Using Engineering and Particle Physics in the Advancement of Green Chemistry

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Introduction

The field of green chemistry aims to make chemical processes less wasteful and hazardous to both humans and the environment. Solvent waste is a leading source of (hazardous) industrial waste.

We are interested in the study of two green solvents: liquid and supercritical CO_2. Fluid CO_2 solvents are reusable and easily removed from a reaction. Additionally, $scCO_2$ has tunable physical properties.

Objectives

Previous experimental data of vinylidene fluoride polymerization shows high rate tunability in the liquid CO_2 range. Also, considerably slower reaction rates were seen in low-density $scCO_2$. To further investigate these phase ranges, our objective was to design and build a new reaction system.

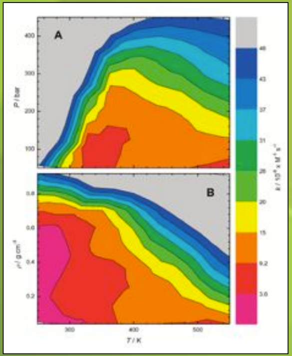

Figure 1: Thermodynamic rate map of Mu + VDF in CO_2 [1].

Method – TF-μSR

μSR is the only technique available to study H atoms in fluid CO_2. Spin polarized muons are fired into the sample to probe hydrogen, and their decay data gives information on reaction kinetics and formed free radicals.

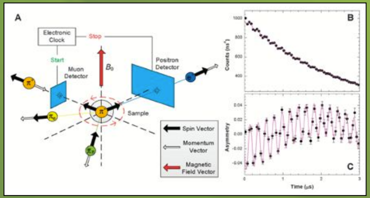

Figure 2: Standard TF-μSR experiment and data.[1]

Design Requirements

To investigate both liquid CO_2 and low-density $scCO_2$, a new reaction system design was proposed based on the following requirements:

1) *Cell temperature must be adjustable between -10°C and 200°C.*
2) *Must have large internal volume to allow for low-density experiments.*
3) *Minimize (electro)magnetic components.*
4) *Window must be thin enough to allow passage of muons, but thick enough to withstand high pressure up to 300 bar.*

Strategy & Design

Our design features removable heating/cooling sheaths and a one-piece window design of 2.0mm thickness. Dimensions were optimized using SRIM, a Monte Carlo simulation of ion thermalization paths.

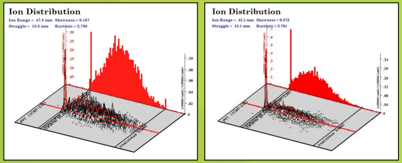

Figure 3: SRIM generated 3D ion range plot of 26 MeV muons in 0.1 g/cm^3 $scCO_2$ with 1.9mm (left) and 2.0mm (right) windows.

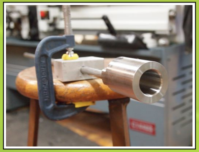

Figure 4: Vessel Body, Support.

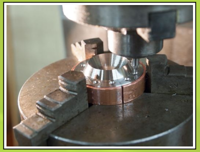

Figure 5: Vessel Window

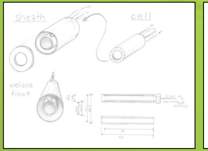

Figure 6: Cooling Sheath

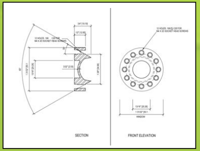

Figure 7: Vessel Window

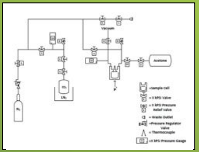

Figure 8: Gas Flow System

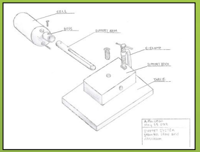

Figure 9: Cell Support Arm

Engineering Analysis

To ensure a safe and functional system, a stress distribution was calculated for our design. A finite element analysis program was used in the analysis.

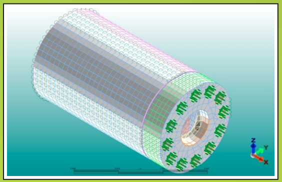

Figure 10: FEA Model of Vessel

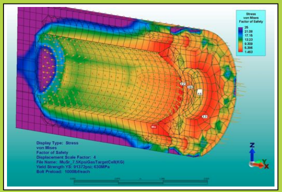

Figure 11: Vessel Cross-section Stress Distribution

Proof of Concept

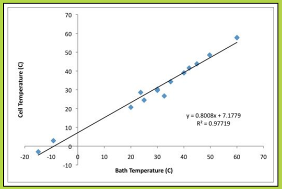

Figure 12: Circulator Calibration with Various CO_2 Densities

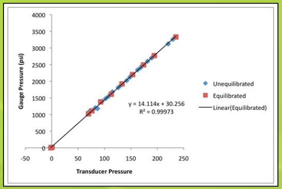

Figure 13: Pressure Transducer Calibration with Mechanical Gauge

Figure 14: Pressure Test Setup

Figure 15: Assembled High-Pressure Vessel

The vessel was successfully tested up to 7100 psi and 200°C without leaks.

Conclusions

We have designed and built a high-pressure reaction vessel and system with dimensions optimized using Monte Carlo simulation. Through offline proof of concept experiments, we have so far proven that our vessel is leakproof and functioning properly. The temperature and pressure control systems were also calibrated for future use.

Future Work

Before our group can examine rate tuneability in liquid CO_2 and low-density supercritical CO_2, the online proof-of-concept experiments must be completed. The goals of these experiments are to:

1) Observe a muon and muonium in situ.

2) Show the momentum at which we have minimal stopping in the window/cell body.

3) Compare TF-μSR experiments in unpurified and purified CO_2.

4) Show that relaxation increases with change of T or P upon the addition of acetone.

Acknowledgements

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References

1. *Khashayar Ghandi et al. Phys. Chem. Chem. Phys., 2012, **14**, 8502-8505.*