Understanding the Molecular Complexity of Astrochemistry
Laboratory synthesis of prebiotic species

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The Open University
Core Questions for Astrochemistry & Astrobiology

• Where are the building blocks of life synthesised? ISM or on planet?

• Are the conditions for such synthesis common/universal?

• European Astrobiology Roadmap. Published 2016
Outline

• Ice studies
• Introduction – summary of previous talks
• Laboratory studies  - Methodology
• Electron induced chemistry  - some conclusions

• Atmospheric Chemistry
• Urey Miller synthesis
• Plasma studies – ion chemistry
## >140 Interstellar and Circumstellar Molecules

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- **Formic Acid**
- **Acetic Acid**
- **Glycolaldehyde**
- **Glycine**
- **Benzene**
- **Cyanopolyynes**

[http://www.cv.nrao.edu/~awootten/allmols.html](http://www.cv.nrao.edu/~awootten/allmols.html)

National Radio Astronomy Observatory
Astrochemistry

- How is such rich inventory of molecules created?

We have a dusty grain picture
Molecular synthesis on dust grains

An ICY GRAIN of INTERSTELLAR DUST:
A ‘crust’ of ice entirely covers the dust. Dominated by H$_2$O, the ice contains many different chemicals - the least volatile molecules in the top layers.

Forming interstellar ice:
Atoms and molecules stick onto the cold dust, reacting to form H$_2$ and H$_2$O or freezing to form “solids”.

Processing interstellar ice:
Atoms and molecules react at the ice surface to form new, more complex molecules. UV photons and electrons from cosmic rays also “kick start” chemical reactions in the ice. When the ice is heated, many molecules are desorbed.
Building products delivered to planets
Remember reactions can occur in gas phase

First molecules had to be formed without dust (no stars)

And molecules of life may be synthesised in planetary atmospheres
Ion–molecule reactions

Ion-Molecule reactions are a typical example of a reaction that do not require energy input.

e.g.

\[ \text{NH}_3^+ + \text{H}_2 \rightarrow \text{NH}_4^+ + \text{H} \]

\[ \text{Ar}^+ + \text{H}_2 \rightarrow \text{ArH}^+ + \text{H} \]

\[ \text{He}^+ + \text{H}_2 \rightarrow \text{He} + \text{H}^+ + \text{H} \]

\[ \text{H}_2^- + \text{H} \rightarrow \text{H} + \text{H}_2 + \text{e}^- \]
Ion molecule studies

CRESU (Cinétique de Réaction en Ecoulement Supersonique Uniforme) to study neutral-neutral reactions and energy transfer processes in the gas phase down to temperatures as low as ~10 K. (Rennes)
But lets go back to dust ...

Gas phase experiments can not explain all the chemistry in the ISM

E.g. the formation of H$_2$ .... the most common molecule in the ISM can not be formed in the gas phase in quantities required

Instead it is formed by reactions on the surface of little dust grains ...
Chemistry on Dust grains

• Some of these grains are covered with an icy mantle formed by freezing out of atoms/molecules from the gas phase.

• Hence we need to explore ice chemistry!

• The ices in the mantle are bombarded with cosmic rays, ions, solar UV, electrons.
• Chemistry occurs making molecules.
• Desorption back to gas phase.
How to make the ISM in the lab?

- We need to mimic the conditions of the ISM as accurately as we can

**We need**
- Mimic of dust grain surfaces
- ISM is COLD
- ISM is empty
- To mimic radiation sources

- Plus we need methodology to probe results!
Making nothing !!

- Space is empty

- More empty than we can reproduce in the laboratory

- Typically experiments that explore such astrochemistry are performed at pressures of $P \sim 10^{-8} - 10^{-10}$ mbar
  - Still $> a$ million times higher than ISM!

- But why is this a problem ??
Making nothing

• Even at $P \sim 10^{-8} - 10^{-10}$ mbar there is enough residual gas to freeze out on your sample and form a contamination layer!!

• Thus during your experiment you are depositing molecules from vacuum

• Most common contaminants are WATER, CO/CO$_2$ and hydrocarbons - all molecules you may want to explore and can play role in chemistry

• Best vacuum after bake our remove water and dry pumps reduce hydrocarbon content

• Distinguish background CO/CO$_2$ by using $^{13}$C as target carbon (just more expensive)

• It is expensive making nothing and time consuming !!!
Getting (really) cold

• How do make surface cold --- 10K ??

• Use coolant

• LN$_2$ takes you to 77K

• Liquid helium (4K) but not 100% efficient - IR radiation from walls of chamber heat your sample

• Liquid helium is expensive (more than whisky per litre !) world shortage So recycle it
Dust grain mimics

- What are they made of?
  - Carbon?
  - Silicates

One good substrate is
- Olivine \((\text{Mg,Fe})_2\text{SiO}_4\).

- But is non conducting
Dust Grain mimics

- But the grains are small .... micron or nano scale !!

- To date experiments have used bulk samples Cm in size (few, VERY FEW exceptions)

- Is chemistry and ice morphology same on micron/ nano surface as on bulk ?

- How does struture effect diffusion? Desorption ? etc
Ultrasonic dust trap
• IR of water ice on carbon dust in a levititation trap

• Spectra match water ice spectra recorded by AKARI in star forming region.
What about time?

• In space events take time

• 1 molecule absorbed a day/week/month!
• 1 collision every few years!

• *Yet in lab we speed it up!*

• Are ices formed so slowly in space same morphology as ices formed in lab?
• Do collisions scale with fluence - can we replicate synthesis in lab via fluence?

• *We have to assume time is not important is this true?*

• *This requires modelling of molecule by molecule ice film growth*
How do we monitor chemistry?

- **Spectroscopy** --
  - InfraRed (via FTIR or Raman)
  - or Ultraviolet

- If we desorb from surface may use Mass spectrometry

- **TPD Temperature Programmed Desorption**
**OU Portable System:**
- Transmission UV & FTIR Spectroscopy and Processing
- Designed to be transported to central facilities → Synchrotrons, RAL, QUB

**OU Static System:**
- TPD, RAIRS and Processing
- Molecular synthesis with electrons and photons → E-gun, UV lamp

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**Detectors**
- UV-VIS / FTIR detector
- PMT

**Sources**
- UV-VIS / FTIR spectrometer
- Synchrotron
- E-gun, Synchrotron, ECR

**Cryogen inlet via transfer line**
**Temp controller**
**Cryostat**
**To pumping station**
**Thermocouples**
**Copper sample mount**
**MgF2/ZnSe substrate**
**Resistive heater**
**Ion gauge**

**Proposed TSP**
- Rotary feed-through
- Z-axis feed-through

**Sample mount and substrate**
**Gate valve**
**Mag-lev turbo-pump**
**Mass spectrometer**
**Cryostat**

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**300mm**
Energetic Sources

• VUV light

• Ion beams

• Electron beams
Experimental studies of chemical processing of astrochemical ices

First we need to find a mimic of star light!

Stars are fuelled by nuclear reactions

We can’t use these in a laboratory
“The Ultimate Synchrotron Radiation Source”
ISA- Institute for Storage Ring Facilities,
Department of Physics and Astronomy, University of Aarhus

Energy = 580 MeV
Circumference = 45.7 m
Lifetime = Infinite (top-up)
How do we study Cosmic Ray (CR) induced chemistry?

To study CR chemistry we need to:

1. Produce beams of CRs – protons, alpha particles and electrons
2. Accelerate CRs to high energies

Use particle accelerators - Van der Graf Accelerators
Modern version ECRIS Ion source

9.0 – 10.5 GHz Electron Cyclotron Resonance Ion Source at Belfast
Role of electrons?

- To date most processing studies have explored **UV irradiation** - ok for diffuse clouds but dense clouds?

- UV and cosmic rays induce secondary electrons
Cosmic rays as secondary electron source

- Major product of cosmic rays are Secondary electrons and they can induce chemistry

- Indeed one CR may produce an avalanche of $10^4$ electrons whose energy vary from close to CR energy to thermal energy.
So what do we know?

- Experiments are exploring synthesis routes.
Irradiation of $\text{H}_2\text{O}:\text{CO}_2$ ice

Before irradiation
Irradiation of H$_2$O:CO$_2$ ice

After irradiation for 1 hour
Warm-up + Irradiation of H$_2$O:CO$_2$ ice
Warm-up after $H^+$
Irradiation of $H_2O$:CO$_2$
ice
Warm-up after H$^+$
Irradiation of H$_2$O:CO$_2$
ice
Warm-up after $H^+$
Irradiation of $H_2O:CO_2$
ice
Warm-up after $\text{H}^+$ Irradiation of $\text{H}_2\text{O}:\text{CO}_2$ ice
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Irradiation of H₂O:CO₂
ice
Warm-up after H$^+$ Irradiation of H$_2$O:CO$_2$ ice
Warm-up after Irradiation of H$_2$O:CO$_2$ ice
This is Martian ice cap
Electron Induced Chemistry

- Give some examples of how electrons irradiating ice can induce synthesis.

‘Simple’ single ices are not simple!

- Oxygen → to ozone
- Methanol ice → CO and CO$_2$
  H$_2$CO and ‘complex’
Mechanisms for electron induced astrochemistry

Look at irradiation of pure ices of oxygen

Ozone formation
Ozone monomer synthesis

- Inter cluster chemistry!

\[(O_2\ldots O_2) \rightarrow (O_2\ldots O\ldots O) \rightarrow (O_3\ldots O)\]
Ozone monomer synthesis

Temperature dependence;

- Yield largest at lowest Temperature
Ozone complexes

- $O_3 \cdot O$
- $O_3 \cdot O_3$
Co-deposit two isomers
Formation of isotopomers (11K)
Desorption

Mass spectrometric detection of species but

- Heating can induce synthesis eg ozone formation in O₂ ice

- Molecules can fragment upon desorption - mass fragment may not be parent species

![Graph showing column density vs time with temperature program started and stopped points marked.](image)
So can we go on to make building blocks of life?

- How to create an amino acid?
- How to create a sugar in space?
- Synthesis in the ice mantles?
Formation of ethylene glycol in pure methanol ice

HOH$_2$C-CH$_2$OH

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Fig. 6-3: Ethylene glycol was observed after irradiation of pure CH$_3$OH with 1 keV e$^+$ at 30 K and then annealing process.
Formation of methyl formate CH₃OHCO

Fig. 6-9: Irradiation of 1:1 binary mixture of NH₃:CH₃OH with 1 keV e⁻ at 30 K

Fig 6-14: Formation of CH₂OHCO during the irradiation of binary mixture NH₃:CH₃OH with 1 keV e⁻ at 30 K
Create exotic compounds - Ammonium carbamate

Fig. 5-5: IR spectra of NH₃:CO₂ (1:1), (a) post-irradiation (58 min) and after warm-up (220 - 270 K); and (b) comparing Frasco's actual 1964 experimental spectrum at 248 K.
Formation of formamide HCONH$_2$ from irradiation of ammonia/methanol ice
Irradiation of methylamine and carbon dioxide ice makes glycine simple amino acid.

Effects of Irradiation

Figure 3 – Pristine CH₃NH₂ & CO₂ mixture

Figure 4 – 100 minute after irradiation of the mixture
Electron Induced Chemistry - summary

- Experiments reveal;
- Electron irradiation is efficient at molecular synthesis (>UV per event ?)
- Can make complex molecules from simple ‘ingredients’
- Low energy electrons (< photo dissociation energy) meV to 10eV can induce chemistry (via dissociative electron attachment == anion chemistry_so no barriers)
Electron induced chemistry

- Can make larger molecules e.g. methyl formate; ethylene glycol; ammonium carbamate; glycine; all from simple Methanol/ammonia ices

- Strong Temperature and morphology dependence

- Does it play a role in ISM Chemistry Question to answer?
So big question?

- How large are the molecules you can synthesize?
- Can make sugar and amino acids
- RIBOSE (Meinert et al Science 352, pp. 208-212 (2016))
- How do you make the real ‘building blocks of life’? The nuclear bases?
Nucleobases

- Adenine
- Guanine
- Thymine
- Cytosine

- Paired in DNA
- AT
- CG
but there is still much to do /learn

There are known knowns; there are things we know that we know.

There are known unknowns; that is to say, there are things that we now know we don’t know.

But there are also unknown unknowns – there are things we do not know we don’t know.

-Donald Rumsfeld
Route 2 to molecular synthesis;  
Form on the planet  
The UREY MILLER Experiment

\[ \text{CH}_4 + \text{NH}_3 + \text{H}_2 + \text{energy} \rightarrow \text{Glycine} \]

Glycine  
Amino acid
Urey Miller Results

• Urey-Miller was run for a week, liquid was extracted from the flask
• Analysed with paper chromatogram
• 3 Amino acids identified, Glycine, α-alanine and β-alanine
• Hence it is possible to form prebiotic molecules from basic chemistry
Planetary atmospheres

- One area of current and developing research is the study of planetary atmospheres
- Observational studies
- Models (e.g. climate models of Mars and Venus)
Exploring Titan’s atmosphere -- the early Earth?

Cassini-Huygens Mission to Titan
Titan’s atmosphere – chemical composition

Titan’s atmosphere – chemical composition

CH$_4$, C$_2$H$_6$, CO$_2$, $^{40}$Ar detected at surface

Other Possible Species:
- Cyanogen (C$_2$N$_2$)
- Acetylene (C$_2$H$_2$)
- Vinylethylene (C$_4$H$_4$)
- Propene (C$_3$H$_6$)
- Butane (C$_4$H$_{10}$)
- Benzene (C$_6$H$_6$)

Titan’s surface

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<tr>
<td>Acetonitrile</td>
<td>CH\textsubscript{3}CN</td>
<td>5 p.p.b.</td>
</tr>
<tr>
<td>Carbon dioxide</td>
<td>CO\textsubscript{2}</td>
<td>10 p.p.b.</td>
</tr>
<tr>
<td>Cyanoacetylene</td>
<td>HC\textsubscript{3}N</td>
<td>10 p.p.b.</td>
</tr>
<tr>
<td>Methylacetylene</td>
<td>CH\textsubscript{3}C\textsubscript{2}H</td>
<td>5 p.p.b.</td>
</tr>
<tr>
<td>Cyanogen</td>
<td>C\textsubscript{2}N\textsubscript{2}</td>
<td>5 p.p.b.</td>
</tr>
<tr>
<td>Water vapour</td>
<td>H\textsubscript{2}O</td>
<td>8 p.p.b.</td>
</tr>
<tr>
<td>Diacetylene</td>
<td>C\textsubscript{4}H\textsubscript{2}</td>
<td>1 p.p.b.</td>
</tr>
</tbody>
</table>
Question

- What organic chemistry may be prevalent?

- Can prebiotic molecules form in a Titan atmosphere?
Use laboratory plasma/discharge as mimic of planetary atmosphere

- Study discharge in $\text{N}_2/\text{CH}_4$
- Make Titan dust analogues
- Detect anions
Molecular formation in glow discharge

Form main hydrocarbons and nitriles seen in Titan atmosphere
Neutral chemistry
Optical Emission --
FTIR –or GC-MS
Neutral Chemistry – GC-MS

Ethane, ethene, cyanogens, propene, propane, propyne, propadiene, butenyene, butadiene, butadiyne, Acetonitrile, propenenitrile, benzene and toluene.
What about anions in Titan atmosphere?

The anion spectrum recorded by the Cassini-Huygens mission at an altitude of 953 km (Coates et al. (2007)).
Experimental set-up for exploring anion formation in point-to-plane corona discharge
Mass spectra of negative ions extracted from point-to-plain corona discharges.

Q = 100 ml/min
I = 0.8 mA
U = 2.7 kV
Anions in the discharge

The detection of CN-, CH₂CN-, C₃N-, CH₂CN- and C₅N- anions provides good evidence of the presence of HCN, CH₃CN, HC₃N, and HC₅N neutrals.
Anions in Titan’s atmosphere

![Graph showing energy/charge and relative product yield]
Future Research/Challenges in astrochemistry

- **Reproduceability**
- Different experiments do not agree on molecules synthesized and/or concentrations of species produced.

- **Cross Sections/rate constants**
- Ill defined and hence hard to produce data for models

- **Same ingredients mixed and bake make different cakes**
Future Research/Challenges in astrochemistry

- Do experiments mimic nature?
- ISM surface chemistry is on micron sized dust grains - is this replicated in bulk ice films?
- Time – do laboratory experiments replicate the slow, unimolecular events of space?
- Planetary atmospheres – How to make a real mimic (no walls) Terrestrial atmospheric chambers
Future Research/Challenges in astrochemistry

- Understand the physics/chemistry
- Role of (secondary) electrons and electron induced mechanisms compared to direct ion bombardment and UV absorption
- Morphology – Surface cluster chemistry and three body reactions
- Synthesis when/where – Direct or through diffusion of radicals (In-situ spectroscopy vs TPD results)
Future Research/Challenges in astrochemistry

- How do we know we have ‘got it right’
- Experiments tested against/predict observations
- ALMA spatial chemical maps.
Future Research/Challenges in astrochemistry

- **Towards biology**
- **Energetic processing of**
  prebiotic and biological molecules
- **Look for routes of**
  formation and stability

![Graph showing the destruction of OCN during the irradiation of NH₃:CO₂ (1:1 ratio).](image)
Future Research/Challenges in astrochemistry

astrobiology

- How do prebiotic molecules assemble to form biomolecular systems e.g. DNA
- Role of the host planet?
- Is life inevitable or rare?
Core Questions for Astrochemistry & Astrobiology

• Where are the building blocks of life synthesised? ISM or on planet?

• Are the conditions for such synthesis common/universal?

• European Astrobiology Roadmap. Published 2016
and finally thanks to.. 

- Bhala Sivaraman
- Binu Nair
- PRL India