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Measurement and Modeling of Transport Properties of Low-GWP Refrigerant Mixtures

(Outcomes of NEDO Project in Japan)

Akio MIYARA, Monjur MORSHED, Md. Jahangir ALAM
Atiqur R TUHIN, Silvia SUDIRMAN

Department of Mechanical Engineering, Saga University

佐賀大学
SAGA UNIVERSITY

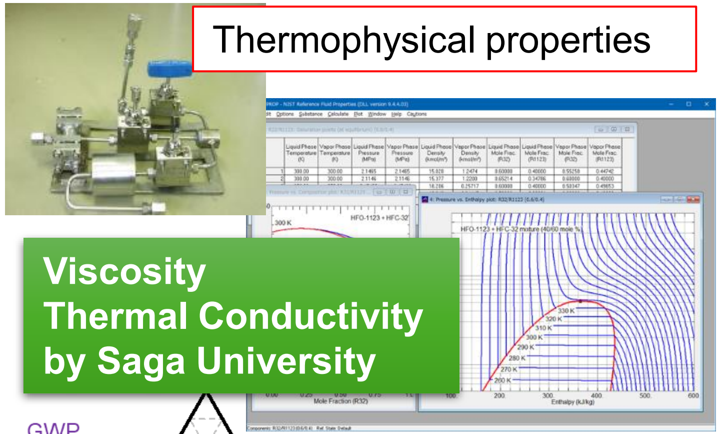
Outline of NEDO Project from 2018 to 2022

Development of Technology and Assessment Techniques for Next-Generation Refrigerants with a Low GWP Value

https://www.nedo.go.jp/english/activities/activities_ZZJP_100140.html

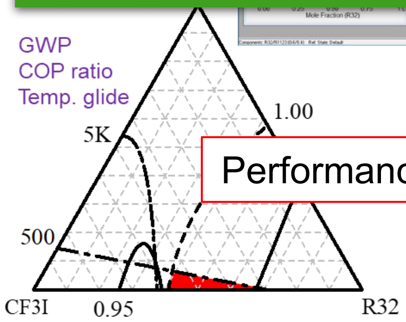
Refrigerants

Thermophysical properties

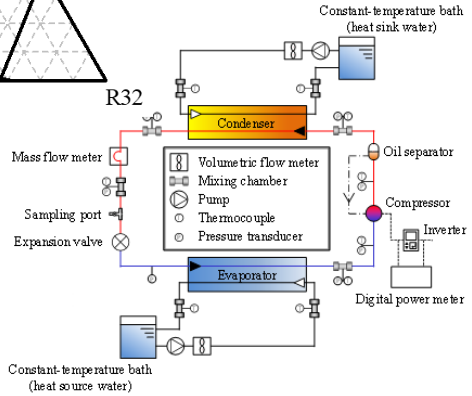


Viscosity
Thermal Conductivity
by Saga University

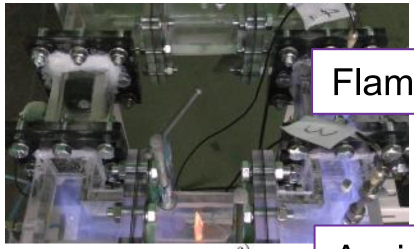
GWP
COP ratio
Temp. glide



Performance evaluation



Safety evaluation
Risk assessments



Flammability evaluation

Accident scenarios

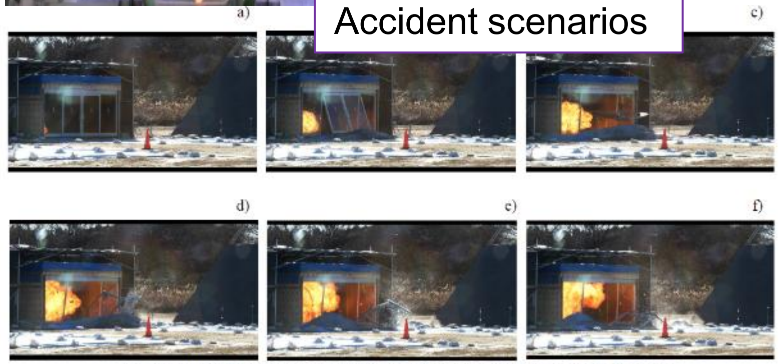
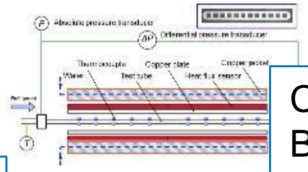
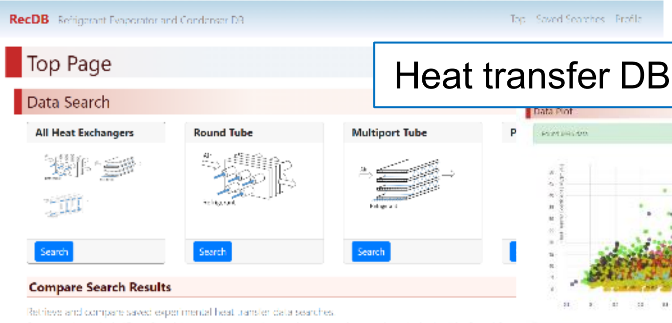


Fig. 4-11 Glass door behavior observed after propane gas ignition (200 ms interval).

Equipment



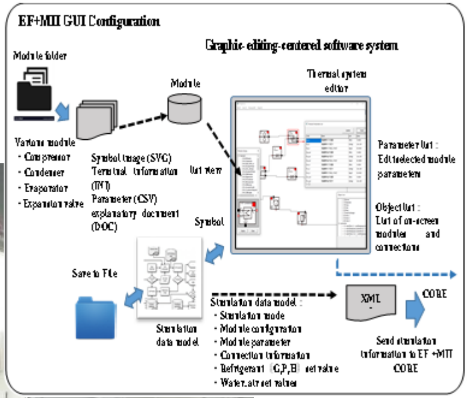
Condensation
Boiling



Heat transfer DB

Plate Heat Exchanger
Heat Transfer Database
by Saga University

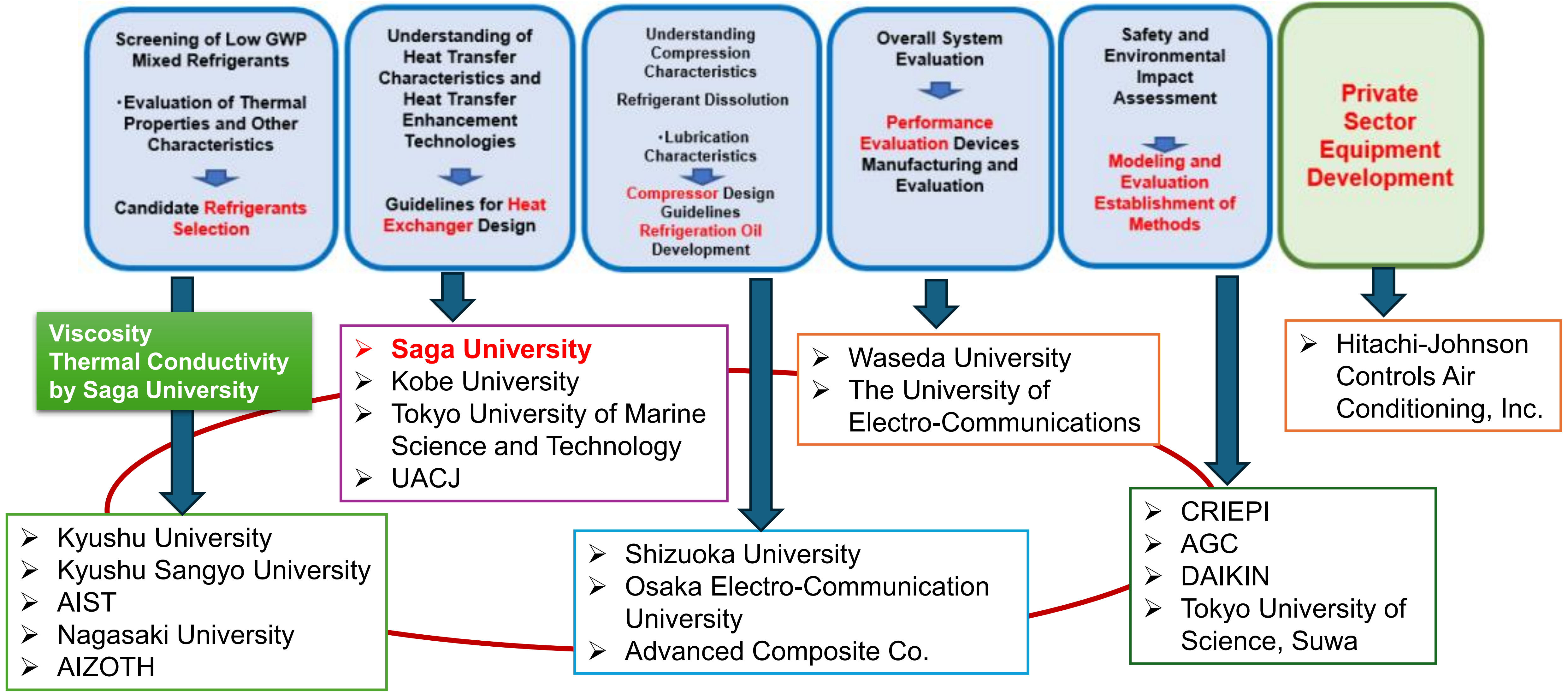
System evaluation



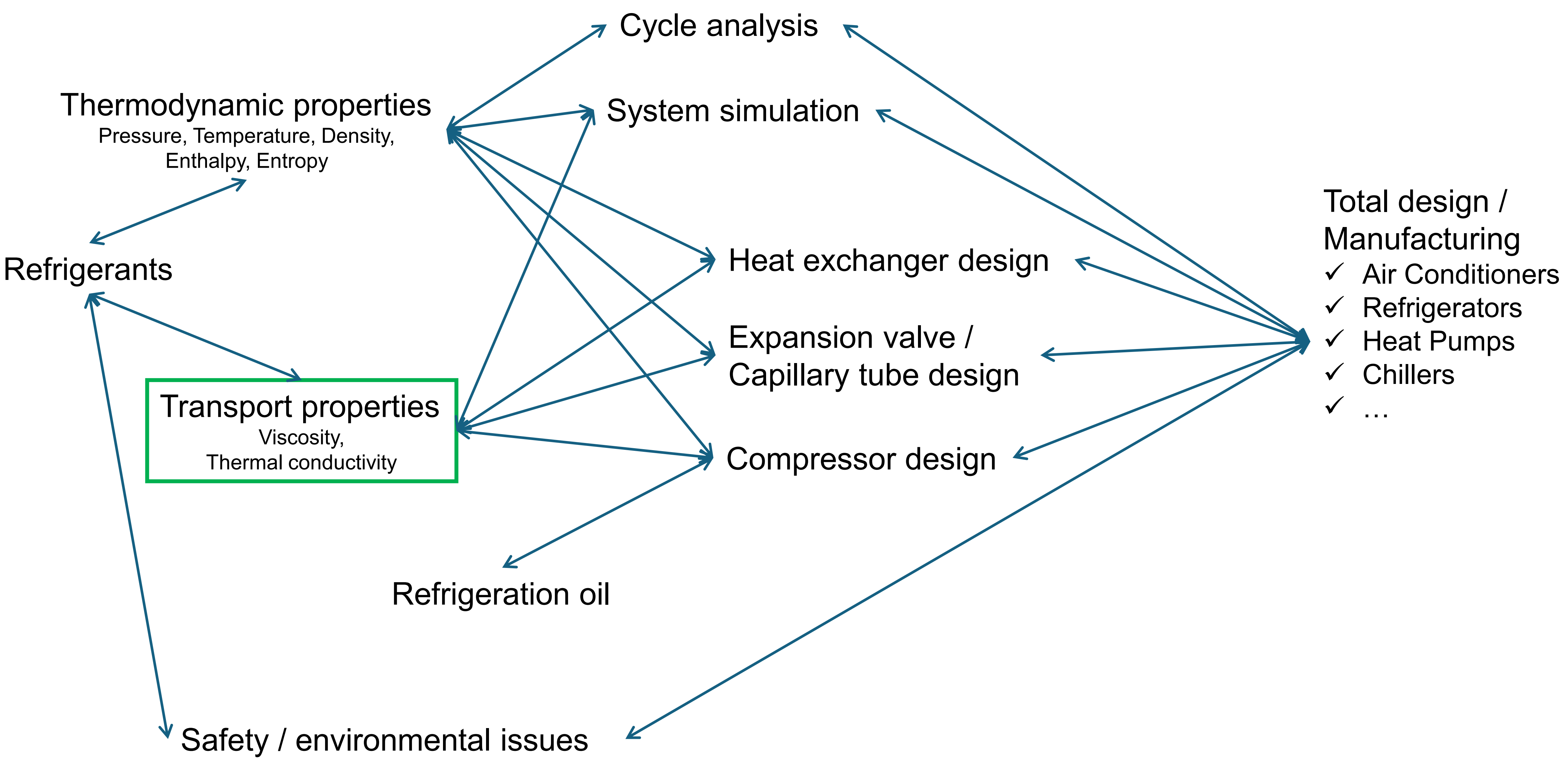
Outline of NEDO Project from 2023 to 2027

Development of Refrigeration and Air-Conditioning Technologies for Practical Use of Next-Generation Low-GWP Refrigerants

https://www.nedo.go.jp/english/activities/activities_ZZJP_100244.html



Refrigerants to Total Design/Manufacturing - Role of transport properties -



Target Refrigerants - Needs for HFO-based low-GWP refrigerants -

Natural refrigerants
CO_2, HCs, NH_3,

HFO-based low-GWP refrigerants
(pure, mixture)

Household refrigerators and freezers

Room air conditioners
(household A/C units)

Cold storage warehouses
(refrigerated and frozen storage facilities)

Heat pumps for swimming pool heating

Packaged air conditioners
(for shops, offices)

Food processing refrigeration equipment
(ice cream machines, blast freezers)

Heat pump food dryers
(for agricultural and marine products)

Ground-source heat pumps

Commercial refrigerators and freezers
(showcases, chest freezers, etc.)

Central air-conditioning systems
(chillers + air handling units)

Variable refrigerant flow systems
(VRF, multi-split for buildings)

Ice makers

Automotive air conditioners

Industrial refrigeration systems
(chemical plants, low-temperature laboratories)

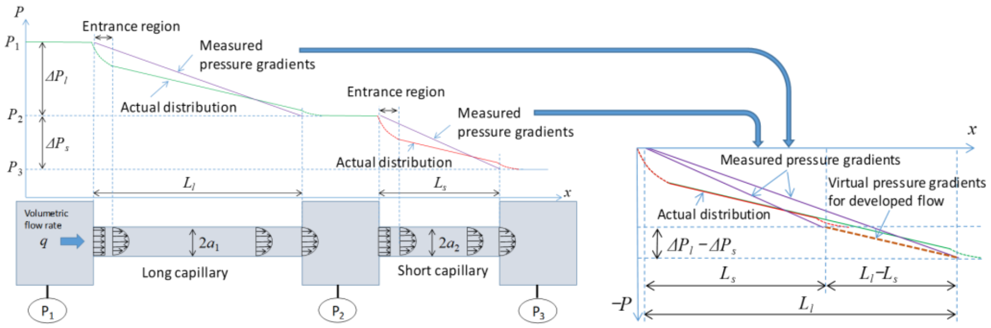
Heat pump water heater
(Air-to-water, Water-to-water, etc.)

Special-purpose air conditioners
(clean rooms, hospitals, server rooms)

Industrial process heat pumps
(waste heat recovery, process heating in factories)

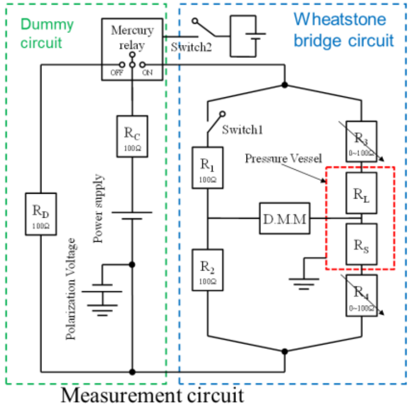
Measurement of Transport Properties

Viscosity Measurement - Tandem capillary tube method -



Hagen-Poiseuille theory	Correction of end effects	Cancel of end effects	Correction for diameter difference
$\eta = \frac{\pi a^4 \Delta P}{8qL}$	$\eta = \frac{\pi a^4 \Delta P}{8q(L+na_l)} - \frac{m\rho q}{8\pi(L+na)}$	$\eta = \frac{\pi a^4 (\Delta P_l - \Delta P_s)}{8q(L_l - L_s)}$	$\eta = \frac{\pi (a_l^4 \Delta P_l - a_s^4 \Delta P_s)}{8q(L_l - L_s)}$

Thermal Conductivity Measurement - Transient hot wire -



Measurement circuit

Thermal Conductivity as;

$$\Delta T(t, r_o) = \frac{Q}{4\pi\lambda}\left(\ln t + \ln \frac{4\alpha}{r_o^2 C}\right)$$

Differentiation by logarithmic time

$$\lambda = \frac{Q}{4\pi} \Big/ \frac{d(\Delta T)}{d(\ln t)} = \frac{Q/4\pi}{dT/d\ln t} = \frac{Q}{4\pi}\left(\frac{dE/dT}{dE/d\ln t}\right) = \frac{Q}{4\pi}\cdot\frac{dE}{dT}\Big/\frac{dE}{d\ln t}$$

$$Q = \frac{RI^2 t}{lt} = \frac{R_{l,0} + R_{s,0}}{(l_l + l_s)}\left(\frac{V_b}{R_3 + R_{l,0} + R_4 + R_{s,0}}\right)^2$$

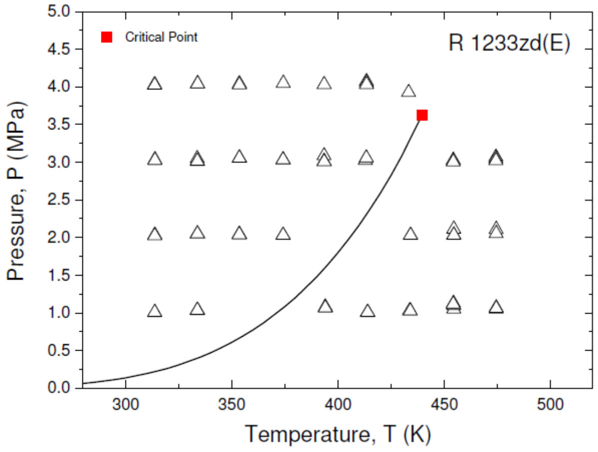
$$E = V_b\left(\frac{R_3 + R_L}{(R_3 + R_L) + (R_4 + R_S)} - \frac{R_1}{R_1 + R_2}\right)$$

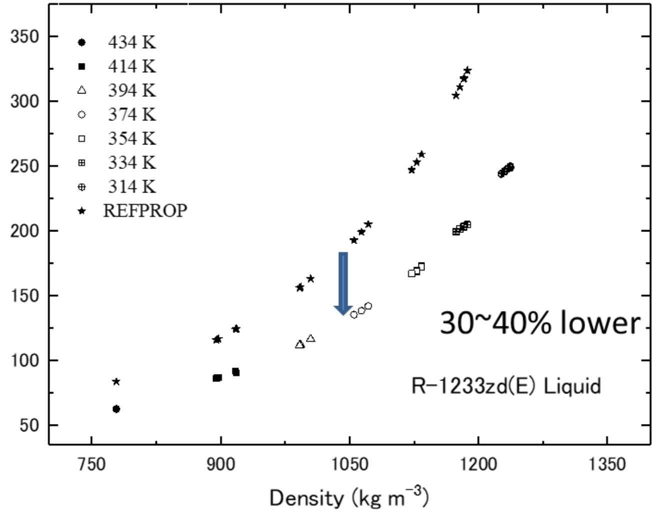
Therefore,

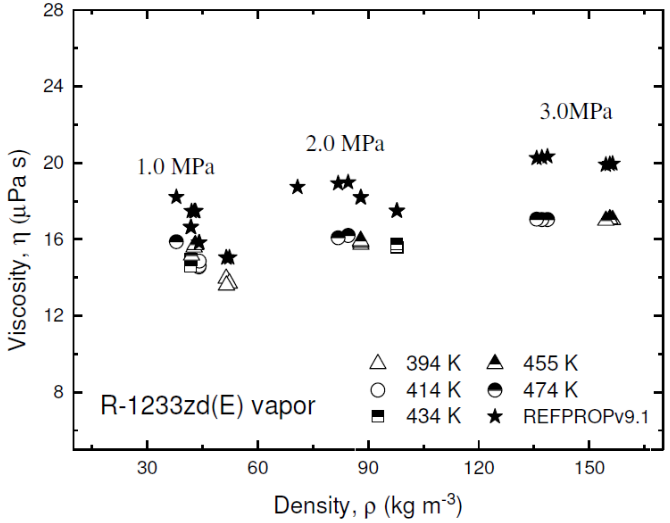
$$dE/dT = V_b \frac{d}{dT}\left(\frac{R_3 + R_L}{(R_3 + R_L) + (R_4 + R_S)}\right)$$

$$= \frac{V_b}{(R_3 + R_L) + (R_4 + R_S)}\left(\frac{dR_L}{dT} - \left(\frac{dR_L}{dT} + \frac{dR_S}{dT}\right)\frac{R_3 + R_L}{(R_3 + R_L) + (R_4 + R_S)}\right)$$

$$\lambda = \frac{R_{l,0} + R_{s,0}}{4\pi(l_l + l_s)}\left(\frac{V_b}{R_3 + R_{l,0} + R_4 + R_{s,0}}\right)^2 \frac{V_b}{R_3 + R_l + R_4 + R_s}\left(\frac{dR_l}{dT} - \left(\frac{dR_l}{dT} + \frac{dR_s}{dT}\right)\frac{R_3 + R_l}{R_3 + R_l + R_4 + R_s}\right)\Big/\frac{dE}{d\ln t}$$







Measured Refrigerants

Name	Property	Validation	On going	Published	High temp. side	Low temp. side
R134a; R32	Viscosity and Thermal C.	√				
R1336mzz(E); R1233zd(E);	Viscosity and Thermal C.			√ [1, 2, 3, 4]	√	
R356mmz; R1224yd(Z); R1336mzz(Z)	Viscosity and Thermal C.			√ [5, 6, 7, 8]	√	
R1234ze(Z); 3,3,4,4,5,5-HFCPE; CF_3I	Viscosity and Thermal C.			√ [9, 10, 11, 12, 13]	√	
R1123+R32	Viscosity			√ [14]	√	√
R1132(E)	Viscosity			√ [15]	√	√
R1132(E)	Thermal C.		√			
R474A (R1132(E)+R1234yf)	Viscosity		√		√	
R474A (R1132(E)+R1234yf)	Thermal C.		√			
R454A	Viscosity and Thermal C.		√			
R454B	Viscosity			√ [16]		
R454B	Thermal C.		√			
R454C	Viscosity			√ [16]		
R454C	Thermal C.		√			
R1123	Viscosity		√			
R1123	Thermal C.			√ [17]		√

Calculation of Transport Properties - ECS model for viscosity -

Viscosity Calculation Model

$$\eta(T,\rho) = \eta_d(T) + \Delta\eta_r(T,\rho) + \Delta\eta_c(T,\rho)$$

$\Delta\eta_c \rightarrow 0$

Dilute gas term:

Chapman-Enskog theory

$$\eta_d(T) = F_{Chung} \frac{C\sqrt{MT}}{\sigma^2 \Omega^{(2,2)}}$$

ECS model - for Residual Viscosity -

$$\Delta\eta_r(T,\rho) = \Delta\eta_{r,ECS}(T,\rho) = \Delta\eta_0(T_0,\rho_0) \cdot F_\eta(T,\rho)$$

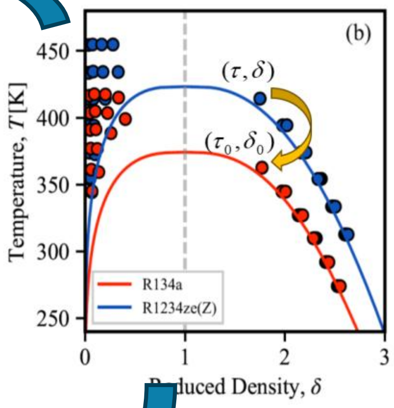
Target fluid residual viscosity

Reference fluid residual viscosity (viscosity of R134a)

Factor of ECS model

$$\alpha_0^r(\tau_0,\delta_0) - \alpha^r(\tau,\delta) = 0$$

$$Z_0(\tau_0,\delta_0) - Z(\tau,\delta) = 0$$



$$F_\eta(T,\rho) = f^{1/2} h^{-2/3} \left(\frac{M}{M_0}\right)^{1/2}$$

$$f = \frac{T}{T_0} \quad h = \frac{\rho_0}{\rho} \quad \tau = \frac{T_{crit}}{T} \quad \delta = \frac{\rho}{\rho_c}$$

Fitting with experimental data

$$\Delta\eta_r(T,\rho) = \Delta\eta_{r,ECS}(T,\rho) = \Delta\eta_0(T_0,\rho_\eta) \cdot F_\eta(T,\rho)$$

New density fitted by experimental data

$$\rho_\eta(T,\rho) = \rho_0(T,\rho) \cdot \psi(\delta)$$

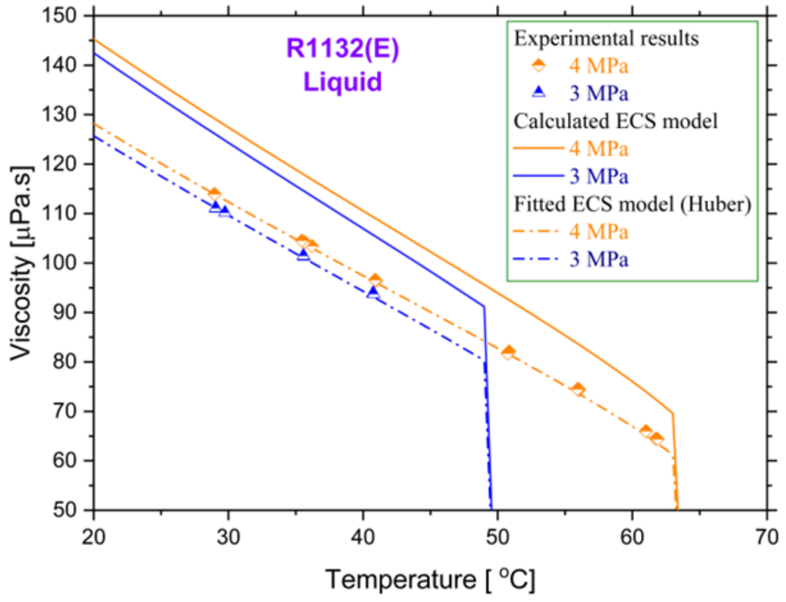
Viscosity shape factor: $\psi(\delta) = \sum_{k=0}^{n} \beta_k \delta^k$

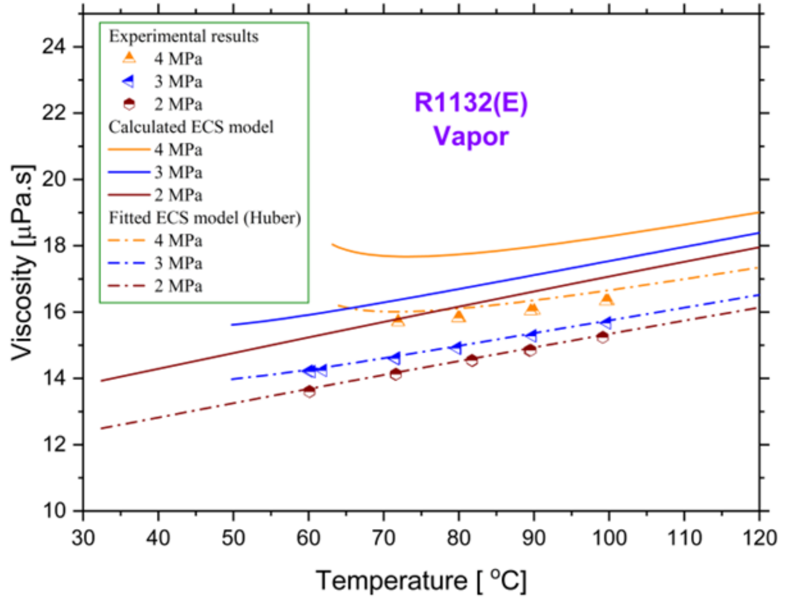
β_k : fitting parameters

Final equation of ECS model:

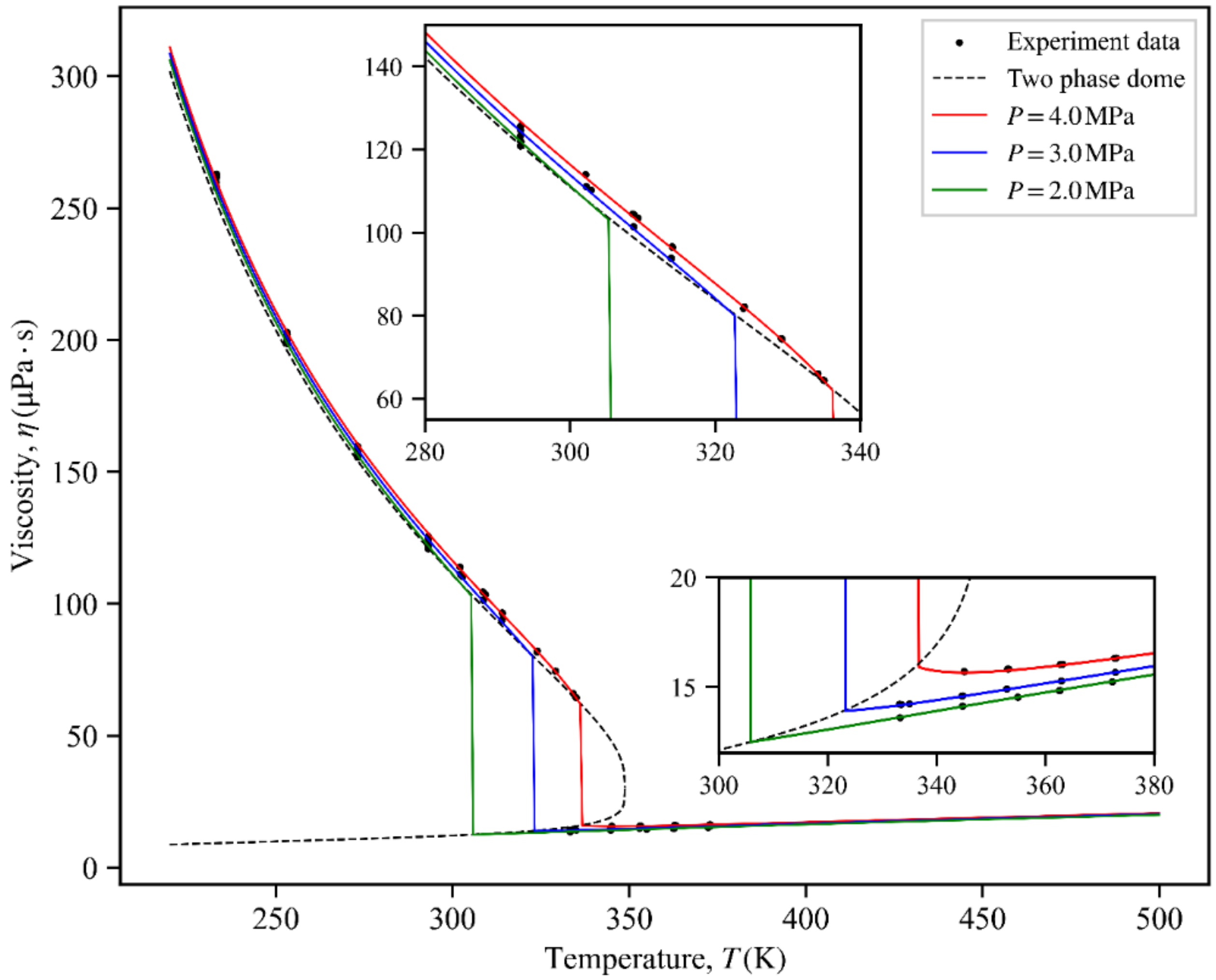
$$\eta(T,\rho) = F_{Chung} \frac{C\sqrt{MT}}{\sigma^2 \Omega^{(2,2)}} + \Delta\eta_0\left(T_0, \rho_0 \sum_{k=0}^{n} \beta_k \delta^k\right) \cdot F_\eta(T,\rho)$$

Viscosity Values Before and After Fitting



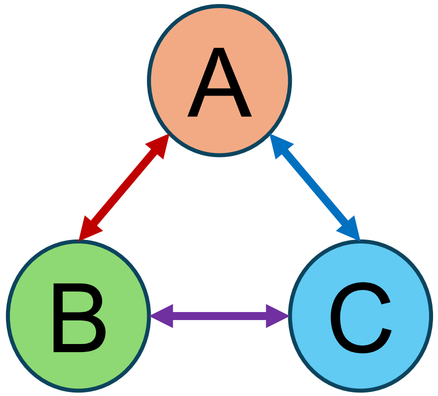


Overview of measured and predicted viscosities after fully optimized fitting



Viscosity Modeling of Mixture Refrigerants

- Viscosity model of a general mixture using ECS is complex compared to pure fluids
  - It requires viscosity binary interaction parameters among the constituent fluids
  - For example, if a mixture consists of 3 components (say A, B, and C), ECS requires viscosity binary interaction parameters for each pair (A-B, A-C, and B-C)



  - For each pair of fluids, there exists four binary interaction parameters
    - ***Two*** for dilute-gas viscosity and ***two*** for residual contribution
  - These parameters are found by fitting appropriate experimental data

Viscosity Modeling of Mixture Refrigerants

- Viscosity of a binary mixture is given by

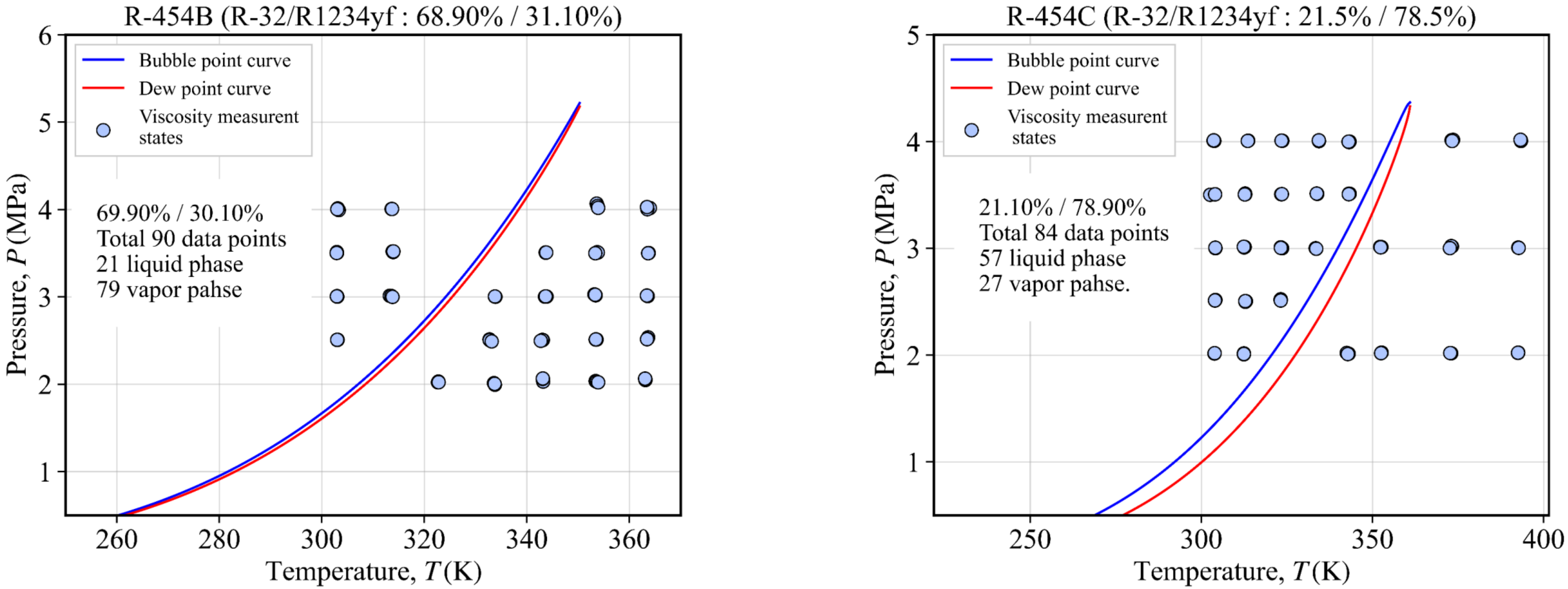
$$\eta(T,\rho,\vec{z}) = \underbrace{\frac{\left(z_1^2 H_{22} + z_2^2 H_{11} - 2 z_1 z_2 H_{12}\right)}{H_{11}H_{12} - H_{12}^2}}_{\text{Dilute-gas contribution}} + \underbrace{\Delta\eta_0(T_0,\rho_0)\cdot F_\eta(T,\rho,\vec{z})}_{\text{Residual contribution}}$$

- There are four adjustable parameters in the model
  - k_σ and k_ε: belongs to dilute-gas part
  - k_f and k_h: belongs to residual part
  - These are found by fitting to experimental data
  - They are set to zero if no experimental data is available

- Nitrogen is used as the reference fluid in the residual part

Viscosity Modeling of Mixture Refrigerants

- Let us take the example for R32 & R1234yf mixture



State points where viscosity measurements have been carried out
R-454B (on the left) and R-454C (on the right)

Viscosity Modeling of Mixture Refrigerants

- Let us take the example for R32 & R1234yf mixture

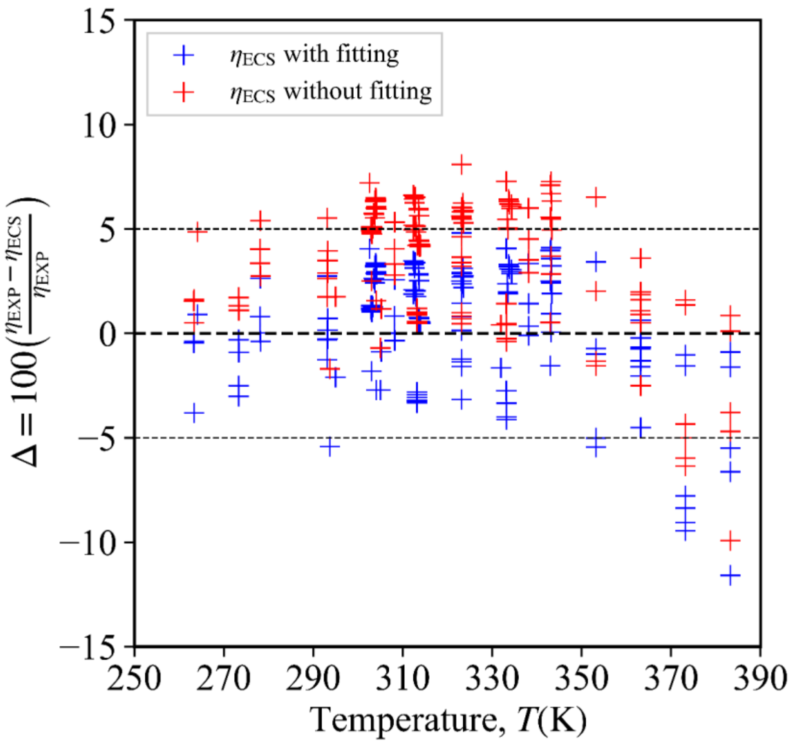


Fig. 2 Deviation plot before and after fitting

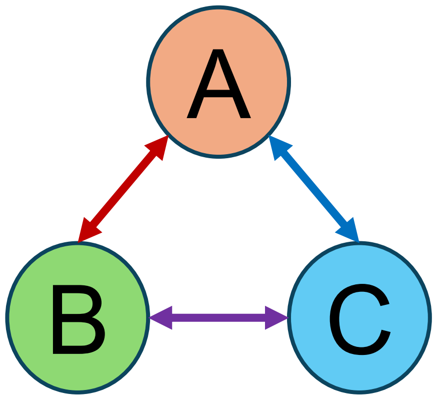
- The fitting parameters were found to be

k_σ	0.085644
k_ε	-0.589745
k_f	0.241787
k_h	-0.030763

- Before fitting AAD = 4.0% and after fitting AAD = 2.3%

Thermal Conductivity Modeling of Mixture Refrigerants

- Like viscosity, thermal conductivity modeling of a general mixture using ECS is complex compared to pure fluids
  - For each pair of fluids in a mixture, there exists three binary interaction parameters
    - ***One*** for dilute-gas thermal conductivity and ***two*** for residual contribution
- At present we are in the process of developing fitting code for thermal conductivity of mixtures



$$\lambda_m(T,\rho,\vec{z}) = \underbrace{\lambda_m^*(T,\vec{z}) + \lambda_m^{\text{int}}(T,\vec{z})}_{\text{Dilute-gas contribution}} + \overbrace{\Delta\lambda_m^{\text{r}}(T,\rho,\vec{z})}^{\text{Residual contribution}} + \underbrace{\Delta\lambda_m^{\text{crit}}(T,\rho,\vec{z})}_{\text{critical enhancement}}$$

$$\lambda_m^*(T,\vec{z}) + \lambda_m^{\text{int}}(T,\vec{z}) = \sum_{j=1}^{n} \frac{z_j\left(\lambda_j^*(T) + \lambda_j^{\text{int}}(T)\right)}{\sum_{i=1}^{n} z_i \phi_{ij}}$$

$$\Delta\lambda_m^{\text{r}}(T,\rho,\vec{z}) = \Delta\lambda_{ref}^{\text{r}}\left(T_{ref},\rho_{ref},\right) \cdot F_\lambda(T,\rho,\vec{z})$$

Average of Olchowy-Sengers model

Summary

- **Viscosity was measured by the tandem capillary tube method for pure and mixture refrigerants**
  - ✓ Measured refrigerants: R1234ze(Z), R1336mzz(Z), R1233zd(E), HFE-356mmz, R1224yd(Z), R1123+R32, R1336mzz(E), "3,3,4,4,5,5-HFCPE", CF_3I, R1132(E), R454B, R474A,
- **Thermal conductivity was measured by the transient hot wire method for pure and blended refrigerants**
  - ✓ Measured refrigerants: R1234ze(Z), R1336mzz(Z), R1233zd(E), HFE-356mmz, R1224yd(Z), R1336mzz(E), "3,3,4,4,5,5-HFCPE", R32+R1234ze(E), CF_3I, R1132(E), R454B, R454C, R474A,
- **Calculation models were developed for pure and blended refrigerants**
  - ✓ ECS models of viscosity and thermal conductivity for pure refrigerants has been developed.
  - ✓ ECS model of viscosity and thermal conductivity for mixture refrigerants has been developed.

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