ML driven high-pressure Li-Cu-H ternary hydrides discovery

NOTE

- The study and results presented here represent an initial exploratory investigation.

- While the predictions are grounded in well-established theoretical principles and robust machine-learning models, further detailed analysis—including literature review, additional calculations, and deeper physical interpretation—remains to be carried out.

Workflow

- Generated ~2,000 Li–Cu–H structures using **MatterGen**, each with lattice constants and atomic coordinates (POSCAR format).
- Used **MatterSim + MLIP** (trained on >5M structures) to fully relax all candidates under pressures from **0 to 100 GPa**.
- Collected stress, energy, enthalpy, and other properties from the relaxed structures.
- Ran **DFT calculations** for pure Li, Cu, and H reference phases to obtain their accurate energetic baselines at the same pressures.
- Evaluated **formation enthalpy** for every Li–Cu–H compound using the appropriate elemental and molecular reference states.

Workflow

- Li-Cu-H formation/reaction

$$a\,Li(s) + b\,Cu(s) + (c/2)\,H_2(g) \rightarrow Li_aCu_bH_c$$

- Formation enthalpy

$$\Delta H_f(P) = [\, H_{tot}(cell, P) - (\, a \cdot H_{Li}(P) + b \cdot H_{Cu}(P) + (c/2) \cdot H_{H2}(P)\,)\,] / (a + b + c)$$

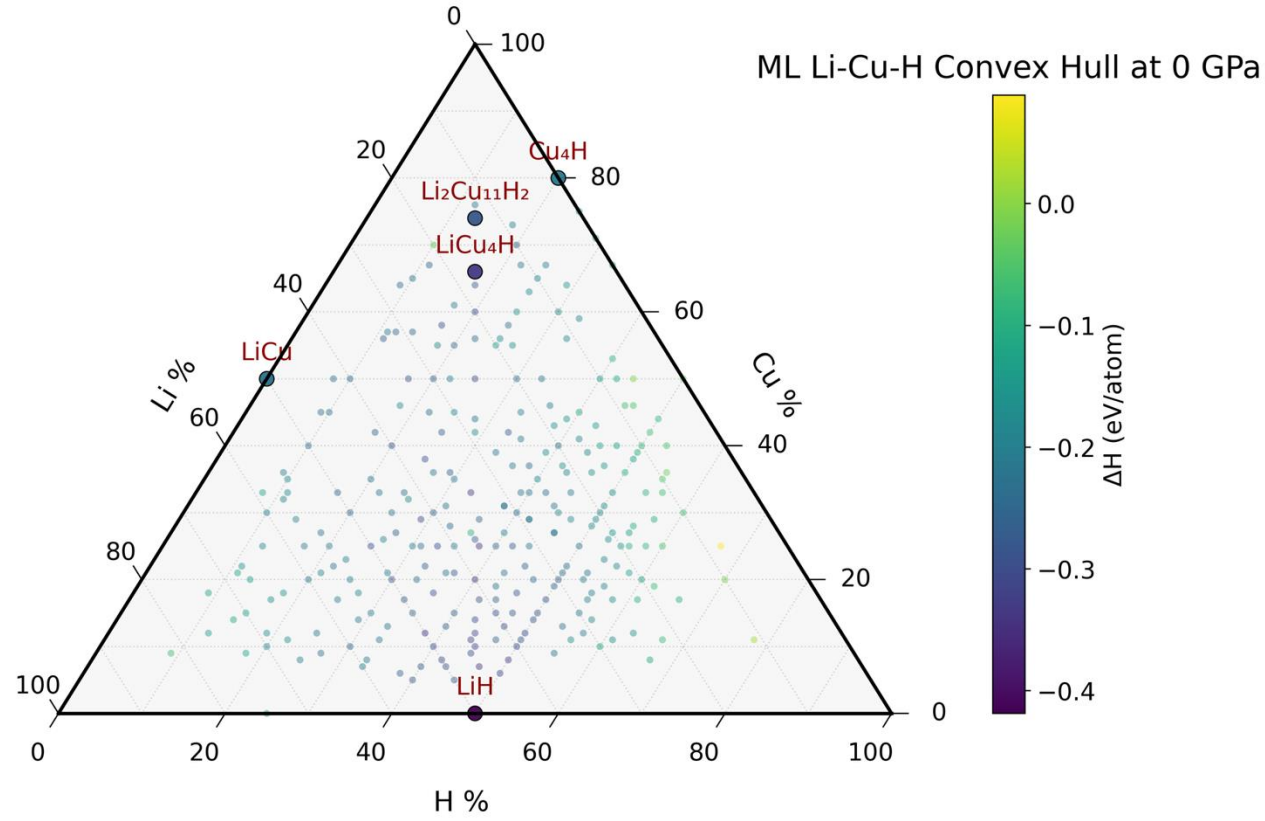
Workflow

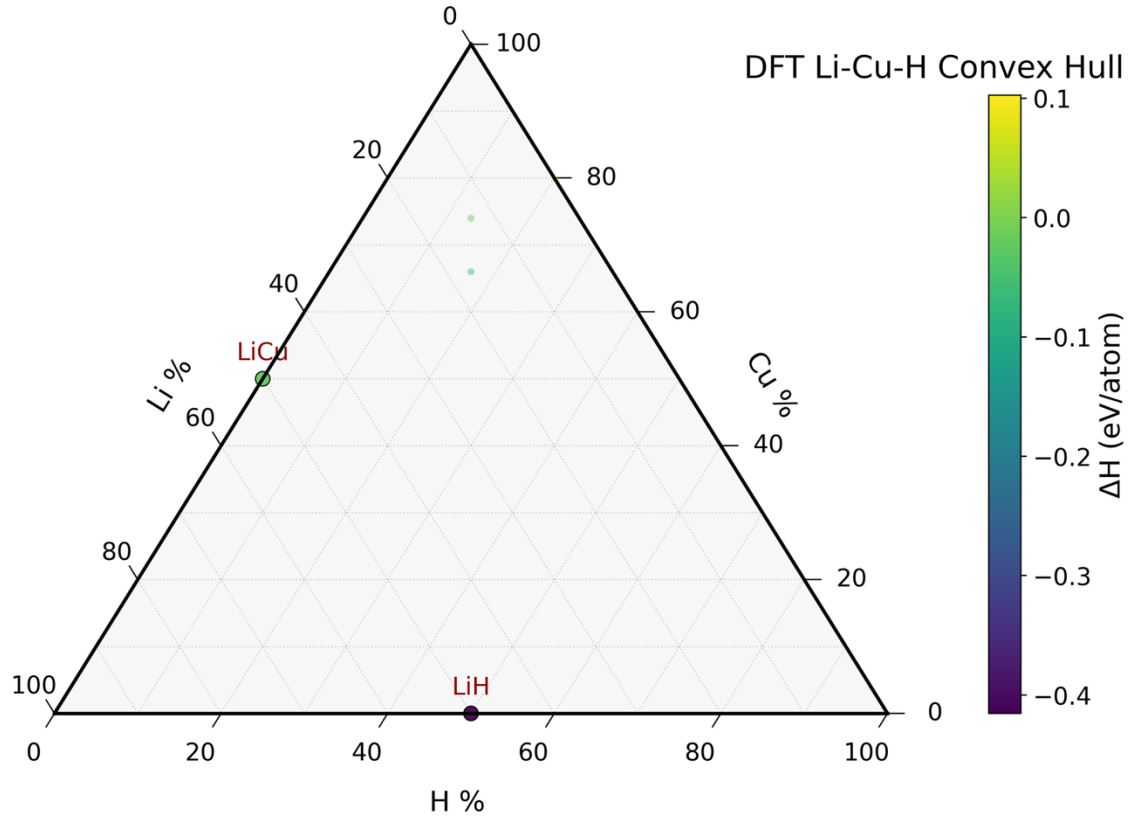
- ML-predicted stable Li–Cu–H phases → re-optimized using **full DFT relaxation**
- Post-relaxation **SCF step** yields accurate energies & enthalpies
- **Formation enthalpy recomputed** using identical reference scheme
- **DFT convex hull** built at each pressure
- **ML vs. DFT stability** compared for consistency and validation

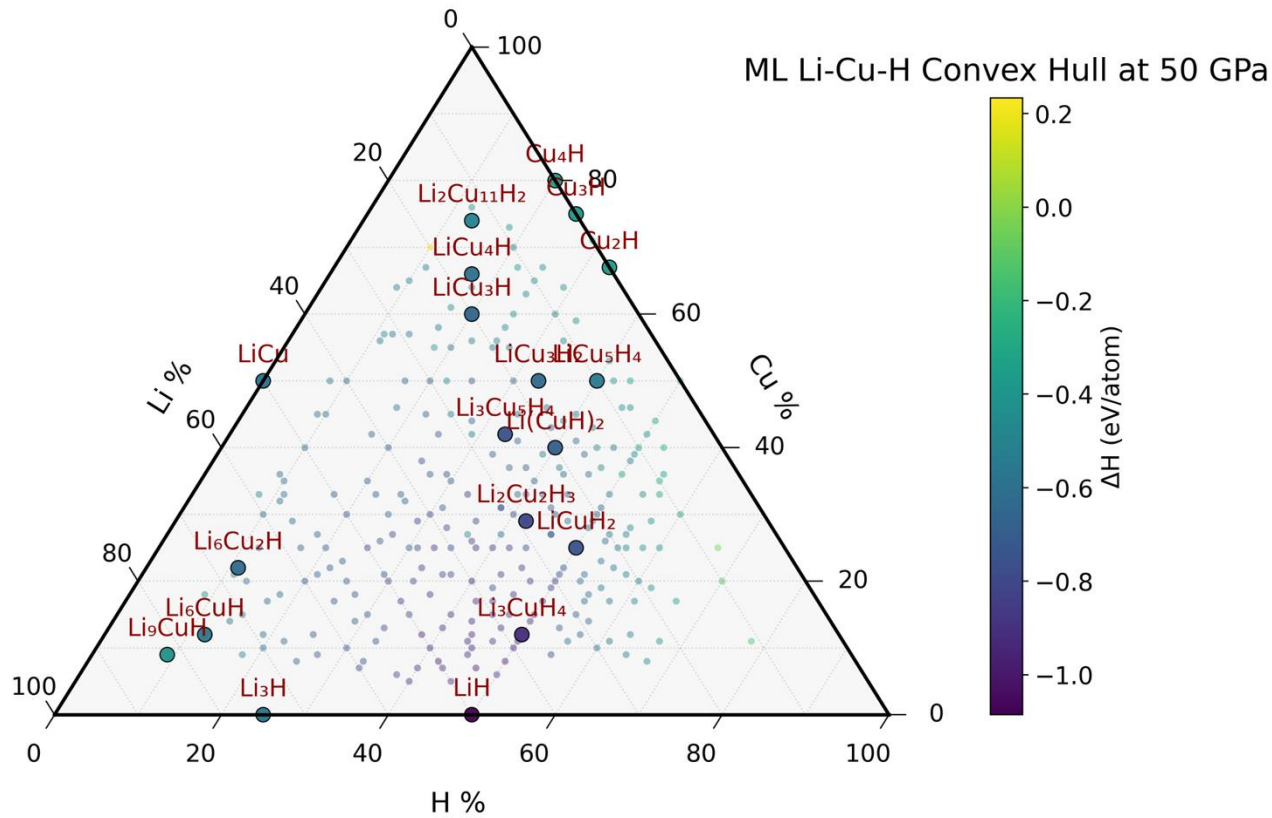
Overall Workflow

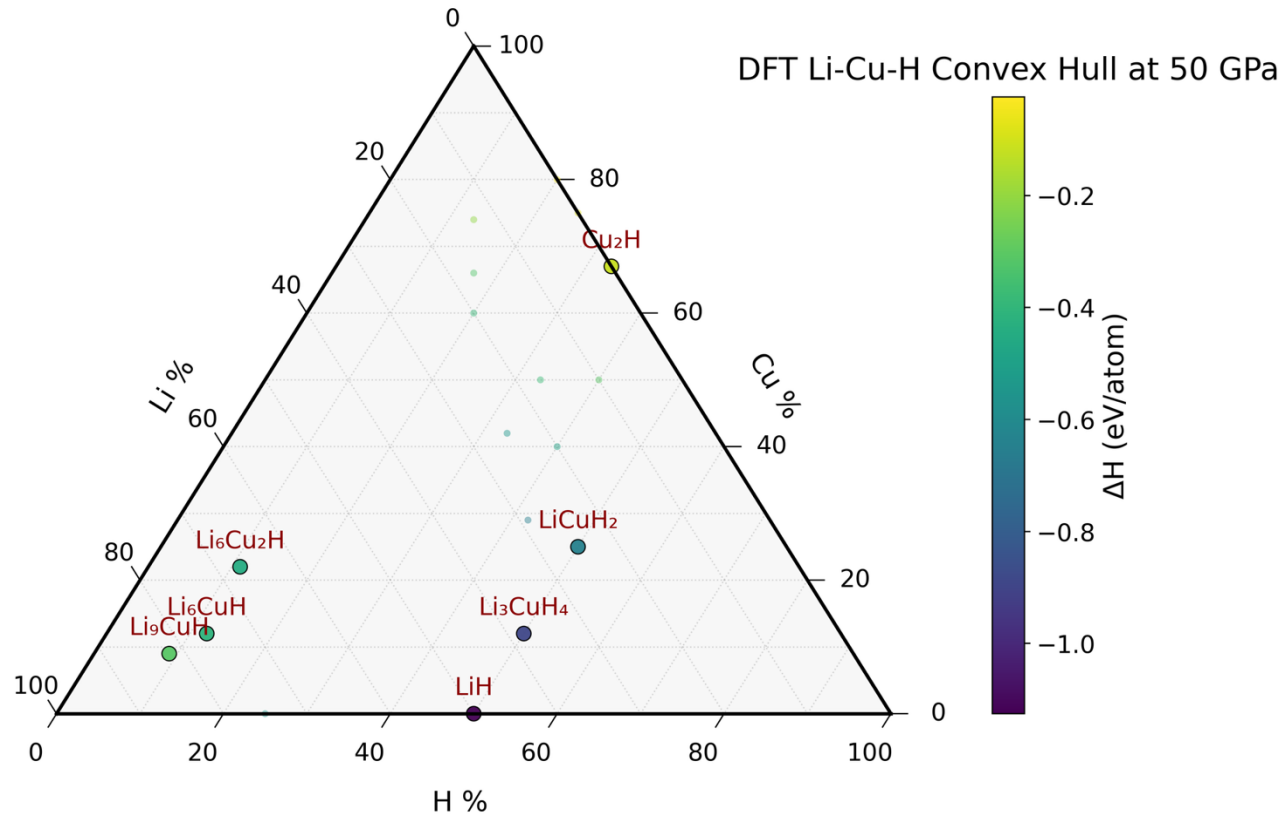
1. **Structure Generation**
   - Use MatterGen to create ~2000 Li–Cu–H candidate structures.
2. **ML Relaxation (MatterSim)**
   - Relax structures with MLIP at pressures from 0 to 100 GPa.
   - Extract enthalpy, stress, energy.
3. **ML Convex Hull Construction**
   - Compute ΔHf for all relaxed structures.
   - Identify ML-stable compositions.
4. **DFT Verification**
   - Fully relax ML-stable candidates with DFT at their target pressures.
   - Perform SCF step to obtain accurate enthalpy.
5. **DFT Convex Hull Construction**
   - Recompute ΔHf using DFT energetics.
   - Compare DFT hull with ML hull.
6. **Cross-Validation**
   - Confirm which phases remain stable.
   - Analyze pressure-dependent phase stability.

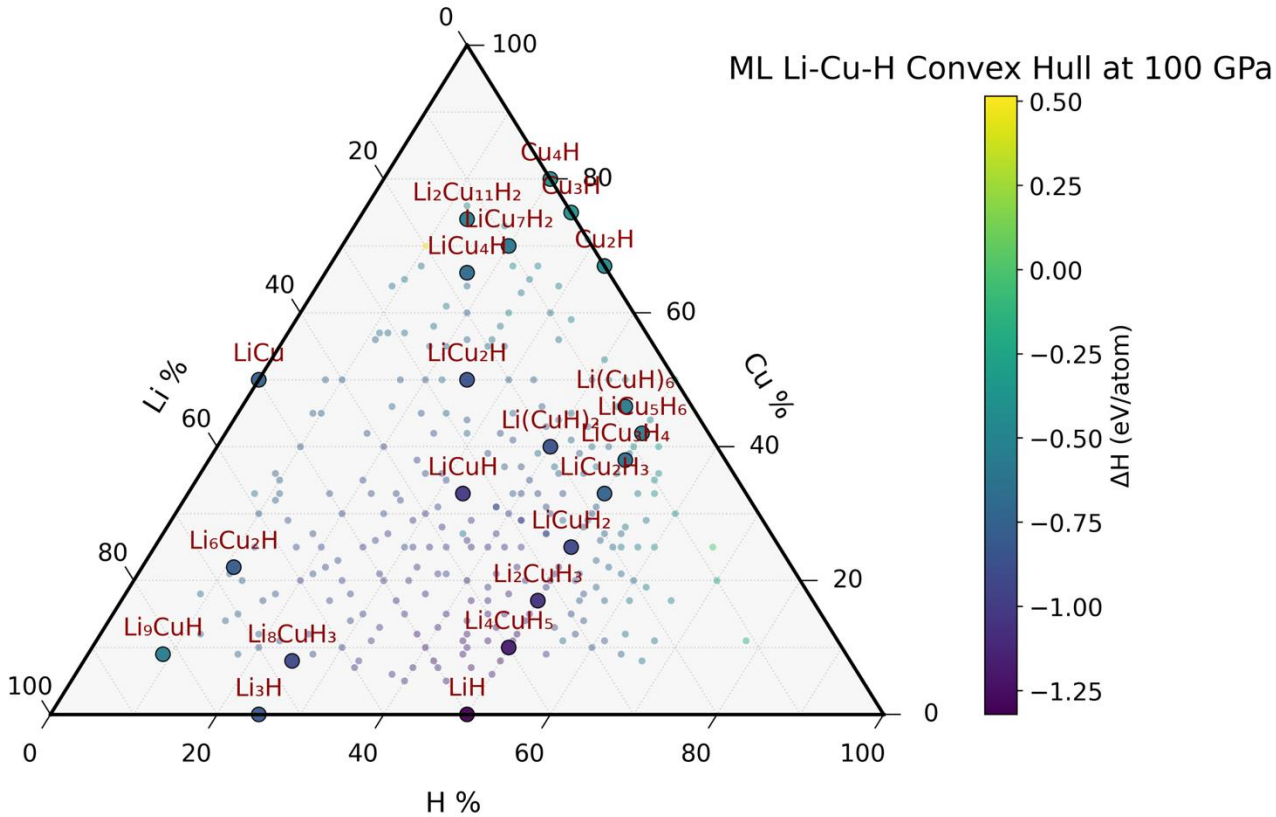
Results

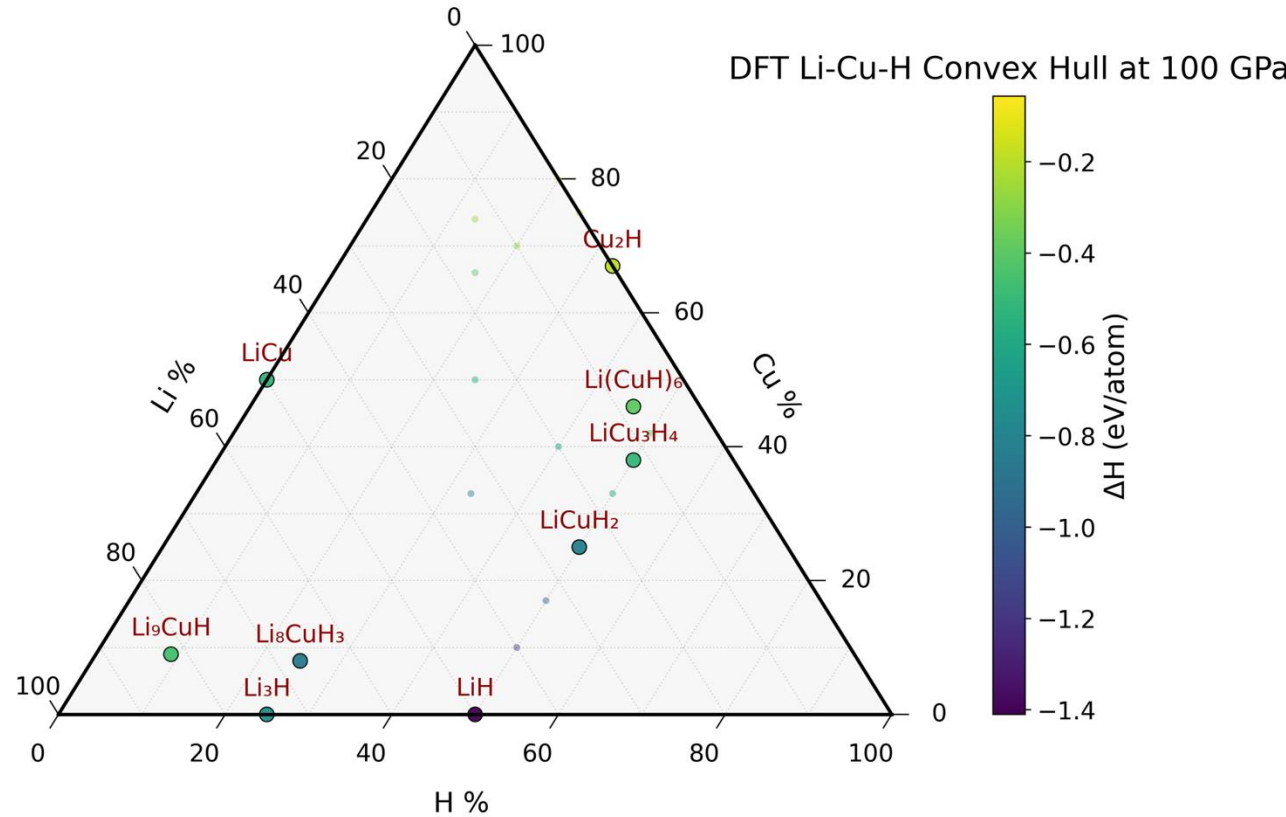

ML Li-Cu-H Convex Hull at 0 GPa
Cu₄H
Li₂Cu₁₁H₂
LiCu₄H
LiCu
LiH
Li %
Cu %
H %
ΔH (eV/atom)
0.0
−0.1
−0.2
−0.3
−0.4


DFT Li-Cu-H Convex Hull at 0 GPa
LiCu
LiH
Li %
Cu %
H %
ΔH (eV/atom)
0.1
0.0
−0.1
−0.2
−0.3
−0.4


ML Li-Cu-H Convex Hull at 50 GPa
Li %
Cu %
H %
ΔH (eV/atom)
Cu₄H
Li₂Cu₁₁H₂
Cu₃H
LiCu₄H
Cu₂H
LiCu₃H
LiCu
LiCu₃H₂
LiCu₅H₄
Li₃Cu₅H₄
Li(CuH)₂
Li₂Cu₂H₃
LiCuH₂
Li₆Cu₂H
Li₆CuH
Li₉CuH
Li₃CuH₄
Li₃H
LiH


DFT Li-Cu-H Convex Hull at 50 GPa
Li %
Cu %
H %
ΔH (eV/atom)
Cu₂H
LiCuH₂
Li₆Cu₂H
Li₆CuH
Li₉CuH
Li₃CuH₄
LiH


ML Li-Cu-H Convex Hull at 100 GPa
Li %
Cu %
H %
ΔH (eV/atom)
Cu₄H
Li₂Cu₁₁H₂
Cu₃H
LiCu₇H₂
Cu₂H
LiCu₄H
LiCu
LiCu₂H
Li(CuH)₆
LiCu₅H₆
Li(CuH)₂
LiCu₃H₄
LiCuH
LiCu₂H₃
LiCuH₂
Li₆Cu₂H
Li₂CuH₃
Li₄CuH₅
Li₉CuH
Li₈CuH₃
Li₃H
LiH


DFT Li-Cu-H Convex Hull at 100 GPa
Li %
Cu %
H %
ΔH (eV/atom)
Cu₂H
LiCu
Li(CuH)₆
LiCu₃H₄
LiCuH₂
Li₉CuH
Li₈CuH₃
Li₃H
LiH

Detail Results

- All convex hull plots stored in *images* folder

- Data including stable structures, space group, energies, enthalpies, … in

outputs_hull_mattersim and

outputs_hull_DFT folders

and <*P_GPa*> and *structures* sub-folders inside

Key findings

- At low pressure, as validated by ML plus DFT, the Li–Cu–H phase space is dominated by binary compounds, with LiH and LiCu forming the only stable phases.

- As pressure increases, particularly beyond ~50 GPa, the energetic landscape shifts, enabling the stabilization of Li–Cu–H ternary compositions.