Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils

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Graphene has been attracting great interest because of its distinctive band structure and physical properties. Today, graphene is limited to small sizes because it is produced mostly by exfoliating graphite. We grew large-area graphene films of the order of centimeters on copper substrates by chemical vapor deposition using methane. The films are predominantly single-layer graphene, with a small percentage (less than 5%) of the area having few layers, and are continuous across copper surface steps and grain boundaries. The low solubility of carbon in copper appears to help make this growth process self-limiting. We also developed graphene film transfer processes to arbitrary substrates, and dual-gated field-effect transistors fabricated on silicon/silicon dioxide substrates showed electron mobilities as high as 4050 square centimeters per volt per second at room temperature.

papers:
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• growth process
• growth results
• growth details
• transferring grown graphene
• transport property
graphene synthesis:

- exfoliation (<1000μm² size)
- chem. reduction of graphite oxide (contamination)
- UHV annealing of SiC (strongly bound single layer)
- growth in metal substrates:

  here: 25μm Cu foils as substrate/catalyst
  @~1000°C in glass tube furnace
  H₂/CH₄ gas atmosphere

result: carbon single layer in cm-range (!)
process

process flow:

1. load the substrate (Ni, Cu, Co..)
2. heating the furnace (40 mTorr)
3. induce carbon source (gas)
4. cool down to RT

As-received Cu

Cu + Graphene

H₂ = 2 sccm

CH₄ = 35 sccm

P = ~40 mTorr

P = ~500 mTorr

Growth

Temperature (°C)

Time (min)
growth in time

SEM pictures

1min

2.5min

10min

60min
growth results
testing the growth mode

high solubility of C in the metal:

Dissolution

Surface segregation

Precipitation

1st isotope

2nd isotope

cooling down

low solubility of C in the metal:

Surface adsorption

1st isotope

2nd isotope

if atoms can't diffuse into the metal:
- isotopes don't mix
- growth only occurs on uncovered surface
self-limiting growth on catalytic Cu surface

different on Ni surface:
due to higher solubility the carbon diffuses and forms multilayer graphene by precipitation
transfere on substrate

PDMS or PMMA stamping:

Scoop floating graphene with desired substrate:
graphene transferred to Si chip + lithographic patterning

do double-gated FET device:
- topgate (Al₂O₃ dielectric)
- backgate (doped Si / SiO₂)
- room temp. measurements
  mobility ~ 4000 cm²/Vs
  with $\mu \sim 3 \times 10^{11}$ cm⁻²

$\rightarrow$ quality comparable to conventional flakes
Conclusion

- CWD growth on Cu is self-limiting
- this leads the cm-sized continuous graphene "sheets"
- transfer to arbitrary substrates is possible
- good quality according to transport and Raman measurements

The way is free for wafer-sized graphene production!