *Rhamnus frangula* L (RfL) and *Helianthus tuberosus* L (HtL) in order to obtain rich extracts compatible to their incorporation in LDH; 2) the selection of the operational parameters required for the immobilization of the highest amount of these phytocomplexes in LDH-type matrices (containing Mg/Ca/Al/Fe); 3) the selection of the hydrogel type and of the most adequate synthesis route required for an efficient encapsulation of the phytocomplexes immobilized in the LDH-matrix (RfL-LDH; HtL-LDH) yielding HYG-RfL-LDH and HYG- HtL-LDH; 4) the determination of the adsorption capacity of the vegetal residue remained after the extraction of bioactive phytocomplexes for the retention of heavy metals pollutants (Fe, Ni, Cu, Cr) and organic dyes (indigo carmine) from wastewaters aiming to minimize the wastes while achieving a dual action technology; 5) the assessment of the pH influence on active ingredients in vitro release from RfL-LDH; HtL-LDH; 6) the design of the 2 laboratory technologies (TRL3) for the obtaining of a) HYG-RfL-LDH and b) HYG-HtL-LDH and the valorization of the spent vegetal materials as pollutants adsorbents; 7) the optimization of the operational parameters governing the extraction of the active phytocomplexes from RfL and HtL; 8) setting up the two optimized double action laboratory technologies applied for the obtaining of hydrogel encapsulated phytocomplexes imobilised in LDH matrix; 9) the post optimality assessment of the active ingredients in vitro release from HYG-RfL-LDH and HYG-HtL-LDH depending on the pH in order to determine the appropriate amounts of composites that could be utilized in the formulation of food supplements; 10) the demonstration of the functionality of the two technologies; 11) the validation of the above mentioned technologies (TRL4). Other indirect objectives are: a) increasing the quality and diversity of food

supplements; b) decreasing the adverse side effects of concentrated phytocomplexes; c) increasing the competitiveness of the Romanian products on the international market; d) improving the scientific visibility at national and international level.

Rezultatele estimate

(în română)

Model experimental pentru tehnologia 1 HYG-RfL-LDH

Model experimental pentru tehnologia 2 HYG-HtL-LDH

Tehnologie de laborator 1 HYG-RfL-LDH

Tehnologie de laborator 2 HYG-HtL-LDH

Raport de demonstrare si documente de validare (TRL4) tehnologie 1 HYG-RfL-LDH

Raport de demonstrare si documente de validare (TRL4) tehnologie 2 HYG-HtL-LDH

4 comunicari

2 articole trimise spre publicare

2 cereri de brevet.

Rezultatele estimate

(în engleză)

Experimental model of technology 1 HYG-RfL-LDH

Experimental model of technology 2 HYG-HtL-LDH

Laboratory technology 1 HYG-RfL-LDH

Laboratory technology 2 HYG-HtL-LDH

Demonstration report and validation document (TRL4) for technology 1 HYG-RfL-LDH

Demonstration report and validation document (TRL4) for technology 2 HYG-HtL-LDH

4 communications

2 articles submitted for publication

2 applications for patent

Raport stiintific etapa 1

SURSA DE FINANȚARE – UEFISCDI Programul PNIII

Cod proiect - PN-III-P2-2.1-PED-2021-1870 –

Nr. Contract 646PED/2022

Titlu proiect:

Ecotehnologie de obținere a unor fitoingredienți încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari –

Acronim - DUACTIONMER

Consortiu:

Coordonator (CO) - Universitatea din București – Director de proiect Conf. Dr. Rodica Zăvoianu

Partener 1 (P1) - INCDPC-ICECHIM București – Responsabil proiect partener 1 - Dr. Ing. Andrei Sârbu

Partener 2 (P2) - Centrul de cercetare și prelucrare a plantelor medicinale Plantavorel SA (P2) – Responsabil proiect Partener 2 – Dr. Ing. Carmen Țebrencu

Stadiul de implementare al proiectului

**Etapă 1 -2022 CERCETARE INDUSTRIALĂ ASUPRA CONCEPTULUI FORMULAT AL
TEHNOLOGIEI 1 ȘI DEZVOLTAREA MODELULUI EXPERIMENTAL**

Durata 21 iunie 2022-31.12.2022

Finanțare etapă 1 – 160195,00 lei

Sursa 1 – buget de stat -146765,00 lei

Sursa 2 – cofinanțare – 13430,00 lei

REZUMAT RAPORT ȘTIINȚIFIC

În cadrul primei etape a proiectului PN-III-P2-2.1-PED-2021-1870-DUACTIONMER în perioada iunie-decembrie 2022 s-au efectuat o serie de activități de **cercetare industrială în vederea dezvoltării modelului experimental al ecotehnologiei 1 HYG-RfL-LDH de obținere a unor fitoingredienți din *Rhamnus Frangula L* (RfL) încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari (LDH).**

La **activitatea 1.1., P2** a efectuat **30** de experimente pentru a elabora **modelul experimental tehnologic și analitic privind modulul a₁ al ecotehnologiei de extracție a fitocomplexilor din RfL** prin 2 metode de extracție neconvenționale (a₁₁- extracție asistată de microunde (MAE) și a₁₂ – extracție asistată de ultrasunete (UAE)) și **a livrat la CO o șarjă de soluție extractivă RfL obținută prin MAE în vederea imobilizării fitocomplexului extras în diverse matrici LDH (pentru dezvoltarea modulului b₁ al ecotehnologiei) și o șarjă de material vegetal epuizat în vederea testării capacității de adsorbție a diferiților poluanți (pentru modulul d₁ al ecotehnologiei).** În vederea dezvoltării modulului **b₁ al ecotehnologiei**, CO a efectuat sinteza a **8** compuși LDH, care au fost calcinați înainte de a fi folosiți la imobilizarea fitoextractului RfL prin metoda reconstrucției și care au fost caracterizați prin difracție de raze X, DRIFT, spectroscopie DR-UV-Vis și analiză termică diferențială (TG-DTA) și a predat la P1 un număr de **16** probe LDH precursor și LDH-RfL pentru a fi utilizate la elaborarea **modulului c₁ al ecotehnologiei**. P1 a evaluat posibilitatea incorporării compușilor LDH precursori și LDH-RfL în două tipuri diferite de hidrogel efectuând **10** experimente cu hidrogel pe bază de polimer natural biocompatibil și **18** experimente cu hidrogel compozit biocompatibil în scopul **dezvoltării modulului c₁ al ecotehnologiei**.

La **activitatea 1.2., CO** a efectuat **39** experimente pentru **evaluarea tehnico-științifică și testarea capacității de adsorbție a resturilor vegetale epuizate după extracția fitocomplexilor din RfL** prin metoda MAE pentru 4 poluanți din ape reziduale: indigo carmin, Cr, Ni și Fe. Capacitatea de adsorbție a materialului vegetal epuizat a fost determinată în urma dozării prin spectrofotometrie UV-Vis

a poluanților rămași în apă la sfârșitul testelor. **Rezultatele obținute au permis elaborarea modelului experimental pentru modulul d₁ al ecotehnologiei.**

Prin urmare, la finalizarea activităților 1.1. și 1.2 s-a putut elabora integral modelul experimental pentru ecotehnologia 1 HYG-RfL-LDH urmând ca acesta să fie perfecționat în cadrul etapei următoare a proiectului.

P2 a efectuat **activitatea 1.3.** de cercetare industrială premergătoare pentru elaborarea modulului a₂ al modelului experimental al ecotehnologiei 2 **de obținere a unor fitoingredienți din *Helianthus tuberosus* L (HtL) încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari (LDH).** S-au testat 4 modalități de inactivare enzimatică a tuberculilor HtL care au fost apoi supuși procedurii de extracție a fitocomplecșilor biologic activi prin 3 metode convenționale diferite ca număr de etape (în două trepte de extracție, într-o treaptă de extracție cu prelucrarea soluției extractive și într-o singură treaptă de extracție de lungă durată). Atât solidele cât și soluțiile extractive rezultate au fost caracterizate fitochimic.

Activitatea 1.4. a constat în diseminarea rezultatelor după cum urmează:

2 articole ISI

1. I. E. Neblea, A.-M. Gavrila, T. V. Iordache, A. Zaharia, P. O. Stănescu, I. C. Radu, S. G. Burlacu, G. Neagu, A.L. Chiriac,* A. Sârbu*, Interpenetrating networks of bacterial cellulose and poly (ethylene glycol) diacrylate as potential cephalixin carriers in wound therapy, J. Polymer Research (2022) 29: 406 <https://doi.org/10.1007/s10965-022-03250-9>, (IF 3,097, zona galbena)

2. R. Zăvoianu, M. Tudorache, V.I. Pârvulescu, B. Cojocaru, O.D. Pavel, New MgFeAl-LDH Catalysts for Claisen–Schmidt Condensation, Molecules, 2022, 27, 8391. <https://doi.org/10.3390/molecules27238391>. (IF 4,972, zona galbena)

3 Comunicări orale la Conferința Nationala de Chimie ediția XXXVI, Călimănești-Caciulata Vâlcea, 4-7 oct. 2022 (autori de la CO, P1 și P2)

a) Beads based on sodium alginate and titania composites for heavy metals removal from simulated waste waters, A. Miron, A.L. Neagu, S. Dolana, A. Zaharia, A.-M. Gavrila, A. Sârbu, T.-V. Iordache, A.-L. Chiriac (P1)

b) Molecularly imprinted supermacroporous cryogels for Penicillin G adsorption, M. V. Dumitru, T.-V. Iordache, I. E. Neblea, E. B. Stoica, T. Sandu, A. Sârbu, A.-M. Gavrila, A.L. Chiriac (P1)

c) Removal of indigo carmine from wastewater by adsorption on vegetal wastes, O.D. Pavel (CO), R. Zăvoianu (CO), D. Buha, E.M. Ungureanu, A. Cruceanu (CO), A. Tîrșoagă (CO), O.T. Apreutesei (P2)

2 postere; i) A non-traditional perspective in the synthesis of Fe-LDH type materials, O. D. Pavel, R. Zăvoianu, A. Tîrșoagă, A. Cruceanu, B. Cojocaru, R. Bîrjega, V. I. Pârvulescu **la Conferința Nationala de Chimie** ediția XXXVI (autori CO) ; **ii)** Hydrogels based on natural polymers dopped with LDH and phytoextract – M. V. Dumitru, T. V. Iordache, A. L. Neagu, A. Zaharia, R. Zăvoianu, O. Pavel, S. Teodor, A. Sârbu, SICHEM 2022, Universitatea Politehnica Bucuresti, 17-18 noiembrie 2022 (autori P1 și CO)

Abstractele pentru comunicările și posterele prezentate sunt atașate la finalul acestui document.

În cadrul **activității 1.5.** trei membri din echipa CO au făcut o vizită de lucru la P2 pentru a prelua probele de extract și material vegetal uzat și a se instrui cu privire la protocoalele de lucru pentru controlul analitic al fitocomplecșilor extrași din RfL.

Pe baza celor prezentate mai sus se poate considera că toate obiectivele propuse pentru această etapă au fost atinse și chiar depășite în ceea ce privește diseminarea la care erau prevăzute doar 2 comunicări în această etapă.

Hydrogels based on natural polymers dopped with LDH and phytoextract

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Abstract: *New composite hydrogels based on Chitosan and hydrotalcite dopped with phyoextracts were prepared with the aim of possible slow release of bioactive substances.*

Key words: Chitosan, hydrogels, LDH, phytoextract

Introduction: Lately, with the rapid development of industry, hydrogels have attracted more and more attention due to their large specific surface area and unique structure. Hydrogels are three-dimensional network-structured polymer formed by a polymer main chain and a hydrophilic functional group through covalent bonds, hydrogen bonds, or physical winding cross-linking [1-2]. Hydrogels have been used for many years in controlled drug delivery, water purification (for heavy metals, dyes, antibiotics) tissue regeneration, etc. Natural polymers (chitosan alginate, biocelulose) are the most attractive materials used to replace synthetic materials [3]. In this work, new hydrogels based on chitosan (CS) were successfully prepared by adding layered double hydroxides (hydrotalcite-LDH) and LDH dopped with phytoextract during the process of CS hydrogel preparation.

Experimental and Modelling: For the development of the aimed hydrogels two polymers were needed: chitosan (CS) and poly(vinylalcohol) (PVA). Two solutions of these polymers were prepared and mixed, followed by the addition of hydrotalcites (LDH) and LDH with a phytoextract and, at the end, the crosslinker was introduced.

Results and discussions: In order to highlight the new developed hydrogels, the samples were characterized using various modern techniques such as FTIR, swelling degree. FTIR spectra confirmed the occurrence of characteristic bands for the involved raw materials.

Conclusions: The aimed hydrogels based on natural polymers were succesfully developed and characterized by FTIR and also their swelling degree capacity was studied in time.

Acknowledgement: The study was funded by the Ministry of Education and Research through Ececutive Unit for Financing Higher Education, Research, Development and Innovation (UEFISCDI), grant. No. 646 PED/2022 DUACTIONMER and grant no. 15PFE/2021 NeXT-BExcel.

References:

- [1] Wang R., Zhang X., Zhu J., Bai J., Gao L., Liuc S., Jiao T., Facile preparation of self-assembled chitosan-based composite hydrogels with enhanced adsorption performances, *Colloids and Surfaces A* 598 (2020) 124860.
- [2] Peppas N. A., Hoffman A S., Hydrogels, 153-155
- [3]. R. Jayakumar et al., Biomedical applications of chitin and chitosan-based nanomaterials—A short review, *Carbohydrate Polymers*, 82,2010, p. 227-232.

A NON-TRADITIONAL PERSPECTIVE IN THE SYNTHESIS OF Fe-LDH TYPE MATERIALS

Octavian Dumitru Pavel¹, Rodica Zăvoianu¹, Alina Tîrșoagă¹, Anca Cruceanu¹, Bogdan Cojocaru¹, Ruxandra Bîrjega², Vasile I. Pârvulescu¹

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The aim of this work was the obtaining of Fe modified layered double hydroxides (LDH) materials (Mg,Fe/Al=3 and Mg/Fe=1 molar ratio) using two non-traditional approaches in LDH synthesis: *i*) the mechano-chemical method and *ii*) the use of an organic base (tetramethylammonium hydroxide - TMAH) as precipitation agent.¹For comparison, Fe-LDH was also prepared by the traditional method: co-precipitation at pH of 10 in the presence of inorganic alkalis (NaOH/Na₂CO₃).Fe-LDH materials were subsequently calcined at 460 °C for 18 h to obtain the corresponding mixed oxides. The mixed oxides were then rehydrated with distilled water to reconstruct the layered structure of the parent LDH based on the memory effect.All materials were characterized by XRD, IR and UV-Vis spectroscopy, N₂ adsorption-desorption isotherms and basic sites determination by irreversible adsorption of organic molecules with different pK_a. Thus, a higher purity was remarkable for the compounds obtained by co-precipitation compared to those obtained by the mechano-chemical method regardless of the nature of the base.The presence of impurities in Fe-LDH obtained by both routes is related to the presence of Fe₃O₄ diffraction lines in the XRD patterns.The contaminating Fe₃O₄ phase is preserved even after the reconstruction of the layered structure. The catalytic activity in Claisen-Schmidt condensation between cyclohexanone and benzaldehyde followed the same order as the basicity of the samples: mixed oxides > reconstructed > dried, regardless the preparation method.

Acknowledgement: This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CCCDI - UEFISCDI, project number PN-III-P2-2.1-PED-2021-1870, within PNCDI III. This work was supported by a grant of the Ministry of Research, Innovation and Digitization, CNCS/CCCDI – UEFISCDI, project number PN-III-P4-ID-PCE-2020-2207, within PNCDI III.

References

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BEADS BASED ON SODIUM ALGINATE AND TITANIA COMPOSITES FOR HEAVY METALS REMOVAL FROM SIMULATED WASTE WATERS

Andreea Miron^{1,2}, Ana-Lorena Neagu¹, Sorin Dolana¹, Anamaria Zaharia¹, Ana-Mihaela Gavrilă¹, Andrei Sarbu¹, Tanta-Verona Iordache¹, Anita-Laura Chiriac^{1*}

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One of the major environmental issues is heavy metal pollution, hence new techniques must be developed to minimize the effects on the aquatic environment [1]. Sodium alginate (SA) is a natural polymer that is frequently used in wastewater treatment due to the remarkable properties in heavy metals absorption [1-2]. Titanium dioxide (TiO₂) presents good compatibility and mechanical properties, thermal stability and efficiency in removal of heavy metal [2].

Hybrid inorganic-organic composites can be used as innovative additive for beads and can act as effective sorbents for the removal of numerous pollutants, particularly for heavy metals. The most significant benefit of inorganic-organic hybrids is combining properties in a single material. An efficient and inexpensive method to obtain composites materials is host-guest polymerization.

Therefore, the aim of this study is the obtaining of sodium alginate beads based on inorganic-organic composites/or titanium oxide as reference for the removal of heavy metals from simulated waters. The composites synthesis process is very straightforward and assumes impregnation of the inorganic host into a solution containing a monomer and a polymerization initiator, after which template polymerization of the monomer in the inorganic pores is conducted. For this part of the research a TiO₂ obtained by hydrothermal process was used as an inorganic matrix and a vinyl monomer, acrylonitrile, as an organic host [3]. In order to evaluate the adsorption properties of adsorbents, modern techniques were used. Physical, morphological and textural properties were performed using X-Ray fluorescence (XRF), Brunauer-Emmett-Teller (BET) analysis and Scanning Electron Microscopy (SEM). Ultraviolet-visible spectroscopy (UV-Vis) was used to determine the metals retention over time. The effect of the parameters varied during the synthesis of beads upon the final properties were investigated through Fourier-Transform Infrared Spectroscopy (FTIR) and Thermal Gravimetric Analysis (TGA). The results showed interesting adsorption performances of the new biocomposites material.

Acknowledgement: This work was supported by a grant of the Ministry of Research, Innovation and Digitization - UEFISCDI, project number 646PED/2022 - DUACTIONMER and PN-III-Human Resources Programme-YOUNG RESEARCH TEAMS- PN-III-P1-1.1-TE-2021-0915, grant no. 135/2022 project – I-ON-MEM.

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MOLECULARLY IMPRINTED SUPERMACROPOROUS CRYOGELS FOR PENICILLIN G ADSORPTION

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Since their first appearance in the 1980s, interest in natural polymers has grown rapidly at worldwide level, especially in the medical field (tissue engineering, controlled drug delivery). Due to their outstanding properties such as high biocompatibility and low toxicity, this type of polymers can be used to obtain many types of materials for different applications¹. Unfortunately, the water pollution became a problem for humans, and not only. Higher concentrations of antibiotics were found in all water sources². Molecularly imprinted polymers (MIP) are polymeric materials based on the polymerization of a functional monomer/polymer and a cross-linking agent in the presence of a template molecule. After removing the molecule (template), an imprinted polymer with selective cavities and high affinity for the template molecule was obtained³. So, in order to solve the water pollution problem, this study was focused on using natural polymers to obtain molecularly imprinted supermacroporous cryogels for Penicillin G retention.

To develop the aimed molecularly imprinted supermacroporous cryogels, bacterial cellulose and two types of chitosan were used: commercial chitosan (CC) and laboratory prepared chitosan from commercial chitin (CCHC1). The cryogels were lyophilized and tested for their adsorption capacity towards PG.

To highlight the imprinting process and study the structure of supermacroporous cryogels, the samples were characterized through various modern techniques (FTIR, TGA, SD, UV-Vis). The characteristic bands for the involved raw materials and the obtained cryogels were confirmed through FTIR spectroscopy. The template molecule effect and the stability of the samples were studied by Thermogravimetric Analysis. The molecularly imprinted supermacroporous cryogels (**MIP1-MIP4**) were shown to exhibit greater thermal stability than their non-imprinted counterparts (**NIP1-NIP4**). The swelling behavior of the new cryogels revealed their high ability to absorb a large amount of water. Additionally, the UV-Vis results confirmed that supermacroporous cryogels were able to bind PG from aqueous solutions.

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- [3] D. Lia et al., Water Research, **42**, 2008, p. 307-317.

REMOVAL OF INDIGO CARMINE FROM WASTEWATER BY ADSORPTION ON VEGETAL WASTES

**Octavian Dumitru Pavel¹, Rodica Zăvoianu¹, Diana Buha¹, Elena Mihaela Ungureanu¹,
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Lately there is an increased trend to find bio-derived cheap adsorbents.¹ This research evaluates the adsorption capacity of vegetal wastes originating from the processing of sea-buckthorn (*Hippophaes rhamnoides*) and topinambur (*Helianthus tuberosus* L) for indigo carmine (IC) dye pollutant from wastewater. Two different vegetal waste adsorbents were recovered after the juice extraction from fresh sea-buckthorn berries: A1Hr – solid separated after the mechanical cold pressing and A2Hr – solid deposit after the centrifugation of the extracted juice at 4000 rpm for 20 min. Two other vegetal waste adsorbents were collected after inulin extraction from topinambur bulbs, i.e. A1HtL (after extraction in water under stirring) and A2HtL-US (after ultrasound extraction in water). All solids were dried at 90°C for 24 h before the adsorption tests which were performed at 25°C in brown tapered bottles under mild stirring (150 rpm) using synthetic wastewater samples having an initial concentration of IC (14 mg/L) and various adsorbent concentrations in the range of 0.1 - 5 g/L. Wastewater samples were analyzed by UV-Vis spectrometry at 1 h time intervals until 6 h and also after 24 h and 48 h. The adsorbents were characterized before and after the tests by DRIFTS and DR-UV-Vis. The correlation of the adsorption tests results and the characteristics of the solids revealed by DRIFTS and DR-UV-Vis indicated that IC adsorption depended on the aromatics content in the vegetal waste. Richer in aromatics HtL-adsorbents showed higher adsorption capacity than sea-buckthorn-derived ones, e.g. A2HtL-US (9.9 mgIC/g) > A2HtL (8.8 mgIC/g) > A2Hr (7.4 mgIC/G) > A1Hr (5.2 mgIC/g). The resulting spent adsorbents may be further used as additives in colored wrapping paper manufacture.

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FINANCING SOURCE – UEFISCDI Program PNIII

Project Code - PN-III-P2-2.1-PED-2021-1870 –

Contract No. - 646PED/2022

Project Title:

Ecotechnology for obtaining hydrogel encapsulated phytoingredients based on bioactive complexes immobilised in layered double hydroxides matrices

Acronym - DUACTIONMER

Consortium:

Coordinator (CO) – University of Bucharest – Project Manager Conf. Dr. Rodica Zăvoianu

Partner 1 (P1) - INCDPC-ICECHIM Bucharest – Project responsible for partner 1 - Dr. Ing. Andrei Sârbu

Partner 2 (P2) – Research and Processing Center for medicinal plants Plantavorel SA (P2) – Project responsible for Partner 2 – Dr. Ing. Carmen Tebrencu

The implementation status of the project:

Stage 1 -2022 INDUSTRIAL RESEARCH ON THE FORMULATED CONCEPT OF THE TECHNOLOGY 1 AND THE DEVELOPMENT OF THE EXPERIMENTAL MODEL

Period: 21 June 2022-31 December 2022

Stage 1 Financing – 160195,00 lei

Source 1 – buget -146765,00 lei

Sursa 2 – cofinancing – 13430,00 lei

SUMMARY OF THE SCIENTIFIC REPORT

In the first step of the project PN-III-P2-2.1-PED-2021-1870-DUACTIONMER, during the period June-December 2022, according to the project implementation plan, the consortium formed by the research teams of the **University of Bucharest (CO), INCDPC-ICECHIM Bucharest (P1) and the Center for research and processing of medicinal plants Plantavorel SA (P2)** carried out a series of industrial research activities to **develop the experimental model of eco-technology 1 consisting in the obtaining of phytoingredients from *Rhamnus Frangula L* (RfL) encapsulated in a hydrogel containing bioactive complexes immobilized in layered double hydroxides (LDH) matrices.**

At **activity 1.1., P2** carried out **30** experiments to develop the **experimental technological and analytical model** regarding **module a₁ of the eco-technology of extracting phytocomplexes from RfL** through 2 non-conventional extraction methods (**a₁₁- microwave-assisted extraction (MAE) and a₁₂ – extraction assisted by ultrasound (UAE)**) and delivered to CO a batch of RfL extractive solution obtained by MAE to immobilize the extracted phytocomplex in various LDH matrices (**for the development of module b₁ of the eco-technology**) and a batch of exhausted plant material to test the capacity of adsorption of different pollutants (**for module d₁ of eco-technology**). **To develop module b₁ of the eco-technology, CO** synthesised **8** LDH-type compounds, which were calcined before being used to immobilize the RfL phytoextract by the reconstruction method and further characterized by X-ray diffraction, DRIFT, DR-UV-Vis spectroscopy and differential thermal analysis (TG-DTA). **CO** provided to **P1 16** LDH-precursor materials and LDH-RfL samples to be used in developing **the c₁ module of eco-technology**. For the further development of module c₁ of the eco-technology, P1 evaluated the possibility of incorporating the LDH-precursor materials and the LDH-RfL compounds into two different types of hydrogel by performing **10** experiments with biocompatible natural polymer hydrogel and **18** experiments with biocompatible composite hydrogel.

At **activity 1.2., CO** performed **39** experiments for the technical-scientific evaluation and testing of the adsorption capacity of plant residues exhausted after the extraction of phytocomplexes from RfL by the MAE method for 4 pollutants from wastewater, namely: indigo carmine, Cr, Ni and Fe. The

adsorption capacity of the spent plant material was determined by dosing the pollutants remaining in the water at the end of the tests and analyzing them using UV-Vis spectrophotometry. The obtained results allowed the development of the experimental model for the **d₁** module of eco-technology.

Therefore, by fulfilling the tasks in activities 1.1 and 1.2, the experimental model for the eco-technology 1 HYG-RfL-LDH was fully developed and it will be further optimized in the next stage of the project implementation.

P2 has performed **activity 1.3**, which was the preliminary industrial research needed for developing module **a₂** of the experimental model of eco-technology 2 consisting in obtaining some phytoingredients from *Helianthus tuberosus* L (HtL) encapsulated in hydrogel based on bioactive complexes immobilized in the layered double hydroxide (LDH) matrices. Four ways of enzymatic inactivation of HtL tubers were tested, which were further subjected to the extraction procedure of the biologically active phytocomplexes by using 3 conventional methods having different numbers of stages (in two extraction stages, in one extraction stage followed by the analysis of the extractive solution and in one long extraction step). Both the solids and the resulting extractive solutions were phytochemically characterized.

Activity 1.4. consisted of disseminating the results as it follows

2 ISI publications

1. I. E. Neblea, A.-M. Gavrilă, T. V. Iordache, A. Zaharia, P. O. Stănescu, I. C. Radu, S. G. Burlacu, G. Neagu, A.L. Chiriac,* A. Sârbu*, Interpenetrating networks of bacterial cellulose and poly (ethylene glycol) diacrylate as potential cephalixin carriers in wound therapy, *J. Polymer Research* (2022) 29: 406 <https://doi.org/10.1007/s10965-022-03250-9>, (IF 3,097, yellow zone) authors from P1
2. R. Zăvoianu, M. Tudorache, V.I. Pârvulescu, B. Cojocaru, O.D. Pavel, New MgFeAl-LDH Catalysts for Claisen–Schmidt Condensation, *Molecules*, 2022, 27, 8391. <https://doi.org/10.3390/molecules27238391>. (IF 4,972, yellow zone) authors from CO

3 oral presentations at the XXXVI Edition of the National Conference of Chemistry Călimănești-Caciulata Vâlcea, 4-7 oct. 2022 (authors from CO, P1 and P2)

- a) Beads based on sodium alginate and titania composites for heavy metals removal from simulated waste waters, A. Miron, A.L. Neagu, S. Dolana, A. Zaharia, A.-M. Gavrilă, A. Sârbu, T.-V. Iordache, A.-L. Chiriac (P1)
- b) Molecularly imprinted supermacroporous cryogels for Penicillin G adsorption, M. V. Dumitru, T.-V. Iordache, I. E. Neblea, E. B. Stoica, T. Sandu, A. Sârbu, A.-M. Gavrilă, A.L. Chiriac (P1)
- c) Removal of indigo carmine from wastewater by adsorption on vegetal wastes, O.D. Pavel (CO), R. Zăvoianu (CO), D. Buha, E.M. Ungureanu, A. Cruceanu (CO), A. Tîrșoagă (CO), O.T. Apreutesei (P2)

2 posters; i) A non-traditional perspective in the synthesis of Fe-LDH type materials, O. D. Pavel, R. Zăvoianu, A. Tîrșoagă, A. Cruceanu, B. Cojocaru, R. Bîrjega, V. I. Pârvulescu **at the XXXVI Edition of the National Conference of Chemistry** (authors CO) ; **ii)** Hydrogels based on natural polymers dopped with LDH and phytoextract – M. V. Dumitru, T. V. Iordache, A. L. Neagu, A. Zaharia, R. Zăvoianu, O. Pavel, S. Teodor, A. Sârbu, **SICHEM 2022, University Politehnica Bucharest, 17-18 november 2022** (authors P1 and CO)

The abstracts for the above mentioned oral presentations and posters are attached at the end of this document.

Within **activity 1.5.**, three team members of CO made a work visit to P2 to collect the extract samples and the spent plant material and train on the working protocols needed for the analytical control of phytocomplexes extracted from RfL.

Based on the above information, it can be considered that all the objectives proposed for the current stage have been reached and even exceeded in terms of dissemination which targeted initially only 2 communications.

Hydrogels based on natural polymers dopped with LDH and phytoextract

Marinela V. Dumitru¹, Tanta Verona Iordache¹, Ana L. Neagu¹, Anamaria Zaharia¹, Rodica Zavoianu², Octavian Pavel², Sandu Teodor¹, Andrei Sarbu^{1*}

¹National Research-Development Institute for Chemistry and Petrochemistry-ICECHIM, Splaiul Independentei 202, district 6, 060021, Bucharest, Romania

² University of Bucharest, 050663 Bucharest, Romania

Abstract: *New composite hydrogels based on Chitosan and hydrotalcite dopped with phyoextracts were prepared with the aim of possible slow release of bioactive substances.*

Key words: Chitosan, hydrogels, LDH, phytoextract

Introduction: Lately, with the rapid development of industry, hydrogels have attracted more and more attention due to their large specific surface area and unique structure. Hydrogels are three-dimensional network-structured polymer formed by a polymer main chain and a hydrophilic functional group through covalent bonds, hydrogen bonds, or physical winding cross-linking [1-2]. Hydrogels have been used for many years in controlled drug delivery, water purification (for heavy metals, dyes, antibiotics) tissue regeneration, etc. Natural polymers (chitosan alginate, biocelulose) are the most attractive materials used to replace synthetic materials [3]. In this work, new hydrogels based on chitosan (CS) were successfully prepared by adding layered double hydroxides (hydrotalcite-LDH) and LDH dopped with phytoextract during the process of CS hydrogel preparation.

Experimental and Modelling: For the development of the aimed hydrogels two polymers were needed: chitosan (CS) and poly(vinylalcohol) (PVA). Two solutions of these polymers were prepared and mixed, followed by the addition of hydrotalcites (LDH) and LDH with a phytoextract and, at the end, the crosslinker was introduced.

Results and discussions: In order to highlight the new developed hydrogels, the samples were characterized using various modern techniques such as FTIR, swelling degree. FTIR spectra confirmed the occurrence of characteristic bands for the involved raw materials.

Conclusions: The aimed hydrogels based on natural polymers were succesfully developed and characterized by FTIR and also their swelling degree capacity was studied in time.

Acknowledgement: The study was funded by the Ministry of Education and Research through Ececutive Unit for Financing Higher Education, Research, Development and Innovation (UEFISCDI), grant. No. 646 PED/2022 DUACTIONMER and grant no. 15PFE/2021 NeXT-BExcel.

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A NON-TRADITIONAL PERSPECTIVE IN THE SYNTHESIS OF Fe-LDH TYPE MATERIALS

Octavian Dumitru Pavel¹, Rodica Zăvoianu¹, Alina Tîrșoagă¹, Anca Cruceanu¹, Bogdan Cojocaru¹, Ruxandra Bîrjega², Vasile I. Pârvulescu¹

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The aim of this work was the obtaining of Fe modified layered double hydroxides (LDH) materials (Mg,Fe/Al=3 and Mg/Fe=1 molar ratio) using two non-traditional approaches in LDH synthesis: *i*) the mechano-chemical method and *ii*) the use of an organic base (tetramethylammonium hydroxide - TMAH) as precipitation agent.¹For comparison, Fe-LDH was also prepared by the traditional method: co-precipitation at pH of 10 in the presence of inorganic alkalis (NaOH/Na₂CO₃).Fe-LDH materials were subsequently calcined at 460 °C for 18 h to obtain the corresponding mixed oxides. The mixed oxides were then rehydrated with distilled water to reconstruct the layered structure of the parent LDH based on the memory effect. All materials were characterized by XRD, IR and UV-Vis spectroscopy, N₂ adsorption-desorption isotherms and basic sites determination by irreversible adsorption of organic molecules with different pK_a. Thus, a higher purity was remarkable for the compounds obtained by co-precipitation compared to those obtained by the mechano-chemical method regardless of the nature of the base. The presence of impurities in Fe-LDH obtained by both routes is related to the presence of Fe₃O₄ diffraction lines in the XRD patterns. The contaminating Fe₃O₄ phase is preserved even after the reconstruction of the layered structure. The catalytic activity in Claisen-Schmidt condensation between cyclohexanone and benzaldehyde followed the same order as the basicity of the samples: mixed oxides > reconstructed > dried, regardless the preparation method.

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BEADS BASED ON SODIUM ALGINATE AND TITANIA COMPOSITES FOR HEAVY METALS REMOVAL FROM SIMULATED WASTE WATERS

Andreea Miron^{1,2}, Ana-Lorena Neagu¹, Sorin Dolana¹, Anamaria Zaharia¹, Ana-Mihaela Gavrila¹, Andrei Sarbu¹, Tanta-Verona Iordache¹, Anita-Laura Chiriac^{1*}

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SURSA DE FINANȚARE – UEFISCDI Programul PNIII**Cod proiect - PN-III-P2-2.1-PED-2021-1870 –****Nr. Contract 646PED/2022****Titlu proiect:*****Ecotehnologie de obținere a unor fitoingredienți încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari –*****Acronim - DUACTIVMER****Consortiu:****Coordonator (CO) - Universitatea din București –** Director de proiect Conf. Dr. Rodica Zăvoianu**Partener 1 (P1) - INCDPC-ICECHIM București –** Responsabil proiect partener 1 - Dr. Ing. Andrei Sârbu**Partener 2 (P2) - Centrul de cercetare și prelucrare a plantelor medicinale Plantavorel SA (P2) –** Responsabil proiect Partener 2 – Dr. Ing. Carmen Țebrencu**Stadiul de implementare al proiectului****ETAPA 2/2023 – ELABORAREA ECOTEHNOLOGIILOR DE LABORATOR INTEGRATE
HYG-RfL-LDH ȘI HYG-HtL-LDH****Durata 3 Ianuarie 2023-29.12.2023****Finanțare etapa 2 – 391626,00lei****Sursa 1 – buget de stat -345176,00 lei****Sursa 2 – cofinanțare – 46450,00 lei****REZUMAT RAPORT ȘTIINȚIFIC****ETAPA 2/2023 – ELABORAREA ECOTEHNOLOGIILOR DE LABORATOR INTEGRATE
HYG-RfL-LDH ȘI HYG-HtL-LDH**

În cadrul etapei 2 a proiectului PN-III-P2-2.1-PED-2021-1870-DUACTIVMER în perioada ianuarie-decembrie 2023 consorțiul format din echipele de cercetători **Universitatea din București (CO), INCDPC-ICECHIM București (P1) și Centrul de cercetare și prelucrare a plantelor medicinale Plantavorel SA (P2)**, conform planului de realizare al proiectului, a efectuat o serie de activități de **cercetare industrială în vederea: dezvoltării modelului experimental al ecotehnologiei HYG-HtL-LDH de obținerea unor fitoingredienți din *Helianthus tuberosus L* (HtL) încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari (LDH) (activitățile 2.1, 2.2, 2.3); elaborarea tehnologiei de laborator HYG-RfL-LDH pentru obținerea unor fitoingredienți din *Rhamnus frangula L* (RfL) încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari (LDH) (activitatea 2.4) și elaborarea tehnologiei de laborator HYG-HtL-LDH (activitatea 2.5). Partenerul P2 a efectuat activitățile de cercetare industrială 2.6 pentru caracterizarea fitochimică completă și stabilirea caracteristicilor extractelor HtL și 2.7 pentru verificarea funcționalității modulului (a₁) al tehnologiei de extracție a fitocomplecșilor din RfL și stabilirea caracteristicilor acestora și activitățile de dezvoltare experimentală 2.8 pentru demonstrarea și validarea modulului (a₁) al tehnologiei HYG-RfL-LDH și activitatea 2.9. pentru demonstrarea și validarea modulului (a₂) al tehnologiei HYG-HtL-LDH.**

Pentru a elabora **modelul experimental al ecotehnologiei HYG-HtL-LDH** cercetările privind **modulul a₂** al ecotehnologiei în vederea extracției fitocomplecșilor din HtL au fost efectuate de **P2** care a evaluat 5 surse de materie primă vegetală dintre care a selectat materialul cu concentrația cea mai mare de inulină și a efectuat 30 de experimente de extracție prin 2 metode neconvenționale (extracție asistată de ultrasunete (UAE) și extracție asistată microunde de (MAE)). În urma testelor s-a selectat soluția extractivă (SE) cea mai bogată în inulină (SE-MAE) care a fost livrată la CO în vederea imobilizării

inulinei în diverse matrici LDH (pentru dezvoltarea **modulului b₂** al ecotehnologiei) împreună cu două șarje de material vegetal epuizat (A1HTL-US, A2-HTL-MAE) (pentru **modulul d₂** al ecotehnologiei). Pentru **modulul b₂** al ecotehnologiei, **CO** a sintetizat **6** compuși LDH în care a incorporat SE-MAE prin metoda reconstrucției – probele R2-I, R3-I, R4-I și prin metoda coprecipitării probele P2-I, P3-I, P4-I. Solidele au fost caracterizate prin DRX, DRIFT, spectroscopie DR-UV-Vis și analiză termică diferențială (TG-DTA) și a efectuat teste de eliberare la pH-controlat a inulinei din solid. S-au a predat la P1 un număr de **9** probe LDH precursor (H2,H3,H4) și cele 6 probe cu inulină pentru a fi utilizate la elaborarea modulului c₂ al ecotehnologiei. **Modulul c₂ al ecotehnologiei.** a fost elaborat de **P1** care a evaluat posibilitatea incorporării compușilor LDH precursori și LDH-HtL în două tipuri diferite de hidrogel efectuând **9** experimente cu hidrogel pe bază de polimer natural biocompatibil și **9** experimente cu hidrogeluri compozite semi-interpenetrante. După analizarea compozitelor prin determinarea gradelor de gonflare, comportării reologice și SEM, s-au selectat 6 probe care au fost transmise la CO pentru testele de eliberare la pH-controlat a inulinei din solid. **Modulul d₂** al tehnologiei a fost realizat de **CO** care a efectuat **54** experimente pentru determinarea capacității de adsorbție a resturilor vegetale epuizate A1HTL-US (15 teste), A2-HTL-MAE (39 teste) pentru 5 poluanți din ape reziduale: indigo carmin (IC), Cr, Cu, Fe și Ni.. S-a constatat capacitatea de adsorbție mai mare a A2-HTL-MAE pentru IC și selectivitatea acestuia pentru adsorbția Fe.

Elaborarea tehnologiei de laborator **HYG-RfL-LDH** s-a efectuat pe baza modelului experimental propus în etapa 1, și a experimentelor suplimentare efectuate de CO pentru eliberarea controlată a fitocomplexului *RfL* din compușii *RfL*-LDH și hidrogelurile corespunzătoare acestora și a testelor suplimentare pentru adsorbția Cu din apele reziduale pe materialul vegetal epuizat. Tehnologia cuprinde **4 module: (a₁)** extracția complexilor bioactivi din *Rhamnus Frangula L (RfL)* realizat de P2, **(b₁)** – imobilizarea complexilor bioactivi din *RfL* în matrici LDH realizat de **CO**; **(c₁)** încapsularea în hidrogel a complexilor bioactivi din *RfL* imobilizați în matrici LDH realizat de **P1**; **(d₁)** valorificarea materialului vegetal epuizat după extracția compușilor bioactivi din *RfL* ca material adsorbant pentru poluanți din ape. P2 a demonstrat și validat modulul (a₁) cu ajutorul metodei de validare prospectivă efectuând o serie de 3 experimente.

Tehnologia de laborator **HYG-HtL-LDH** s-a dezvoltat pe baza modelului experimental descris mai sus. Demonstrarea și validarea modulului a₂ al tehnologiei a fost realizată de P2 conform metodei aplicate și la tehnologia **HYG-RfL-LDH**

Activitatea 2.10. Diseminarea s-a materializat în:

participări cu 4 prezentări orale, 1 conferință invitată și 3 postere la conferințe:

- i) 18th INTERNATIONAL CONFERENCE ON CHEMISTRY AND THE ENVIRONMENT (ICCE 2023) Venice Italy 11 -16 June 2023 R. Zăvoianu, A.-E. Stamate, O.D. Pavel, A. Cruceanu, R. Bîrjega, I.A. Brezeștean, A. Ciorîță, Efficient Removal of AB74 Dye from Wastewater Using Hybrid Catalysts: Comparative Studies on Ce-LDH-GO and Mo-LDH-GO Composites Prezentare orală (OP107)
- ii) 15th European Congress on Catalysis Prague Czech Republic 27 August-1 September 2023 O.D. Pavel, B.-C. Jurca, R. Zăvoianu, R. Bîrjega, B. Cojocar, V.I. Pârvulescu, Organic alkalis as an alternative for eco-friendly mechano-chemical synthesis of Layered Double Hydroxides-type catalysts
- iii) International Conference „Clean Nature For Health“ 10 - 13 September 2023, Velingrad, Bulgaria A. Sârbu, T. V. Iordache, A. M. Gavrilă, E. B. Stoica, B. Tsyntsarski, S. Teodor, G. Georgiev, I. Stoycheva, Molecularly Imprinted Polymers Preparation and their Use for Environment and Health- Own Result, invited lecture
- iv) The Days of Technical Sciences Academy of Romania, Brasov, Romania, 5-6 October 2023; A. Sârbu, S. Teodor, A. L. Chiriac, T. V. Iordache, F. Yardim, A. Sirkecioglu, N. Petrov, B. Tsintsarski, R. Zăvoianu, Polymer bioactive membrane for the treatment of waste waters from hotels. Prezentare orală
- v) PRIORITIES OF CHEMISTRY FOR A SUSTAINABLE DEVELOPMENT PRIOCHEM-XIXth Edition, 11-13 octombrie 2023; A.-Lorena Neagu, A. Zaharia, M. V. Dumitru, I. E. Neblea, S. Dolana, T.V. Iordache, C. E. Țebrencu, R. Zăvoianu, O. D. Pavel, A. Țișoagă, A. Sârbu, Drug-Release System

Based on Biocompatible Hydrogel Charged With Layered Double Hydroxide Containing Encapsulated *Rhamnus Frangula* L. Phytoextract, prezentare orală

vi). 3rd International Conference on Bioengineering and Polymer Science, Bucharest, June 7-11, 2023 ROMANIA; A. Zaharia, A. L. Neagu, O. D. Pavel, A. Tîrșoagă, I. E. Neblea, S. V. Dolana, T. V. Iordache, R. Zăvoianu, A. Sârbu, Layered Double Hydroxides - Polyethylene Glycol Diacrylate Composite Hydrogels for the Controlled Release of *Rhamnus Frangula* L Phytoextract- poster

vii) 18th INTERNATIONAL CONFERENCE ON CHEMISTRY AND THE ENVIRONMENT (ICCE 2023) Venice Italy 11 -16 June 2023 A. Tîrșoagă, R. Zăvoianu, O.D. Pavel, A. Cruceanu, O.T. Apreutesei, C. Tebrencu, Using lignocellulosic vegetal wastes as adsorbents for transition metals from wastewaters - poster

viii) 5th Euro-Mediterranean Conference for Environmental Integration (EMCEI-2023), 2-5 octombrie 2023, A. Zaharia, I. E. Neblea, A.-L. Chiriac, A.-M. Gavrilă, S. Dolana, A. L. Neagu, B. Trică, A. Sârbu, F.-X. Perrin, T.-V. Iordache, Polyethylene glycol diacrylate nanogels - a versatile system for the molecularly imprinting of biomolecules – poster

Publicarea unui articol într-o revistă ISI (Q1): A.-L. Neagu, A. Zaharia, O. D. Pavel, A. Tîrșoagă, I. E. Neblea, S. V. Dolana, C. E. Tebrencu, T.-V. Iordache, A. Sârbu, R. Zăvoianu, Synergistic Sustained Drug Release System based on immobilized *Rhamnus Frangula* L phytoextract into Layered Double Hydroxide covered by Biocompatible Hydrogel, *Pharmaceutics*, 2023, 15(7), 1888; <https://doi.org/10.3390/pharmaceutics15071888>.

Publicarea unui articol într-o revistă ISI (Q2): O.D. Pavel, A.E. Stamate, R. Zăvoianu, A. Cruceanu, A. Jurca, R. Birjega, I.A. Brezestean, A. Ciorîță, D.C. Culiță, A.P.S. Dias, Mo-LDH-GO Hybrid Catalysts for Indigo Carmine Advanced oxidation, *Materials*, 2023, 4, 3205, doi: 10.3390/ma16083025

Depunerea a două cereri de brevet la OSIM:

1. Sârbu A., Zăvoianu R., Pavel O. D., Zaharia A., Neagu A. L., Jurca A., Tebrencu C., Apreutesei O. T., Dumitru M. V., Sandu T., Iordache T. V., Hidrogeluri compozite biocompatibile, conținând hidroxizidublu lamelari, pentru eliberarea controlată a substanțelor bioactive din fitoextracte de crușin și procedeu de obținere, A 2023 -00108/07.03.2023.

2. Sârbu A., Zăvoianu R., Zaharia A., Pavel O. D., Dumitru M. V., Sandu T., Jurca A., Tebrencu C. E., Stamate A. E., Iordache T. V., Chiriac A. L., Gavrilă A.-M., Hidrogeluri cu rețele semi-interpenetrate, conținând hidroxizi dubli lamelari, pentru eliberarea controlată de inulină și procedeu de obținere.

În cadrul **activității 2.11**, trei membri din echipa CO au făcut o vizită de lucru la P2 pentru discutarea rezultatelor obținute la demonstrarea și validarea modulului (a_1) al tehnologiei HYG-RfL-LDH și la demonstrarea și validarea modulului (a_2) al tehnologiei HYG-HtL-LDH.

Pe baza celor prezentate mai sus se poate considera că obiectivele propuse pentru această etapă au fost atinse și chiar depășite în ceea ce privește diseminarea.

In English

FINANCING SOURCE – UEFISCDI Program PNIII

Project Code - PN-III-P2-2.1-PED-2021-1870 –

Contract No. - 646PED/2022

Project Title:

Ecotechnology for obtaining hydrogel encapsulated phytoingredients based on bioactive complexes immobilised in layered double hydroxides matrices

Acronym - DUACTIONMER

Consortium:

Coordinator (CO) – University of Bucharest – Project Manager Conf. Dr. Rodica Zăvoianu

Partner 1 (P1) - INCDPC-ICECHIM Bucharest – Project responsible for partner 1 - Dr. Ing. Andrei Sârbu

Partner 2 (P2) – Research and Processing Center for Medicinal Plants Plantavorel SA (P2) – Project responsible for Partner 2 – Dr. Ing. Carmen Țebrencu

The implementation status of the project:

Stage 2 -2023 SETTING UP OF THE INTEGRATED LABORATORY ECOTECHNOLOGIES HYG-RfL-LDH AND HYG-HtL-LDH

Period: 1 January 2023-31 December 2023

Stage 2 Financing – 391626,00 lei

Source 2 – budget -345176,00 lei

Sursa 2 – cofinancing – 46450,00 lei

SUMMARY OF THE SCIENTIFIC REPORT

STAGE 2/2023 – SETTING UP OF THE INTEGRATED LABORATORY ECOTECHNOLOGIES HYG-RfL-LDH AND HYG-HtL-LDH

During the second stage of the project PN-III-P2-2.1-PED-2021-1870-DUACTIONMER from January to December 2023 the consortium made by joining the research teams from **University of Bucharest (CO), INCDPC-ICECHIM Bucharest (P1) and the Research and Processing Center for Medicinal Plants SA (P2)**, according to the realisation plan of the project, performed a series of **industrial research activities** aimed to: set up the **experimental model of the ecotechnology HYG-HtL-LDH** for the obtaining of hydrogel encapsulated phytoingredients from *Helianthus tuberosus L* (HtL) based on bioactive complexes immobilised in layered double hydroxides matrices (LDH) (activities **2.1, 2.2, 2.3**); **the development of the laboratory technology HYG-RfL-LDH for the obtaining** of hydrogel encapsulated phytoingredients from *Rhamnus frangula L* (RfL) based on bioactive complexes immobilised in layered double hydroxides (LDH) (activity **2.4**) **and the development of the laboratory technology HYG-HtL-LDH (activity 2.5)**. **Partner P2** performed the **industrial research activities 2.6** for the full phytochemical characterization and the setting up of the HtL extracts characteristics and **2.7** for the **functionality checking-up of the module (a₁)** of the technology for the extraction of RfL phytocomplexes and the setting up of their characteristics as well as the **experimental development activities 2.8** to **demonstrate and validate the module (a₁) of the technology HYG-RfL-LDH** and the activity **2.9** to **demonstrate and validate the module (a₂) of the technology HYG-HtL-LDH**.

In order to set up the **experimental model of the ecotechnology HYG-HtL-LDH**, the researches related to the **module a₂** of the ecotechnology targeting the extraction of the phytocomplexes from HtL were performed by **P2** who evaluated 5 sources of vegetal raw material in order to select the one with the highest concentration of inulin and carried out **30** extraction tests using 2 nonconventional methods (ultrasound assisted extraction (UAE) and microwave assisted extraction (MAE)). Following the tests, the extractive solution (SE) having the highest concentration of inulin (SE-MAE) was selected and

delivered to CO in order to be immobilised in different LDH matrices (for setting up the **module b₂** of the ecotechnology) along with two batches of vegetal spent material (A1HTL-US, A2-HTL-MAE) (to be utilized in the **module d₂** of the ecotechnology). For the **module b₂** of the ecotechnology, CO synthesized 6 LDH compounds into which SE-MAE was incorporated by reconstruction– samples R2-I, R3-I, R4-I and by co-precipitation samples P2-I, P3-I, P4-I. The resulting solids were characterized by XRD, DRIFT, ATR-FTIR, DR-UV-Vis spectroscopy, thermodifferential analysis (TG-DTA) and inulin release tests under controlled pH. CO delivered to P1 a number of 9 LDH samples, 3 samples of reference LDH precursors (H2,H3,H4) and the six samples containing inulin in order to be utilized for setting up the module c₂ of the ecotechnology. The **module c₂ of the ecotechnology** was set up by P1 who evaluated the possibility to incorporate of the LDH precursors and LDH-HtL compounds in two different types of hydrogels by performing 9 experiments with a hydrogel based on a natural biocompatible polymer and 9 experiments with semi-interpenetrated composite hydrogels. After analysing the composites by determining the swelling degree, the rheological behaviour and SEM, 6 samples were selected and delivered to CO for the inulin release tests under controlled pH. The **module d₂** of the technology was set up by CO who performed 54 experiments to determine the adsorption capacity of the spent vegetal materials A1HTL-US (15 tests), A2-HTL-MAE (39 tests) for 5 pollutants from waste waters: indigo carmin (IC), Cr, Cu, Fe and Ni. The obtained results indicated a higher adsorption capacity of A2-HTL-MAE for IC and its selectivity for Fe adsorption.

The setting up of the laboratory technology **HYG-RfL-LDH** was based on the experimental model proposed in the first stage of the project and the supplementary experiments performed by CO for the controlled release of RfL phytocomplex from RfL-LDH compounds and the corresponding hydrogels as well as the supplementary tests for Cu adsorption from waste water onto the spent vegetal material. The technology consists of 4 modules: (a₁) the extraction of bioactive complexes from *Rhamnus Frangula L* (RfL) realised by P2, (b₁) – immobilization of bioactive complexes from RfL into LDH matrices realized by CO; (c₁) hydrogel encapsulation of the bioactive complexes from RfL immobilized into LDH matrices LDH realized by P1; (d₁) valorisation of the spent vegetal material after the extraction of the bioactive compounds from RfL as adsorbant for water pollutants. P2 demonstrated and validated the module (a₁) using the prospective validation method by performing a series of 3 experiments.

The laboratory technology **HYG-HtL-LDH** was developed based on the above described experimental model. The demonstration and validation for the module a₂ of the technology was realized by P2 according to the same method applied for the technology **HYG-RfL-LDH**.

Activity 2.10. The dissemination consisted in:

participations with 4 oral presentations, 1 invited lecture and 3 poster presentations at conferences:

- i) 18th INTERNATIONAL CONFERENCE ON CHEMISTRY AND THE ENVIRONMENT (ICCE 2023) Venice Italy 11 -16 June 2023 R. Zăvoianu, A.-E. Stamate, O.D. Pavel, A. Cruceanu, R. Bîrjega, I.A. Brezeştean, A. Ciorîţă, Efficient Removal of AB74 Dye from Wastewater Using Hybrid Catalysts: Comparative Studies on Ce-LDH-GO and Mo-LDH-GO Composites Oral Presentation (OP107)
- ii) 15th European Congress on Catalysis Prague Czech Republic 27 August-1 September 2023 O.D. Pavel, B.-C. Jurca, R. Zăvoianu, R. Bîrjega, B. Cojocaru, V.I. Pârvulescu, Organic alkalis as an alternative for eco-friendly mechano-chemical synthesis of Layered Double Hydroxides-type catalysts
- iii) International Conference „Clean Nature For Health“ 10 - 13 September 2023, Velingrad, Bulgaria A. Sârbu, T. V. Iordache, A. M. Gavrilă, E. B. Stoica, B. Tsyntsarski, S. Teodor, G. Georgiev, I. Stoycheva, Molecularly Imprinted Polymers Preparation and their Use for Environment and Health- Own Result, invited lecture
- iv) The Days of Technical Sciences Academy of Romania, Brasov, Romania, 5-6 October 2023; A. Sârbu, S. Teodor, A. L. Chiriac, T. V. Iordache, F. Yardim, A. Sirkecioglu, N. Petrov, B. Tsyntsarski, R. Zăvoianu, Polymer bioactive membrane for the treatment of waste waters from hotels. Oral Presentation
- v) PRIORITIES OF CHEMISTRY FOR A SUSTAINABLE DEVELOPMENT PRIOCHEM-XIXth Edition, 11-13 octombrie 2023; A.-Lorena Neagu, A. Zaharia, M. V. Dumitru, I. E. Neblea, S. Dolana, T.V. Iordache, C. E. Ţebrencu, R. Zăvoianu, O. D. Pavel, A. Tîrşoagă, A. Sârbu, Drug-Release System

Based on Biocompatible Hydrogel Charged With Layered Double Hydroxide Containing Encapsulated *Rhamnus Frangula* L. Phytoextract, Oral Presentation

vi). 3rd International Conference on Bioengineering and Polymer Science, Bucharest, June 7-11, 2023 ROMANIA; A. Zaharia, A. L. Neagu, O. D. Pavel, A. Tîrșoagă, I. E. Neblea, S. V. Dolana, T. V. Iordache, R. Zăvoianu, A. Sârbu, Layered Double Hydroxides - Polyethylene Glycol Diacrylate Composite Hydrogels for the Controlled Release of *Rhamnus Frangula* L Phytoextract- poster

vii) 18th INTERNATIONAL CONFERENCE ON CHEMISTRY AND THE ENVIRONMENT (ICCE 2023) Venice Italy 11 -16 June 2023 A. Tîrșoagă, R. Zăvoianu, O.D. Pavel, A. Cruceanu, O.T. Apreutesei, C. Tebrencu, Using lignocellulosic vegetal wastes as adsorbents for transition metals from wastewaters - poster

viii) 5th Euro-Mediterranean Conference for Environmental Integration (EMCEI-2023), 2-5 octombrie 2023, A. Zaharia, I. E. Neblea, A.-L. Chiriac, A.-M. Gavrilă, S. Dolana, A. L. Neagu, B. Trică, A. Sârbu, F.-X. Perrin, T.-V. Iordache, Polyethylene glycol diacrylate nanogels - a versatile system for the molecularly imprinting of biomolecules – poster

Publication of a paper in ISI journal (Q1): A.-L. Neagu, A. Zaharia, O. D. Pavel, A. Tîrșoagă, I. E. Neblea, S. V. Dolana, C. E. Tebrencu, T.-V. Iordache, A. Sârbu, R. Zăvoianu, Synergistic Sustained Drug Release System based on immobilized *Rhamnus Frangula* L phytoextract into Layered Double Hydroxide covered by Biocompatible Hydrogel, *Pharmaceutics*, 2023, 15(7), 1888; <https://doi.org/10.3390/pharmaceutics15071888>.

Publication of a paper in ISI journal (Q2): O.D. Pavel, A.E. Stamate, R. Zăvoianu, A. Cruceanu, A. Jurca, R. Birjega, I.A. Brezestean, A. Ciorîță, D.C. Culiță, A.P.S. Dias, Mo-LDH-GO Hybrid Catalysts for Indigo Carmine Advanced oxidation, *Materials*, 2023, 4, 3205, doi: 10.3390/ma16083205

2 Applications for national patents at OSIM:

1. Sârbu A., Zăvoianu R., Pavel O. D., Zaharia A., Neagu A. L., Jurca A., Tebrencu C., Apreutesei O. T., Dumitru M. V., Sandu T., Iordache T. V., Biocompatible composite hydrogels containing layered double hydroxides, for the controlled release of the bioactive substances from alder buckthorn phytoextracts and obtaining procedure, A 2023 -00108/07.03.2023.

2. Sârbu A., Zăvoianu R., Zaharia A., Pavel O. D., Dumitru M. V., Sandu T., Jurca A., Tebrencu C. E., Stamate A. E., Iordache T. V., Chiriac A. L., Gavrilă A.-M., Hidrogels with semi-interpenetrated networks containing layered double hydroxides for the controlled release of inulin and obtaining procedure. A 2023 -00795/05.12.2023.

Within **activity 2.11.**, three team members of CO made a work visit to P2 to discuss the results obtained at the demonstration and validation of the module (a₁) of the technology HYG-RfL-LDH and the demonstration and validation of the module (a₂) of the technology HYG-HtL-LDH.

Based on the above information, it can be considered that all the objectives proposed for the current stage have been reached and even exceeded in terms of dissemination which targeted initially only 2 communications and 1 paper submitted for publication.

SURSA DE FINANȚARE – UEFISCDI Programul PNIII**Cod proiect - PN-III-P2-2.1-PED-2021-1870 –****Nr. Contract 646PED/2022****Titlu proiect:*****Ecotehnologie de obținere a unor fitoingredienți încapsulați în hidrogel pe baza de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari –*****Acronim - DUACTIVMER****Consortiu:****Coordonator (CO) - Universitatea din București –** Director de proiect Conf. Dr. Rodica Zăvoianu**Partener 1 (P1) - INCDPC-ICECHIM București –** Responsabil proiect partener 1 - Dr. Ing. Andrei Sârbu**Partener 2 (P2) - Centrul de cercetare și prelucrare a plantelor medicinale Plantavorel SA (P2) –****Responsabil proiect Partener 2 –** Dr. Ing. Carmen Țebrencu**Stadiul de implementare al proiectului****ETAPA 3/2024 – ELABORAREA ECOTEHNOLOGIILOR DE LABORATOR INTEGRATE HYG-RfL-LDH ȘI HYG-HtL-LDH****Durata 3 Ianuarie 2024-20.06.2024****Finanțare etapa 3 – 106854,00 lei****Sursa 1 – buget de stat -106854,00 lei****Sursa 2 – cofinanțare – 0,00 lei****REZUMAT RAPORT ȘTIINȚIFIC****ETAPA 3/2024 – ELABORAREA ECOTEHNOLOGIILOR DE LABORATOR INTEGRATE HYG-RfL-LDH ȘI HYG-HtL-LDH**

În cadrul etapei 3 a proiectului **PN-III-P2-2.1-PED-2021-1870-DUACTIVMER** în perioada ianuarie-20 iunie 2024 consorțiul format din echipele de cercetători **Universitatea din București (CO)**, **INCDPC-ICECHIM București (P1)** conform planului de realizare al proiectului, a efectuat **patru** activități de **dezvoltare experimentală** după cum urmează: **activitatea 3.1** privind **demonstrarea funcționalității ecotehnologiei HYG-RfL-LDH**, pentru care **P1** a caracterizat extensiv compozitele HYG-RfL-LDH (activitatea 3.1.1.) și **CO** a testat eliberarea ingredientelor active din HYG-RfL-LDH și eficiența adsorbanților resturi vegetale rămase după extracția fitocomplecșilor din *RfL* în eliminarea poluanților din ape reziduale în procese de adsorbție în flux pentru modulul d_1 al tehnologiei (activitatea 3.1.2.); **activitatea 3.2.** pentru **demonstrarea funcționalității ecotehnologiei HYG-HtL-LDH** în cadrul căreia **P1** a caracterizat materialele (activitatea 3.2.1) și **CO** a verificat comportarea materialelor în testele de eliberare in vitro a fitocomplexului HtL și a testat eficiența adsorbanților resturi vegetale rămase după extracția fitocomplecșilor din *HtL* în eliminarea poluanților din ape reziduale în procese de adsorbție în flux pentru optimizarea modulului d_2 al tehnologiei (activitatea 3.2.2.); **activitatea 3.3.** pentru **validarea ecotehnologiei HYG-RfL-LDH** în care CO s-a ocupat de modulul (b_1) – imobilizarea fitoextractului RfL în matrice LDH (activitatea 3.3.1) iar **P1** a validat modulul (c_1) privind încapsularea *RfL*-LDH în hidrogel PEGDA (activitatea 3.3.2) și **activitatea 3.4.** care a constat în validarea ecotehnologiei HYG-HtL-LDH pe baza rezultatelor obținute de CO la imobilizarea fitoextractului *HtL* în matrice LDH pentru modulul (b_2) al tehnologiei (activitatea 3.4.1) și prin validarea modulului (c_2) privind încapsularea *HtL*-LDH în hidrogel PEGDA-chitosan (activitatea 3.4.2).

În cadrul activităților 3.1.1 și 3.2.1, P1 a caracterizat extensiv materialele folosind tehnicile de caracterizarea ATR-FTIR, DR-Uv-vis, BET, analiza TGA, SEM și XRD și a dovedit prin rezultatele obținute că s-a reușit încapsularea fitoingredienților extrași din *RfL* (act. 3.1.1) și respectiv *HtL* (act. 3.2.1) imobilizați în matrice LDH. Prin efectuarea testelor de eliberare controlată a compușilor fitoactivi din hidrogelurile compozite ce conțin *RfL* (activitatea 3.1.2) și respectiv *HtL* (activitatea 3.2.2), CO a dovedit eficiența încapsulării în hidrogel în prepararea unui sistem eficient de eliberare controlată, la pH-ul intestinal a fitocomplecșilor respectivi. În testele realizate la adsorbția în flux a poluanților din ape reziduale sintetice folosind ca adsorbanți resturile vegetale recuperate după extracția fitocomplecșilor *RfL* (activitatea 3.1.2.) și respectiv *HtL* (activitatea 3.2.2), CO a dovedit că în ambele cazuri îndepărtarea poluanților are loc în proporție de peste 90%, conducând la concentrații admisibile de poluant în apa tratată pe durata a

maxim 3 cicluri adsorbție/regenerare. În cazul resturilor vegetale rămase după extracția *RfL* se pot îndepărta cu succes Cr^{6+} , Ni^{2+} și $\text{Fe}^{2+}, \text{Fe}^{3+}$ în timp ce în cazul resturilor vegetale recuperate după extracția *HtL* se pot îndepărta Cu^{2+} , Ni^{2+} și $\text{Fe}^{2+}, \text{Fe}^{3+}$.

Tehnologia **HYG-RfL-LDH** validată la activitatea 3.3. are un dublu efect: i) permite prepararea unui sistem compozit pe bază de hidrogel PEGDA biocompatibil în care este înglobat extract *RfL* imobilizat în matrice LDH care este eficient în eliberarea controlată susținută, la pH-ul intestinal putând fi util în tratamentul tranzitului intestinal lent și ii) permite tratarea apelor contaminate cu Cr^{6+} , Ni^{2+} și $\text{Fe}^{2+}, \text{Fe}^{3+}$ prin valorificarea resturilor vegetale rămase după extracția fitocomplexului *RfL* din scoarța de crușin.

Tehnologia **HYG-HtL-LDH** validată la activitatea 3.4. are un dublu efect: i) permite prepararea unui sistem compozit pe bază de hidrogel PEGDA-Chitosan biocompatibil în care este înglobat extract *HtL* imobilizat în matrice LDH care este eficient în eliberarea controlată susținută, la pH-ul intestinal putând fi util ca prebiotic și ii) permite tratarea apelor contaminate cu Cu^{2+} , Ni^{2+} și $\text{Fe}^{2+}, \text{Fe}^{3+}$ prin valorificarea resturilor vegetale rămase după extracția fitocomplexului *HtL* din topinambur.

Activitatea 3.5. a constatat în diseminarea rezultatelor s-a materializat prin:

Participarea cu o conferință plenară și 2 postere la International Chemical Engineering and Material Symposium, SICHEM 202411-12 April 2024, ISSN 2537-2254

1. Advanced ceramic powders obtained via host guest polymerization **A. Sârbu**, A. L. Chiriac, T. Sandu, A. Miron, L. Mara, A. Dumitru, C. Lazau, T. V. Iordache – conferință plenară
2. Interpenetrating polymer network hydrogels containing LDH, for the controlled release of inulin, M. V. Dumitru, A. Zaharia, O. Pavel, A. L. Neagu, I.-E. Neblea, T. V. Iordache, A. Miron, A. L. Chiriac, A. Sârbu, R. Zăvoianu (poster) SC P03
3. Environmentally responsible polyurethane formulations as alternatives to HTPB binder used in composite propellants, S.V. Dolana, A. Zaharia, T. Rotariu, T.-V. Iordache, A. Sârbu, I. E. Neblea, A. Miron, M. Dumitru, A. Fica (poster) SC P02

Publicarea unui articol ISI în Catalysis Today

Impact of organic/inorganic alkalis in tailoring of Mg/Al-hydrotalcite used in Claisen–Schmidt condensation R. Zăvoianu, S. E. Taha, B. E. Cojocaru, V.I. Pârvulescu, O. D. Pavel - Catalysis Today Volume 435, 1 June 2024, 114693, <https://doi.org/10.1016/j.cattod.2024.114693>

Acceptarea pentru publicare a unui articol în Revue Roumaine de Chimie.

The influence of the precursor type in the synthesis of Mg/Al-hydrotalcite through a non-traditional method used for Claisen–Schmidt condensation, R. Zăvoianu, B. E. Cojocaru, S. G. Ion, A. P. Soares Dias, A. Cruceanu, O. D. Pavel

Pe baza celor prezentate mai sus se poate considera că toate obiectivele propuse pentru această etapă au fost atinse.

In English

FINANCING SOURCE – UEFISCDI Program PNIII

Project Code - PN-III-P2-2.1-PED-2021-1870 –

Contract No. - 646PED/2022

Project Title:

Ecotechnology for obtaining hydrogel encapsulated phytoingredients based on bioactive complexes immobilised in layered double hydroxides matrices

Acronym - DUACTIONMER

Consortium:

Coordinator (CO) – University of Bucharest – Project Manager Conf. Dr. Rodica Zăvoianu

Partner 1 (P1) - INCDPC-ICECHIM Bucharest – Project responsible for partner 1 - Dr. Ing. Andrei Sârbu

Partner 2 (P2) – Research and Processing Center for Medicinal Plants Plantavorel SA (P2) – Project responsible for Partner 2 – Dr. Ing. Carmen Tebrencu

The implementation status of the project:

Stage 3 -2024 SETTING UP OF THE INTEGRATED LABORATORY ECOTECHNOLOGIES HYG-RfL-LDH AND HYG-HtL-LDH

Period: 3 January 2024-20 June December 2024

Stage 2 Financing – 106854,00 lei

Source 2 – budget -106854,00 lei

Sursa 2 – cofinancing –0,00 lei

SUMMARY OF THE SCIENTIFIC REPORT

STAGE 3/2024 – SETTING UP OF THE INTEGRATED LABORATORY ECOTECHNOLOGIES HYG-RfL-LDH AND HYG-HtL-LDH

According to the propose work plan, during the 3rd stage of the project **PN-III-P2-2.1-PED-2021-1870-DUACTIONMER** (January-20 June 2024) the consortium formed by the research teams from **University of Bucharest (CO), INCDPC-ICECHIM Bucharest (P1)** performed **four** experimental-development activities as it follows: **activity 3.1** dedicated to the **experimental demonstration of the functionality** for the dual-action laboratory ecotechnology **HYG-RfL-LDH**, based on the extensive characterization of HYG-RfL-LDH composites (activity 3.1.1. **P1**) as well as on the tests concerning the active ingredients release from HYG-RfL-LDH and the removal efficiency of pollutants from wastewater using the RfL vegetal waste (activity 3.1.2. **CO**); **activity 3.2.** aiming the **experimental demonstration** for the ecotechnology **HYG-HtL-LDH** during which **P1** characterized the materials (activity 3.2.1) while **CO** verified their behavior in the in vitro release tests of the *HtL* phytocomplex and performed the tests for the optimization of the d2 module of the technology in order to increase the adsorption efficiency of the vegetal waste recovered after *HtL* phytocomplex extraction in the wastewater pollutants removal by in flow adsorption processes (activity 3.2.2.); **activity 3.3.** targetting the **validation of HYG-RfL-LDH ecotechnology** included the validation of the module (b_1) related to the immobilisation of RfL phytoextract into LDH matrix (activity 3.3.1-CO) and the validation of the module (c_1) concerning the encapsulation of RfL-LDH into PEGDA hydrogel (activity 3.3.2 **P1**) and **activity 3.4.** consisting of the validation of the ecotechnology HYG-HtL-LDH based on the results obtained by CO for the immobilisation of *HtL* phytoextract into LDH matrixes in the module (b_2) of the technology (activity 3.4.1) and the validation of the module (c_2) related to the encapsulation of *HtL*-LDH into PEGDA-chitosan hydrogel (activity 3.4.2).

During activities 3.1.1 and 3.2.1, P1 extensively characterized the materials using ATR-FTIR, DR-UV-vis, BET, TGA analysis, SEM and XRD techniques and the obtained results confirmed the success of the encapsulation of the LDH immobilised phytoingredients extracted from *RfL* (act. 3.1.1) and *HtL* (act. 3.2.1) respectively. By performing the tests for controlled release of the phytoactive ingredients from the composite hydrogels containing *RfL* (activity 3.1.2) and *HtL* (activity 3.2.2), respectively, CO a proved the efficiency of the hydrogel encapsulation in the preparation of an efficient

system for the slow release of the named phytocomplexes at intestinal pH. The tests performed by CO in the in-flow adsorption of pollutants from synthetic wastewaters using as adsorbents the vegetal wastes recovered after the extraction of *RfL* and *HtL* phytocomplexes (activities 3.1.2. and 3.2.2, respectively) proved that in both cases the pollutants removal exceeds 90% leading to acceptable limits of pollutant concentration in the treated wastewater for a maximum of 3 cycles adsorption/regeneration. The vegetal wastes recovered after *RfL* extraction remove successfully Cr^{6+} , Ni^{2+} and $\text{Fe}^{2+}, \text{Fe}^{3+}$ while those recovered after *HtL* extraction may remove Cu^{2+} , Ni^{2+} and $\text{Fe}^{2+}, \text{Fe}^{3+}$.

The technology **HYG-RfL-LDH** validated in activity 3.3. has a double effect: i) enables the preparation of a biocompatible PEGDA hydrogel composite system entrapping *RfL* extract immobilized in LDH matrix which is efficient in the sustained controlled release at intestinal pH and could be useful for the treatment of laxy transit and ii) allows the treatment of wastewaters contaminated with Cr^{6+} , Ni^{2+} and $\text{Fe}^{2+}, \text{Fe}^{3+}$, hence valorizing the vegetal wastes recovered after the extraction of *RfL* phytocomplex from buckthorn bark.

The technology **HYG-HtL-LDH** validated in activity 3.2. has a double effect: i) enables the preparation of a biocompatible PEGDA-Chitosan hydrogel composite system entrapping *HtL* extract immobilized in LDH matrix which is efficient in the sustained controlled release at intestinal pH and could be useful as prebiotic and ii) allows the treatment of wastewaters contaminated with Cu^{2+} , Ni^{2+} and $\text{Fe}^{2+}, \text{Fe}^{3+}$, thus valorizing the vegetal wastes recovered after the extraction of *HtL* phytocomplex from Jerusalem artichoke.

Activity 3.5. consisted of the results dissemination in

Participation with one plenary lecture and 2 posters at International Chemical Engineering and Material Symposium, SICHEM 202411-12 April 2024, ISSN 2537-2254

1. Advanced ceramic powders obtained via host guest polymerization **A. Sârbu**, A. L. Chiriac, T. Sandu, A. Miron, L. Mara, A. Dumitru, C. Lazau, T. V. Iordache – conferință plenară
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Publishing 1 ISI article în Catalysis Today

Impact of organic/inorganic alkalis in tailoring of Mg/Al-hydrotalcite used in Claisen–Schmidt condensation R. Zăvoianu, S. E. Taha, B. E. Cojocar, V.I. Pârvulescu, O. D. Pavel - Catalysis Today Volume 435, 1 June 2024, 114693, <https://doi.org/10.1016/j.cattod.2024.114693>

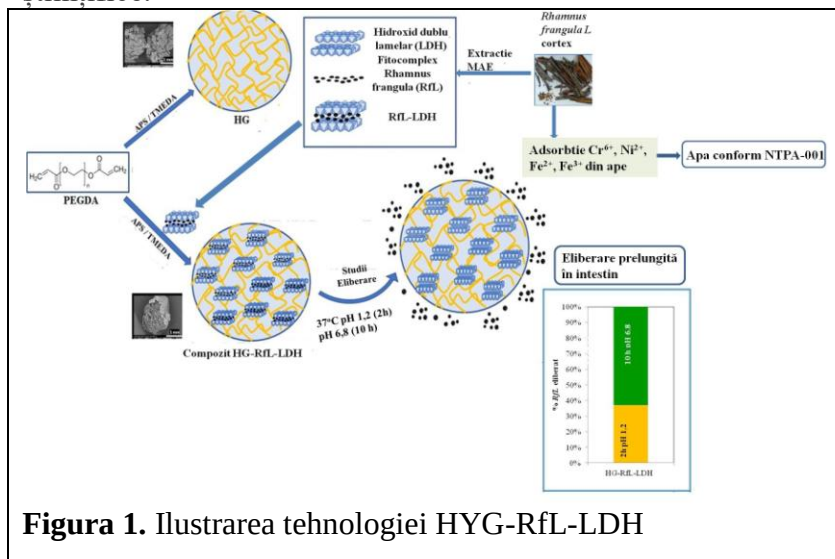
Acceptance for publication of an article în Revue Roumaine de Chimie.

The influence of the precursor type in the synthesis of Mg/Al-hydrotalcite through a non-traditional method used for Claisen–Schmidt condensation, R. Zăvoianu, B. E. Cojocar, S. G. Ion, A. P. Soares Dias, A. Cruceanu, O. D. Pavel

Based on the above presented it may be considered that all the objectives proposed for this stage were reached.

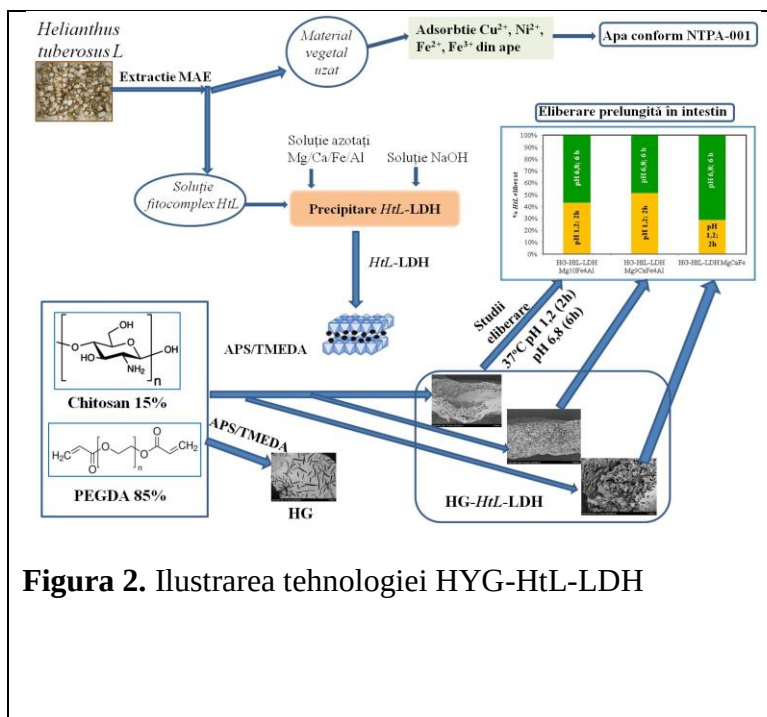
Prezentare succintă a proiectului PN-III-P2-2.1-PED-2021-1870 **Ecotehnologie de obținere a unor fitoingredienți încapsulați în hidrogel pe bază de complecși bioactivi imobilizați în matrice de hidroxizi dubli lamelari (DUACTIVMER) – contract 646PED/2022**

Proiectul a fost realizat de consorțiul format din Universitatea din București (CO), Institutul Național de Cercetare-Dezvoltare pentru Chimie și Petrochimie ICECHIM (P1) și Centrul de Cercetare și Prelucrare a Plantelor Medicinale PLANTAVOREL SA (P2) s-a finalizat prin **validarea a două tehnologii de laborator, depunerea a 2 cereri de brevet înregistrate la OSIM**, publicarea a **6 articole** și prezentarea a **16 comunicări** la manifestări științifice.



producerea contractiilor musculare la nivelul intestinului ușurând astfel tranzitul intestinal se eliberează lent pe parcursul a 10 ore la pH-ul intestinal fără a provoca spasme intestinale dureroase. În urma solubilizării compusului lamelar se eliberează și cantități de oligoelemente care restabilesc echilibrul sărurilor din organism.

b) S-a oferit o variantă de valorificare a unor resturi vegetale care nu își găsesc utilizarea în furajarea animalelor sau ca îngrășământ natural prin folosirea lor ca adsorbanti ai unor poluanți metalici din ape.



Tehnologia HYG-RfL-LDH (figura 1) este o tehnologie cu dublă acțiune care se finalizează prin :

i) obținerea fitoingredienților încapsulați în hidrogel PEGDA biocompatibil pe bază de **compuși bioactivi extrași din scoarța de crușin (*Rhamnus frangula L* cortex)** introduși în spațiul dintre straturile unor compuși lamelari (LDH) ce conțin oligoelemente Mg/Fe/Ca și

ii) prin **utilizarea** resturilor vegetale rămase după extracție **ca adsorbanti pentru urmele de Cr, Ni și Fe din apele reziduale.**

Avantaje : a) În urma încapsulării, compușii bioactivi din crușin care sunt responsabili de

Tehnologia HYG-HtL-LDH (figura 2) este o tehnologie cu dublă acțiune care se finalizează prin :

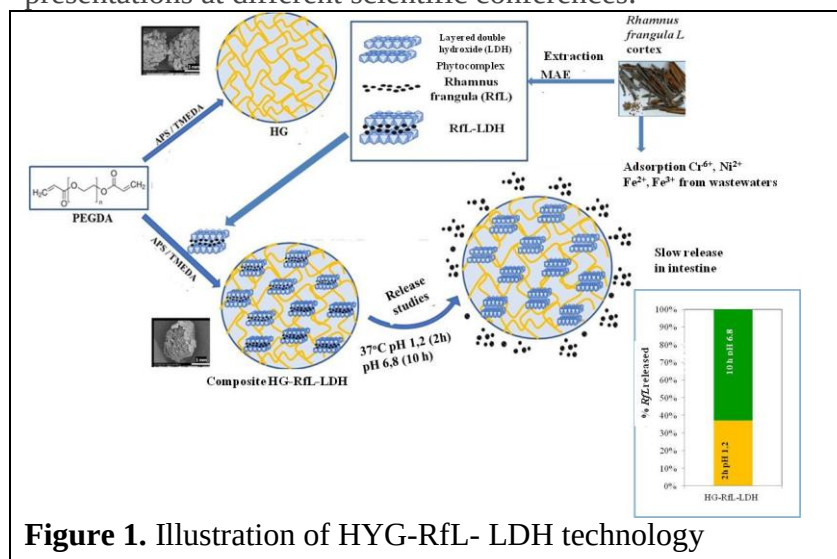
i) obținerea fitoingredienților încapsulați în hidrogel interpenetrant biocompatibil (PEGDA+Chitosan) pe bază de **compuși bioactivi extrași din tuberculi de topinambur (*Helianthus Tuberosus L*)** introduși în sinteza prin precipitare a unor compuși lamelari (LDH) ce conțin oligoelemente Mg/Fe/Ca și

ii) prin **utilizarea** resturilor vegetale rămase după extracție **ca adsorbanti pentru urmele de Cu, Ni și Fe din apele reziduale.**

Avantaje : În urma încapsulării, compușii bioactivi din topinambur (inulina) care au rol de prebiotic și/fibră alimentară se eliberează prelungit pe durata a 6 ore la nivelul intestinului alături de oligoelemente necesare organismului.

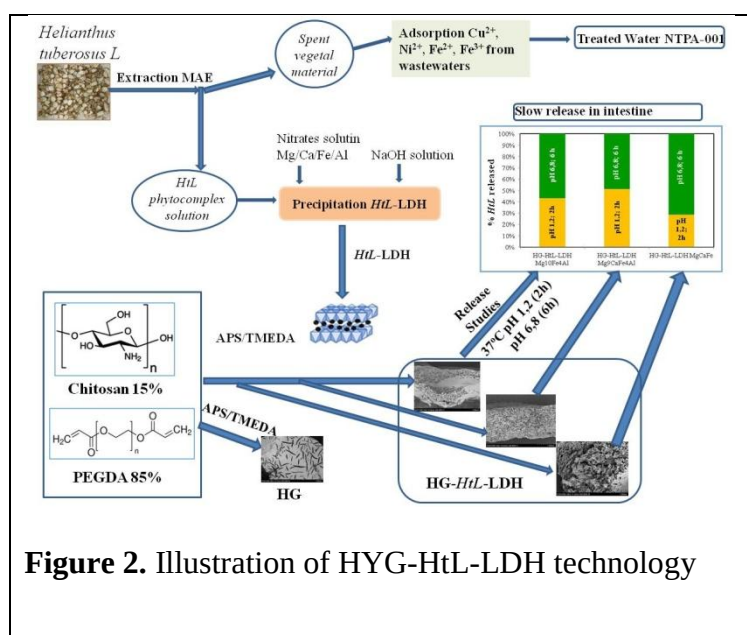
Short presentation of the project PN-III-P2-2.1-PED-2021-1870 **Ecotechnology for obtaining some hydrogel encapsulated phytoingredients based on bioactive complexes immobilised in layered double hydroxide matrixes (DUACTIVMER) – contract 646PED/2022**

The project was developed by a consortium which included the University of Bucharest (CO), the Research & Development National Institute for Chemistry and Petrochemistry ICECHIM (P1) and the Research Center for the Medicinal Plants Processing PLANTAVOREL SA (P2) and was finalized by the **validation of two laboratory technologies**, filling in of **two applications for patents at OSIM**, publishing of **6 ISI articles** and **16 presentations** at different scientific conferences.



Advantages : a) After the encapsulation, the bioactive compounds from buckthorn bark, which are responsible for the peristaltic moves of the bowel enabling the intestinal transit, are slowly released during 10 hours at intestinal pH avoiding painful intestinal spasms. The dissolution of the lamellar compound also releases amounts of oligoelements balancing the electrolytes in the organism.

b) The technology offers a possibility to valorize the spent vegetal material which cannot be used as feed for animals or as natural fertilizer by their utilization as adsorbents for the removal of metal traces from wastewaters.



The technology HYG-HtL-LDH (figure 2) is a double-action technology which is finalized by:

- obtaining of phytoingredients based on **bioactive compounds extracted from Jerusalem artichoke tubers (*Helianthus Tuberosus L*)** introduced in the synthesis by precipitation of some lamellar compounds (**LDH**) containing oligoelements Mg/Fe/Ca encapsulated into a **biocompatible interpenetrant hydrogel (PEGDA+Chitosan)** and
- the **utilization** of the spent vegetal material recovered after the extraction as **adsorbents for traces of Cu, Ni and Fe from wastewaters..**

Advantages : After encapsulation the bioactive compounds from Jerusalem artichoke (inulin) which act as prebiotic and/dietary fiber are slowly released during 6 hours in the intestine along with oligoelements necessary for the organism.