Reproducibile Science in the Neutron Reflection Context

ORSO Workshop 2021

Stuart Prescott

School of Chemical Engineering
UNSW Sydney

s.prescott@unsw.edu.au

@SWPrescott

Slides: `https://deb.li/orso2021`

Reproducibility and neutron reflectometry

Slides: `deb.li/orso2021`

What do we mean by reproducible science

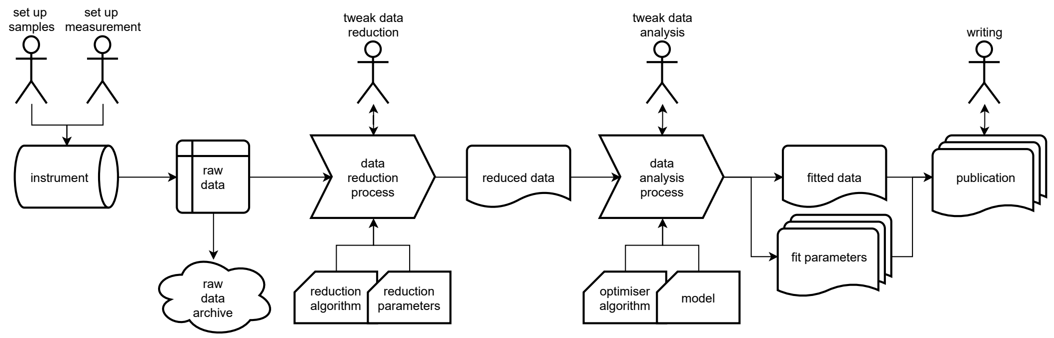
- Transparency in data collection, processing and analysis methods, and derivation of outcomes.
- Publicly available data and associated processing methods.
- Transparent communication of results.

US NSF definition

…reproducibility refers to the ability of a researcher to duplicate the results of a prior study using the same materials as were used by the original investigator. That is, a second researcher might use the same raw data to build the same analysis files and implement the same statistical analysis in an attempt to yield the same results…. **Reproducibility is a minimum necessary condition for a finding to be believable and informative.**

`http://openscience.org/what-exactly-is-open-science/`

What do we mean by reproducible NR?



Can the experiment be repeated?
What are the samples?
How was the instrument used?
Can the raw data be reused?

How is the reduced data made?
Is the reduced data available?

Can the model be reused?
Is the data analysis robust?

Why is reproducibility important for reflectometry

accidents
- mix up run numbers
- reduced data against wrong direct beam
- background subtraction wrong
- auto-reduction done wrong thing

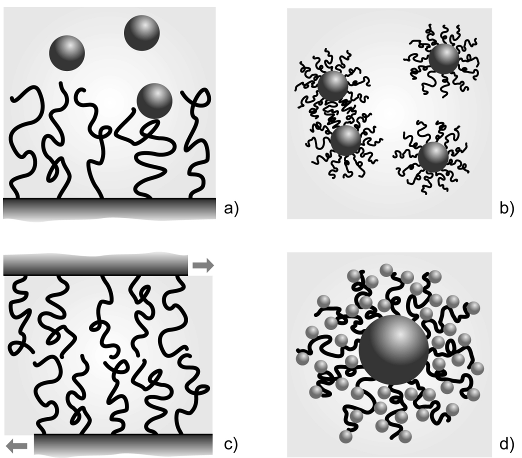
training for new users
- how long does measurement take?
- what footprint/slits/etc are needed?

model reuse
- models are very reusable
- we benefit from access to models
- assists training the next student

data reuse
- reanalyse old data in new study
- next student builds on existing work
- neutrons are scarce, can't just rerun samples
- writing a review paper

data integrity
- clear data pipeline → data hygiene
- prevent data manipulation

Underlying question: what do polymers do in small geometries?



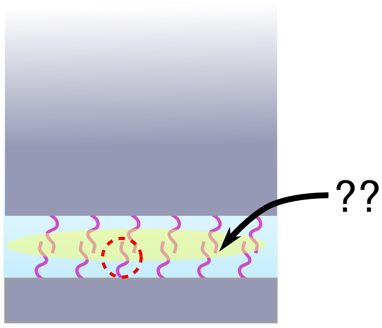
Surface coatings for
- anti-fouling
- stablisation
- lubrication
- selective adsorption

Zooming in on the interface

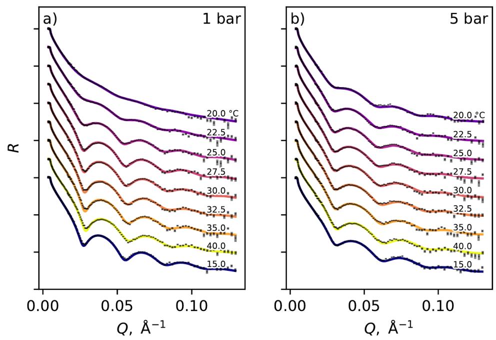
Understanding is poor as interfaces approach to separations about the size of a polymer molecule.

Free draining

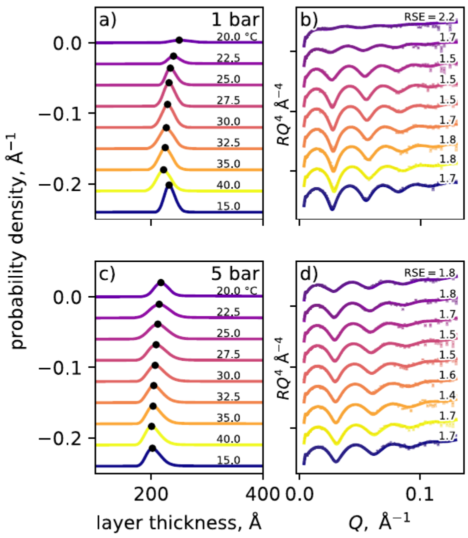
- confinement ≠ pressure
- not hydrostatic pressure
- solvent can escape



What sorts of things to we learn?



Geometric confinement modulates the thermoresponse of PNIPAM
Gresham et al., *Macromolecules*, **2021**, 54, 2541 10.1021/acs.macromol.0c02775



An informal survey of reproducibility

How has practice changed over time?

- these are just ‘typical’ papers for the time
- these are all in good journals
- following (or exceeding) standard practice
- these are all about polymer brushes so as to be somewhat comparable

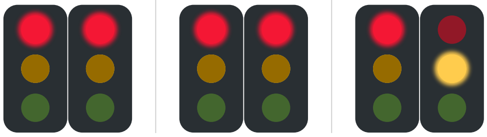
1 “2010 paper” 10.1063/1.4767238
de Vos, WM; Mears, LLE; Richardson, RM; Cosgrove, T; Dalgliesh, R; Prescott, SW
Rev. Sci. Instrum., **2012**, 83, 113903

2 “2015 paper” 10.1021/ma502246r
Abbott, SB; de Vos, WM; Mears, LLE; Cattoz, B; Skoda, MWA; Barker, R; Richardson, RM; Prescott, SW
Macromolecules, **2015**, 48, 2224

3 “2021 paper” 10.1021/acs.macromol.0c02775
Gresham, IJ; Humphreys, BA; Willott, JD; Johnson, EC; Murdoch, TJ; Webber, GB; Wanless, EJ; Nelson, ARJ; Prescott, SW
Macromolecules, **2021**, 54, 2541

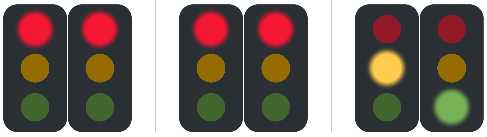
Experimental details in the paper, supp info or references

	2010	2015	2021
Can the experiment be repeated?			
- instrument specified	✓	✓	✓
- measurement times	✗	✗	✗
- footprints, slits, choppers etc	✗	✗	✗
Can the raw data be reused?			
- proposal numbers	✗	✗	✓
- run numbers	✗	✗	✗
- mapping from run numbers to samples	✗	✗	✗
- raw data available?	✗	✗	✗✓



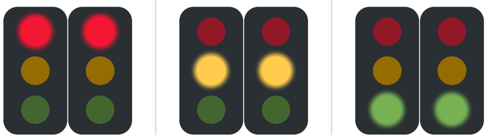
Data reduction details in the paper, supp info or references

	2010	2015	2021
What was done to the data?			
- run number matched to direct beam	✗	✗	✗
- details of reduction published / software	✗✓	✗✓	✗✓
- wavelength ranges used	✓	✓	✓
- handling of overlap/stitch	✗	✗	✗
- parameters for beam/background finding	✗	✗	✗
- binning details	✗	✗	✓
- entirely algorithmic reduction	✗	✗	✗✓
Is the reduced data available for reuse?	✗	✗	✓



Data analysis details in the paper, supp info or references

	2010	2015	2021
Can the model be reused?			
- description of model in words	✓	✓	✓
- code for model	✗	✗	✓
- optimisation details published	✗	✓	✓
Is the data analysis robust?			
- fitting is unique / multimodality checked	✗	✓	✓
- (un)certainty in the model	✗	✗	✓



Detailed view of "2021 paper": experimental details

Neutron Reflectometry. All NR experiments were carried out at the OPAL 20 MW reactor (ANSTO, Sydney, Australia) on the PLATYPUS time-of-flight reflectometer.[30] Specular reflectivity measurements were made at angles of 0.65 and 3.2° for dry samples, 0.8 and 3.8° for unconfined solvated measurements, and 0.35 and 1.5° for confined solvated measurements. An upward reflecting geometry was employed for dry and free-solvent measurements, while confined measurements were conducted in a downward reflecting geometry. These angles resulted in Q ranges of 0.008 to 0.28, 0.009 to 0.31, and 0.004 to 0.13 $Å^{-1}$, respectively.

- ✓ angles
- ✗ slits
- ✗ exposure times

Next steps...

- include addition info in paper / supp info
- the user needs help to know what the important info is
- peer review needs to ask where the info is

Detailed view of "2021 paper": data reduction details

Data reduction followed the standard procedure for PLATYPUS, with a final resolution of $\Delta Q/Q = 8.8\%$;[30] previous experiments have shown this resolution to be sufficient for similar layers.[2,31]

- ✗ doesn't say software was `refnx`
- ✗ beam finder / centre refinement details
- ✗ background subtraction details
- ✗ auto or manual?

The omissions belie significant progress:

- ✓ batch reduction in Jupyter notebook
- ✓ reproducible, automated reduction
- ✓ clear record does exist (just not published!)

Reader doesn't know that method used has been published: Gutfreund et al. *J. Appl. Cryst.* **2018**, 51, 606. 10.1107/S160057671800448X

Next steps...

- publish "standard" reduction info for each instrument (kept up to date): ORSO, arXiv
- tooling produces easy to publish record of sample name, run number, direct beam, and reducer parameters
- include these metadata in the supp info
- peer review needs to ask where the info is

Detailed view of "2021 paper": data analysis details

Data Analysis. The dry brush was modeled as a single slab, while unconfined solvated brushes were modeled by a freeform spline, both following our established analysis protocol.[31,33] The confined data were analyzed using a thickness distribution model with an interfacial volume constraint, implemented in Python through the *refnx* reflectometry analysis package.[26] This model was created to support The full implementation details are included in the Supporting Information.

supp info

1.4 Modeling the confined brush for NR

We start with the oft-utilized[1,5,6,8] space-filling two-component assumption for the confined polymer film:

$$\phi_{\mathrm{poly}} + \phi_{\mathrm{water}} = 1 \tag{S2}$$

$$\rho_{\mathrm{layer}} = \phi_{\mathrm{poly}}\rho_{\mathrm{poly}} + (1 - \phi_{\mathrm{poly}})\rho_{\mathrm{water}} \tag{S3}$$

where ϕ_{poly} and ϕ_{water} are the volume fractions of polymer and water in the layer, and ρ_{layer}, ρ_{poly} and ρ_{water} are the SLDs of the confined layer, pure polymer and pure water respectively. As in previous work[1,5,6,8] we make the further (implicit) assumption that the molar volume of polymer is constant upon solvation:

$$\phi_{\mathrm{poly}} = \frac{d_{\mathrm{dry}}}{s_{\mathrm{SM}}} \tag{S4}$$

where d_{dry} is the thickness of polymer in the 'dry' ($\phi_{\mathrm{poly}} = 1$) state and s_{SM} is the distance between the silica substrate and the confining membrane. To account for the water content of PNIPAM at ambient conditions, the thickness of the polymer in the $\phi_{\mathrm{poly}} = 1$ state was taken as 90 % of the measured dry thickness. The water fraction of dry PNIPAM films

- ✓ short description in paper with more details
- ✓ published code base used for analysis
- ✓ supp info has *lots* more
- ✓ description in words plus actual equations used

Detailed view of “2021 paper”: data analysis details

1.4.1 Programmatic implementation of the distribution function

Written in Python 3.6.[i]

```python
import numpy as np
from scipy.stats import gamma

def dist_pdf(z, loc, scale, a, tail=0, tail_len=0):
    """
    Separation probability density function for confinement cell
    modelling. It is produced by the normalised summation of a
    typical gamma distribution and a custom tail distribution.

    Parameters
    ----------
    z : numpy.array
        Separations at which the function is evaluated.
    loc : float
        The separation at which the gamma-component starts.
    scale : float
        The width of the gamma-component of the distribution.
    a : float, optional
        The shape parameter for the gamma-component of the
        distribution. Lower a results in 'exponential' PDFs,
        whilst higher a results in more 'normal' PDFs.
    tail : float, optional
        The weighting of the tail-component. Higher values result
        in a higher tail. Values of zero result in no tail.
    tail_len : float, optional
        The length of the tail-component.

    Returns probability density at separations provided.

    """
    pdf1 = gamma.pdf(z, loc=loc, scale=scale, a=a)

    tpeak = loc + (a-1)*scale

    # End at tail length + gamma mode
    tcut = tail_len + loc + (a-1)*scale
```

This document has embedded files. Click here to see them

- ✓ description in words plus actual equations used
- ✓ exact code of the model included in PDF
- ✓ exact code of the model attached to the PDF

Current Practice

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Detailed view of “2021 paper”: data analysis details

Notes

The authors declare no competing financial interest.
The reduced NR data files are published on Zenodo at DOI: 10.5281/zenodo.4324395.

Neutron reflectometry datasets for "Geometrical confinement modulates the thermoresponse of a poly(N-isopropylacrylamide) brush"

Gresham, Isaac J.; Humphreys, Ben A.; Willott, Joshua D.; Johnson, Edwin C.; Murdoch, Timothy J.; Webber, Grant B.; Wanless, Erica J.; Nelson, Andrew R.J.; Prescott, Stuart W.

Reflectometry datasets collected from confined and unconfined polymer brushes on the Platypus reflectometer at ANSTO.

The reflectometry data files provided are four-column text files. Column 1 is Q, column 2 is R, column 3 is R error, and column 4 is Q error (resolution).

Files (632.8 kB)

Name	Size	
0bar 15C post-confinement.dat md5:9eade893611dee08474d09506f13fe3d	18.4 kB	Download
0bar 15C pre-confinement.dat md5:d763b8caa0a23da5d8742ee511e2ff10	18.4 kB	Download
0bar 20C pre-confinement.dat	18.4 kB	Download

- ✓ link to Zenodo record for data included in paper
- ✓ all reduced data included in Zenodo record

Current Practice

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Another recent example of excellent practice

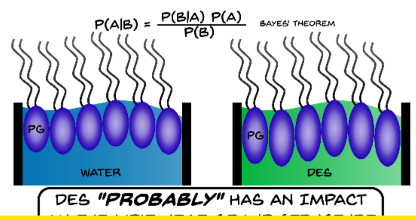
README.md

ESI for "Bayesian determination of the effect of a deep eutectic solvent on the structure of lipid monolayers"

publication DOI 10.1039/C9CP00203K | DOI 10.5281/zenodo.2577796 | arXiv 1810.07616 | license CC-BY-SA-4.0

Andrew R. McCluskey*, Adrian Sanchez-Fernandez, Karen J. Edler, Stephen C. Parker, Andrew J. Jackson, Richard A. Campbell, and Thomas Arnold*.

*a.r.mccluskey@bath.ac.uk/andrew.mccluskey@diamond.ac.uk & tom.arnold@esss.se



- ✓ link to Zenodo record for data included in paper
- ✓ all reduced data included in Zenodo record
- ✓ GitHub repository with
  - all the reduced data
  - all the Python files for the model
  - all the Jupyter notebooks
  - instructions on how to run the analysis

Current Practice

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Jupyter: better than an unconstrained GUI but not a silver bullet

Jupyter is excellent at helping explore data

- interactive
- can see input, process, output, analysis all in one place
- mid-way point between a GUI and code/batch procss

However...

- not human readable file format (JSON with encoded binary blobs for images)
- file format details have changed in the past; reliance on compatibility is a big risk
- cells can be executed out of order or edited after execution
- desire to rereun it all at the end can be CPU-expensive
- difficult to push off to HPC for bigger analyses

(see work of McCluskey et al. to turn ipynb files into Python files as a better record of the final code)

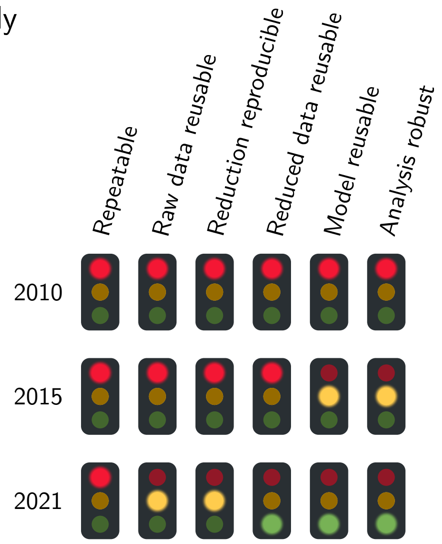
Reproducibility: the good, the bad and the ugly

the good: data analysis

- current good practice gives highly reproducible analysis
- tooling is helping
- still sources of friction
- we need to start asking for this in peer review

the bad: data transformations

- metadata lost in transformations
- tooling is **not** helping
- file formats are **not** helping
- automated part of data reduction is under-specified
- manual part of data reduction is unspecified



Conclusions

1. Can improve the experimental description
   – we can do this today
   – reviewers should start asking for this today
2. Can improve the model description
   – we can do this today
   – reviewers should start asking for this today
3. Need "white paper" standard reduction descriptions that can be cited
   – we can do this 'tomorrow'
   – reviewers can start asking for this soon
4. Need tooling and file formats to improve the reproducibility of the reduction process
   – this will take some time
   – reviewers can start asking for this in the future

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Useful resources for those wanting to read more:

- `http://openscience.org/what-exactly-is-open-science/`
- `https://www.earthdatascience.org/courses/intro-to-earth-data-science/open-reproducible-science/`
- What does research reproducibility mean? Goodman, SN; Fanelli, D; Ioannidis, JPA. *Science Translational Medicine*, **2016**, 8(341) 341ps12. DOI: 10.1126/scitranslmed.aaf5027
- A manifesto for reproducible science. Munafò, MR et al. *Nature Human Behaviour*, **2017**, 1, 0021. DOI: 10.1038/s41562-016-0021