



# Metal–Organic Framework Integrated Layered Double Hydroxide (MOF–LDH) Nanocomposites for Enhanced Chemical Adsorption and Cleaner Wet Processing of Leather

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## Abstract

The leather processing industry relies heavily on wet chemical operations, including soaking, liming, tanning, dyeing, and fatliquoring, where inefficient uptake of chemical agents contributes to high chemical consumption, wastewater generation, and environmental concerns. The development of functional materials capable of improving chemical adsorption during leather processing is therefore essential for sustainable leather manufacture. Metal–Organic Framework–Layered Double Hydroxide (MOF–LDH) nanocomposites represent a promising class of hierarchical materials that combine the tunable layered structure of layered double hydroxides (LDHs) with the high porosity and adsorption capacity of metal–organic frameworks (MOFs). In this study, a MOF–LDH hierarchical composite material was designed and evaluated as an adsorption-enhancing additive for wet leather processing. The combination of LDH interlayer anion exchange capability and MOF porous channels provides multiple active adsorption sites for interaction with tanning agents, dyes, and fatliquoring molecules. Structural characterisation using X-ray photoelectron spectroscopy (XPS), Brunauer–Emmett–Teller (BET) surface analysis, scanning electron microscopy (SEM), and transmission electron microscopy (TEM) demonstrated successful formation of the hierarchical composite structure. The synthesised MOF–LDH particles exhibited nanoscale morphology with an average size of approximately 100 nm and a porous surface architecture. Application of MOF–LDH during chrome tanning significantly improved leather performance. Compared with conventional leather treated with 2% chrome tanning agent, MOF–LDH-assisted tanning increased the shrinkage temperature from 73°C to 89°C, indicating enhanced collagen stabilisation. Dye uptake increased from 87% to 92%, while fatliquor absorption improved from 88% to 94%. The softness value also increased, demonstrating improved physical properties of the finished leather. These improvements are attributed to enhanced adsorption and fixation of ionic chemical species within the collagen matrix. The results indicate that MOF–LDH materials provide an effective strategy for improving chemical utilisation efficiency, reducing chemical discharge, and promoting cleaner production in leather manufacturing.

**Keywords:** metal–organic framework; layered double hydroxide; adsorption; leather tanning; chrome reduction; sustainable processing; nanocomposite.

## 1. INTRODUCTION

### 1.1 Background of Leather Wet Processing

Leather manufacturing is one of the oldest industrial activities and remains economically important worldwide. The transformation of animal hides into durable leather requires a sequence of chemical and mechanical operations, among which wet processing stages determine the final quality and functional properties of leather. These stages include soaking, liming, deliming, bating, tanning, dyeing, and fatliquoring.

Among these operations, tanning is considered the most critical stage because it converts unstable collagen fibres into a stable material resistant to heat, microorganisms, and degradation. Chromium salts, particularly basic chromium sulphate, are widely used because of their high tanning efficiency and ability to produce leather with desirable softness, strength, and hydrothermal stability (Covington, 2009).



However, traditional chrome tanning has limitations. A portion of chromium salts remains unabsorbed and is discharged into wastewater, creating environmental challenges. Improving chromium uptake and reducing chemical waste are therefore major goals in modern leather technology

### 1.1.1 Challenges Associated with Chemical Absorption in Leather Processing

Leather contains a complex collagen fibre network containing functional groups such as amino, carboxyl, and hydroxyl groups. Interaction between these groups and processing chemicals determines absorption efficiency. However, uneven penetration, limited active binding sites, and competition between chemical species often reduce utilisation efficiency.

During dyeing and fatliquoring, poor chemical absorption can lead to:

1. Increased chemical consumption.
2. Higher wastewater pollution load.
3. Uneven colouration.
4. Reduced softness and physical performance.

Therefore, advanced adsorption materials that can promote controlled interaction between chemicals and collagen fibres are receiving increasing attention.

### 1.2 Metal–Organic Frameworks as Advanced Adsorbent Materials

Metal–organic frameworks (MOFs) are crystalline porous materials composed of metal ions or clusters connected by organic ligands. They possess high surface areas, tunable pore structures, and adjustable chemical functionality.

According to Furukawa et al. (2013), MOFs provide exceptional opportunities for adsorption applications because their pore size, surface chemistry, and molecular interactions can be tailored during synthesis.

MOFs have been widely studied for:

- gas separation,
- catalysis,
- drug delivery,
- pollutant removal,
- chemical adsorption.

Their large internal surface areas allow interaction with various molecules through electrostatic forces,

hydrogen bonding, and coordination interactions.

However, pure MOFs may have limitations, including low stability under certain chemical environments. Combining MOFs with layered materials provides an effective approach to improving stability and adsorption performance.

### 1.3 Layered Double Hydroxides and Their Adsorption Mechanism

Layered double hydroxides (LDHs) are inorganic layered materials structurally similar to hydrotalcite. They consist of positively charged metal hydroxide layers balanced by exchangeable interlayer anions.

The general LDH structure provides:

- ion exchange capability,
- surface hydroxyl groups,
- adjustable composition,
- chemical stability.

LDHs have been extensively investigated for adsorption because their interlayer regions can capture negatively charged species through anion exchange mechanisms (Meyn et al., 1990).

In leather processing, these characteristics are valuable because many processing chemicals, including dyes and tanning complexes, contain ionic functional groups.

### 1.4 Synergistic Effect of MOF–LDH Composite Materials

Combining MOFs and LDHs creates a hierarchical material with complementary adsorption mechanisms.

The LDH component provides:

- ionic exchange sites,
- layered diffusion pathways,
- surface hydroxyl groups.

The MOF component provides:

- porous cavities,
- high surface area,
- molecular-level adsorption sites.

The resulting MOF–LDH composite can simultaneously improve adsorption capacity and chemical fixation.

This hierarchical structure offers potential advantages in leather processing by increasing penetration and retention of tanning agents, dyes, and fatliquors within collagen fibres.



## 2. MATERIALS AND METHODS

### 2.1 Materials

The materials used in this study were selected to enable the synthesis of MOF–LDH hierarchical nanocomposites and evaluate their influence on wet leather processing.

Metal salts, including divalent and trivalent metal precursors, were used as the inorganic sources for LDH formation, while organic linkers were used for MOF framework construction. Analytical-grade reagents were employed without further purification.

The leather substrate used for application experiments was chrome-free or low-chrome processed hide material suitable for tanning evaluation. Basic chromium sulphate was selected as the conventional tanning agent for comparison.

The major chemical materials included:

- metal ion precursors for LDH synthesis,
- organic ligands for MOF formation,
- chromium tanning salts,
- commercial dyes,
- fatliquoring agents,
- buffer chemicals for pH adjustment.

### 2.2 Synthesis of MOF–LDH Hierarchical Composite

The MOF–LDH composite was prepared through a controlled in-situ growth strategy to combine the layered structure of LDH with the porous framework of MOF.

#### 2.2.1 Preparation of LDH Structure

The LDH precursor was synthesised using a co-precipitation method. Appropriate metal salt solutions were mixed under continuous stirring while maintaining alkaline conditions.

The precipitation reaction resulted in the formation of positively charged hydroxide layers. Interlayer anions were incorporated to balance the charge between the layers.

The obtained suspension was aged to improve crystallinity and structural organisation. The product was then washed, filtered, and dried.

#### 2.2.2 Construction of MOF Framework

The MOF component was prepared by coordinating metal ions with organic ligands under controlled conditions. During synthesis, coordination between metal centres and organic linkers generated a three-dimensional porous framework.

The MOF structure was subsequently combined with LDH through interfacial assembly, producing the hierarchical MOF–LDH composite.

The expected structure consisted of:

1. LDH nanosheets acting as ion exchange layers.
2. MOF nanoparticles providing porous adsorption cavities.
3. Interface regions providing additional active adsorption sites.

### 2.3 Characterisation of MOF–LDH Composite

#### 2.3.1 X-ray Photoelectron Spectroscopy (XPS)

XPS analysis was performed to determine elemental composition and chemical bonding states.

The presence of characteristic metal, oxygen, and carbon signals confirmed successful formation of the composite material.

The interaction between MOF and LDH components indicated the formation of a stable hierarchical structure.

#### 2.3.2 Brunauer–Emmett–Teller (BET) Surface Area Analysis

The adsorption capacity of porous materials is strongly influenced by specific surface area and pore structure.

BET analysis demonstrated that the synthesised MOF–LDH possessed a specific surface area of approximately 20.60 m<sup>2</sup> g<sup>-1</sup>.

The increased surface area provided additional active regions for interaction with leather-processing chemicals.

The porous structure contributed to:

- improved diffusion of chemical molecules,
- enhanced adsorption efficiency,
- increased retention of treatment agents.



### 2.3.3 Scanning Electron Microscopy (SEM)

SEM was used to investigate morphology and surface structure.

The composite particles displayed a distinct polyhedral morphology with a rough surface texture.

The formation of nanoscale structures increased contact area between the material and chemical species.

### 2.3.4 Transmission Electron Microscopy (TEM)

TEM analysis revealed that MOF-LDH particles were approximately 100 nm in size.

The images confirmed the presence of interconnected structural regions, demonstrating a successful combination of layered and porous components.

## 2.4 Application of MOF-LDH in Leather Tanning

### 2.4.1 Tanning Process

Leather samples were treated using a conventional chrome tanning process and compared with MOF-LDH-assisted tanning.

The experimental groups included:

1. Conventional chrome tanning.
2. MOF-LDH-assisted chrome tanning.

The MOF-LDH additive was introduced during the tanning stage to enhance adsorption and fixation of chromium complexes.

### 2.4.2 Evaluation of Leather Properties

The treated leather samples were evaluated according to physical and chemical performance indicators.

The investigated properties included:

- shrinkage temperature,
- dye absorption,
- fatliquor absorption,
- softness,
- colour characteristics.

## 3. RESULTS AND DISCUSSION

### 3.1 Structural Characteristics of MOF-LDH

The characterisation results confirmed successful synthesis of the hierarchical MOF-LDH material.

The combination of LDH layers and MOF porous structures produced a multifunctional adsorbent.

The LDH structure contributed ion exchange capacity, while MOF pores increased molecular adsorption.

Such synergistic behaviour has been reported as a major advantage of hybrid porous materials (Zhang et al., 2018).

### 3.2 Effect of MOF-LDH on Chromium Tanning

Chromium tanning efficiency depends on the interaction between chromium complexes and collagen functional groups.

The addition of MOF-LDH improved chromium fixation by increasing adsorption sites.

The shrinkage temperature increased from:

73°C (conventional chrome tanning)

to

89°C (MOF-LDH-assisted tanning).

The increase indicates a stronger stabilisation of collagen fibres.

The improved thermal stability can be attributed to enhanced chromium coordination and more uniform distribution within the collagen matrix.

### 3.3 Improvement of Dye Absorption

Dye uptake is controlled by chemical interactions between dye molecules and collagen fibres.

Traditional dyeing processes often suffer from incomplete adsorption, resulting in wastewater colouration.

The MOF-LDH composite increased dye absorption from:

87% to 92%.

The improvement was caused by:

1. Electrostatic attraction between charged groups.
2. Porous adsorption sites from MOF structures.
3. Improved penetration into collagen networks.

The composite acted as a carrier and fixation promoter, increasing the dye's utilisation efficiency.

### 3.4 Improvement of Fatliquor Absorption

Fatliquoring provides flexibility and softness by introducing lubricating molecules between collagen fibres.

The presence of MOF-LDH enhanced fatliquor absorption, increasing it from 88% to 94%.

88% to 94%.



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The improvement resulted from stronger interaction between fatliquor molecules and active sites on the composite surface.

Higher fatliquor retention contributed to improved mechanical flexibility.

### 3.5 Effect on Leather Softness

The softness value increased from:

8.0 mm to 9.5 mm.

This improvement demonstrates better fibre lubrication and reduced internal friction.

The hierarchical porous structure allowed more uniform distribution of fatliquoring substances, producing a softer leather structure.

### 3.6 Mechanism of Enhanced Chemical Adsorption by MOF-LDH During Leather Processing

#### 3.6.1 Interaction Between MOF-LDH and Chromium Complexes

The improvement in chrome tanning efficiency observed after MOF-LDH application can be explained by the interaction between the composite material and chromium tanning species.

Chromium complexes in tanning solutions exist as positively charged and hydrolysed chromium species depending on pH conditions. Collagen fibres contain negatively charged carboxyl groups that interact with chromium ions during tanning.

The MOF-LDH composite provides additional adsorption sites through:

1. Surface hydroxyl groups.
2. Exchangeable ions within LDH layers.
3. Coordinated metal centres within MOF structures.
4. Internal porous cavities.

These active regions increase the concentration of chromium complexes near collagen fibres and facilitate stronger cross-linking reactions.

The porous MOF component improves molecular transport, while the LDH component enhances ionic interactions. This combination results in improved chromium fixation and reduced chromium discharge.

#### 3.6.2 Adsorption Mechanism of Dye Molecules

Leather dyes usually contain sulfonic groups and other ionic functional groups that determine their affinity toward collagen.

During dyeing, MOF-LDH improves adsorption through several mechanisms:

##### Electrostatic Interaction

The positively charged LDH layers attract negatively charged dye molecules, promoting dye retention.

##### Hydrogen Bonding

Hydroxyl groups on LDH surfaces interact with functional groups in dye molecules.

##### Pore Filling

MOF cavities provide nanoscale spaces where dye molecules can be physically trapped.

##### Surface Coordination

Metal centres within the MOF structure may interact with dye molecules through coordination effects. The combined adsorption pathways increase dye fixation and improve colour uniformity.

#### 3.6.3 Interaction with Fatliquoring Agents

Fatliquors are generally composed of oil-based molecules or modified lipid structures.

The efficiency of fatliquoring depends on penetration and distribution within the collagen fibre network.

MOF-LDH improves fatliquor performance through:

- increased surface affinity,
- improved dispersion,
- stronger retention between fibres.

The porous structure acts as a reservoir that facilitates gradual release and distribution of fatliquoring molecules.

Consequently, the treated leather exhibits improved flexibility and softness.



#### 4. ENVIRONMENTAL SIGNIFICANCE OF MOF-LDH-ASSISTED LEATHER PROCESSING

##### 4.1 Reduction of Chemical Waste

Excess chemicals are frequently used in conventional leather processing because collagen fibres absorb only a portion of the applied materials.

Low chemical uptake results in:

- increased wastewater treatment requirements,
- higher production costs,
- environmental pollution.

The use of MOF-LDH improves chemical absorption efficiency, allowing lower chemical dosages while maintaining leather quality.

##### 4.2 Cleaner Chrome Tanning Technology

Chromium pollution remains one of the main environmental concerns associated with leather production.

Although chromium(III) is less toxic than chromium(VI), improper disposal may lead to environmental

accumulation and possible oxidation into more harmful forms.

Improving chromium utilisation through adsorption enhancement reduces:

- chromium concentration in wastewater,
- sludge generation,
- chemical losses.

The MOF-LDH strategy therefore supports cleaner tanning practices.

##### 4.3 Contribution to Sustainable Leather Manufacturing

Sustainable leather production requires improved resource efficiency and reduced environmental impact.

MOF-LDH materials contribute to sustainability by:

1. Increasing chemical utilisation.
2. Reducing pollutant discharge.
3. Improving product quality.
4. Supporting low-waste processing.

The approach aligns with modern principles of green chemistry, including prevention of waste and improved material efficiency.

**Table 1.** Comparison between Conventional and MOF-LDH-Assisted Leather Processing

Parameter	Conventional Chrome Tanning	MOF-LDH Assisted Tanning
Chromium fixation	Moderate	Improved
Shrinkage temperature	73 °C	89 °C
Dye absorption	87%	92%
Fatliquor absorption	88%	94%
Softness	8.0 mm	9.5 mm
Chemical utilization	Lower	Higher
Waste generation	Higher	Reduced

The comparison demonstrates that incorporation of MOF-LDH significantly improves processing efficiency and leather properties.

#### 5. FUTURE PERSPECTIVES

##### 5.1 Optimisation of MOF-LDH Structure

Future research should focus on controlling:

- particle size,
- pore distribution,
- surface functional groups,

- LDH layer composition.

Optimisation of these characteristics may further improve adsorption capacity.

##### 5.2 Application in Other Leather Processing Stages

Although this study focuses mainly on tanning, MOF-LDH materials may also be applied during:

- soaking,
- dyeing,
- retanning,
- finishing.



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Their adsorption capability may improve the fixation of different processing chemicals.

### 5.3 Industrial Scale-Up Challenges

Despite laboratory success, industrial implementation requires evaluation of:

- production cost,
- large-scale synthesis,
- recycling ability,
- long-term stability.

The development of inexpensive and environmentally friendly synthesis routes remains important.

## 6. CONCLUSION

Metal–Organic Framework–Layered Double Hydroxide (MOF–LDH) hierarchical composites represent a promising advanced material for improving chemical absorption during wet leather processing.

The combination of LDH layered structures and MOF porous frameworks provides multiple adsorption mechanisms, including ion exchange, electrostatic attraction, hydrogen bonding, and pore confinement.

Characterisation results confirmed the successful formation of a nanoscale porous composite with favourable structural properties. Application of MOF–LDH during chrome tanning significantly enhanced leather performance.

The addition of MOF–LDH increased shrinkage temperature from 73 °C to 89 °C, improved dye absorption from 87% to 92%, increased fatliquor absorption from 88% to 94%, and improved softness from 8.0 mm to 9.5 mm.

These improvements demonstrate that MOF–LDH can increase chemical utilisation efficiency, reduce processing waste, and support cleaner leather manufacturing.

The application of hierarchical adsorption materials provides a new direction for sustainable leather

technology and reduced-impact industrial processing.

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