

PHD DEFENSE

Contribution to prognostics of PEM fuel cells: approaches based on degradation information at multiple levels

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1 Background & motivation

ADAPTING THE STATE OF THE ART:

- 2 Remaining Useful Life prediction of PEM fuel cells
 - ▶ Particle Filtering-based RUL prediction
 - Taking into account reversible degradation

- Multi-level prognostics using online inspection of degradation covariates
- Multi-level prognostics using different sources of degradation information
- 6 Conclusion & perspectives

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Problem Statement

Part I

Background & Motivation

Proton Exchange Membrane Fuel Cells

Prognostic and Health Management

Problem Statement

Proton Exchange Membrane (PEM) fuel cell

- Energy transition Hydrogen
- Promising alternative transition device
 Fuel Cells
- PEM Fuel Cell

. . .

- ► Low temperature 80°C-100°C
- ► Small size Single cell : 100 cm^2 P \approx 0.4 W/cm^2
- High efficiency η ≈70%
- Tailorable power







Problem Statement

Stack and cell



- ► Anode:
 - $H_2 \to 2H^+ + 2e^-$

Cathode:

 $\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$

Overall:

 $\frac{1}{2}O_2 + H_2 \to H_2O$

Problem Statement

Balance of Plant



Subsystems

- Reactant conditioning tank, pumps, valves,...
- Electronic conditioning regulator, controller, ...
- Thermal management heat exchanger, ...

Problem Statement

Balance of Plant

Heat analyzing

A PEM fuel cell test bench in FCLAB



Challenges

Issues

- Poor health management (Faults)
- Materials and interfaces degradation (Aging)

Consequences

- Performance degradation
- Lifespan limitation

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- Materials and interfaces degradation (Aging)

Consequences

- Performance degradation
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Challenges

- Enhance reliability
- Better manage lifespan
- \Rightarrow Prognostic and Health Management

Proton Exchange Membrane Fuel Cells

Problem Statement

Prognostics and Health Management (PHM)

- Recent dynamic approach to monitor, analyze and master the Remaining Useful Life (RUL) of industrial systems
- Pioneering technique in fuel cells technologies



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Prognostic and Health Management $\circ \bullet$

Problem Statement

Prognostic and Health Management $\circ \bullet$

Problem Statement

Remaining Useful Life (RUL)



Time











Difficulties



- Degradation can be different:
 - ▶ irreversible (*e.g.* due to material deterioration)
 - ► reversible (e.g. due to operating conditions)
- Degradation can be generated at different levels
- \Rightarrow Observed from different measurements

Problem Statement

Degradation measurements

Stack voltage (or power)



Polarization curves



Electrochemical Impedance Spectroscopy (EIS)



- Classical measurements Voltage, Power
- State of Health (SOH) measurements EIS, Polarization

Problem Statement

Degradation measurements

Stack voltage (or power)



Polarization curves



Electrochemical Impedance Spectroscopy (EIS)



- Classical measurements Voltage, Power
- State of Health (SOH) measurements EIS, Polarization
- \Rightarrow Use multiple measurements

1 RUL prediction (on real application data) with taking into account both irreversible and reversible degradation

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 - An approach using both stack power and SOH characterization measurements

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RUL Prognosis on Stack Power

RUL Prognosis Considering Recovery

Part II

Remaining Useful Life Prediction of PEM fuel cell

RUL Prognosis on Stack Power

Particle Filtering-based prognostics



RUL Prognosis on Stack Power

RUL Prognosis Considering Recovery

Particle Filtering-based prognostics



Estimator uses large amount of randomness to approach the truth

- Technique for implementing Recursive Bayesian filter by Monte Carlo sampling
- Nonlinear, non Gaussian model tracking
RUL Prognosis Considering Recovery

Particle Filtering (PF)



Particles with corresponding weights are used to form an approximation of PDF

Split initial state into particles

 $x_{t-1} \to \{x_{t-1}^i\}_{i=1}^N$

Propagate particles through the transition function

$$x_t^i = f(x_{t-1}^i, \omega_{t-1}^i) \to p(x_t | x_{t-1})$$

3 Importance sampling

$$W_t^i \propto p(z_t | x_t^i) \to x_t^{i*}$$

4 Re-sampling

$$\{x_{t-1}^{i*}\}_{i=1}^N \to p(x_t^*|z_t)$$

RUL Prognosis Considering Recovery

Prognostic performance



Prognostic metrics

RUL Prognosis Considering Recovery

Prognostic performance



Prognostic metrics

Accuracy index

$$Acc_{\lambda} = 1 - \frac{|RUL_{\lambda}^* - \widehat{RUL}_{\lambda}|}{RUL_{\lambda}^*}$$

RUL Prognosis Considering Recovery

Prognostic performance



Prognostic metrics

Accuracy index

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• α - λ Accuracy index

 $\alpha A c_{\lambda} = p\left(\alpha_{\lambda}^{-} \le \widehat{RUL}_{\lambda} \le \alpha_{\lambda}^{+}\right)$

RUL Prognosis Considering Recovery

Prognostic performance



Prognostic metrics

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•
$$\alpha \cdot \lambda$$
 Accuracy index
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Steadiness index

$$Std_{\lambda} = \frac{\sqrt{var(\widehat{EOL}_{(\lambda-L):\lambda})}}{EOL^*}$$

RUL Prognosis Considering Recovery

Prognostic performance



Prognostic metrics

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Steadiness index

$$Std_{\lambda} = \frac{\sqrt{var(\widehat{EOL}_{(\lambda-L):\lambda})}}{EOL^*}$$

Precision index

$$Prc_{\lambda} = \frac{\widehat{RUL}_{\lambda}^{CI+} - \widehat{RUL}_{\lambda}^{CI-}}{RUL_{\lambda}^{*}}$$

Application data (test bench FCLAB)

- ► Two PEM fuel cell stacks under operation degrade
- Stack power measurements are used as the degradation indicator



FC2 - Long-term test with high frequencies current ripples



Application data (test bench FCLAB)

- ► Two PEM fuel cell stacks under operation degrade
- Stack power measurements are used as the degradation indicator

Degradation state models

- Pure logarithmic: $x_k = -\alpha \cdot ln(t_k/t_{k-1}) + x_{k-1}$
- Log-linear: $x_k = -\alpha \cdot ln(t_k/t_{k-1}) - \beta \cdot (t_k - t_{k-1}) + x_{k-1}$
- Linear:

$$x_k = -\beta \cdot (t_k - t_{k-1})) + x_{k-1}$$

Polynomial:

 $x_k = \alpha \cdot (t_k - t_{k-1})^2 - \beta \cdot (t_k - t_{k-1}) + x_{k-1}$



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Polynomial:

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RUL Prognosis on Stack Power $\circ \bullet$

RUL Prognosis Considering Recovery

Difficulty

Reversible degradation



RUL Prognosis on Stack Power $\circ \bullet$

RUL Prognosis Considering Recovery

Difficulty

Reversible degradation



Use information from characterization measurement

⇒ Electrochemical Impedance Spectroscopy

RUL Prognosis on Stack Power $_{\rm OO}$

RUL Prognosis Considering Recovery

Electrochemical Impedance Spectroscopy (EIS) data

EIS measurements



Equivalent Circuit Model 1



Polarization resistance

$$R_{pol} = \frac{(R_1 + R_2) \cdot R_3}{R_1 + R_2 + R_3} + R_{el}$$

Kim, T. et al., (2014). A degenerated equivalent circuit model and hybrid prediction for state-of-health (SOH) of PEM fuel cell. In: 2014 International Conference on Prognostics and Health Management. pp. 1-7.

State transition model

Hypothesis

- Reversible degradation is related to R_{pol}
- R_{pol} can be estimated from EIS

$$x_{c} = x_{k} + \alpha_{1}(R_{pol}) \cdot exp(-\beta_{1}(R_{pol}) \cdot (t_{k} - t_{k-1}))$$
$$x_{k} = \alpha \cdot (t_{k} - t_{k-1})^{2} - \beta \cdot (t_{k} - t_{k-1}) + x_{k-1}$$

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 R_{pol} vs. Power (FC1)

 R_{pol} vs. Power (FC2)





State transition model

Hypothesis

- ► Reversible degradation is related to R_{pol}
- ► *R*_{pol} can be estimated from EIS

$$x_c = x_k + \alpha_1 \cdot \frac{R_{pol}(c)}{R_{pol}(0)} \cdot exp\left(-\beta_1 \cdot \frac{R_{pol}(c)}{R_{pol}(0)} \cdot (t_k - t_{k-1})\right)$$

R_{pol} vs. Power (FC1)



R_{pol} vs. Power (FC2)



RUL Prognosis Considering Recovery

Results (FC2)

Estimation with classical model



Estimation with proposed model



RUL predictions with classical model



RUL predictions with proposed model



RUL Prognosis Considering Recovery

Prognostic performance

RUL predictions of FC1



RUL predictions of FC2



Performance metrics

	FC 1				FC 2			
	Acc	aAc	Prc	Std	Acc	aAc	Prc	Std
Classical	0.67	0.29	0.89	0.06	0.48	0.11	0.69	0.17
Proposed	0.86	0.42	0.68	0.06	0.72	0.34	0.74	0.12

Summary

Challenge accomplished:

- RUL predictions of PEM fuel cell application with taking into reversible degradation
- ► Reversible degradation has been investigated by using the information of *R*_{pol}
- Prediction quality has been improved by using additional information from SOH characterization

How to generalize the use of degradation related information \Rightarrow Introduce **covariates** to the degradation model

Problem statement

Problem formulation

Numerical experiment

Results and summary

Part III

Multi-level Prognostics Using Online Inspection of Degradation Covariates

Numerical experiment

Results and summary

Problem statement



Degradation

- Continuously monitored with simple measurements
- Affected by the change of covariates

Assumption

Possible to inspect the covariates with extra cost

Objective

Use the information of covariates for RUL prediction

Numerical experiment

Results and summary

How?

Design a degradation covariate inspection scheme

- When and how to apply an inspection on covariates (that costs)
- Ensure the prognostic quality
- Minimize the inspection cost

Numerical experiment

Results and summary

General modeling assumptions

Numerical experiment

Results and summary

General modeling assumptions

Evolution of the degradation follows a stochastic process:

$$x_k = f_k(x_{k-1}, \Theta_k)$$

Impacts of covariates on the degradation model parameter can be quantified and modeled:

$$\Theta_k = g_k(c_k)$$

Numerical experiment

Results and summary

General modeling assumptions

Evolution of the degradation follows a stochastic process:

$$x_k = f_k(x_{k-1}, \Theta_k)$$

Impacts of covariates on the degradation model parameter can be quantified and modeled:

$$\Theta_k = g_k(c_k)$$

 \Rightarrow Possible to update the degradation model by inspecting the degradation covariate with cost

Problem statement

Problem formulation

Numerical experiment

Results and summary

Cost of the covariate inspection

Global cost

$$J = J_{inspection}(N_{ins}) + J_{estimation}(\hat{\epsilon})$$

- Inspection incurs application cost
- Inaccurate estimation leads to prognostic quality cost

Method

Two decision variables:

- ► inter-inspection period $\tau \rightarrow$ number of inspections N_{ins}
- error threshold $ET \rightarrow$ estimation error $\hat{\epsilon}$



Numerical experiment

Results and summary

Covariates inspection schemes



Periodic inspection

Performed regularly at fixed time intervals $\boldsymbol{\tau}$

Online inspection

Triggered online when the accumulated estimation error reaches an error threshold ET

Numerical experiment

Results and summary

Degradation example



Simulated by a state transition function ²:

$$x_k = x_{k-1} \cdot exp(-b_k(\mathbf{c}_k) \cdot \Delta t)$$

The presence of covariate c_k impacts the degradation behavior by changing trend parameter b_k .

²An, D., Choi, J.H., and Kim, N.H. (2013). Prognostics 101: A tutorial for particle filter-based prognostics algorithm using Matlab.*Reliab. Eng. Syst. Sat.*, 115, 161-169.

Problem statement

Problem formulation

Numerical experiment

Results and summary

Degradation example



Degradation estimation examples

Problem statement

Problem formulation

Numerical experiment

Results and summary

Decision variables

Periodic inspection



Inter-inspection period $\tau^* \simeq 100h$

Online inspection



Error threshold $ET^* = 4\%$ Minimum inter-inspection period $\tau_h^* = 50h$

Numerical experiment

Results and summary

Different inspection schemes

Periodic covariate inspection



Online covariate inspection



Error of periodic inspection



Error of online inspection



Numerical experiment

Results and summary

RUL predictions results

RUL predictions without inspection



 \sim with periodic inspection

\sim with online inspection





Numerical experiment

Results and summary 000

Cost and prognostic performance

	Without inspection	Periodic \sim	Online \sim
Acc	0	0.54	0.27
αAc	0	0.53	0.36
$\hat{\epsilon}(\%)$	69.19	20.73	28.91
N_{ins}	-	215	110
J (€)	692	422	399

Problem statement

Problem formulation

Numerical experiment

Results and summary

Summary

Challeng accomplished:

- RUL prediction using the information from inspections of covariate
- ► Two covariate inspection schemes: online and periodic
- Online inspection scheme helps to minimize the cost by avoiding unnecessary inspection

 \Rightarrow A different way of using degradation related information at different levels

Part IV

Multi-level Prognostics Using Different Sources of Degradation Information³



³Work in this section has been partly done during an international mobility in the **Laboratory of signal and risk analysis (LASAR**) at *Politecnico di Milano*

Ensemble-based Approach

Numerical Experiment

Problem statement

Stack voltage vs. time

Polarization measurement



Degradation of a stack observed from different measurements:

- External measurement: measured frequently, "poor quality" (voltage)
- Internal characterization: less frequently, "good quality" (polarization)

Objective

Aggregate RUL predictions from both measurements to improve global RUL quality

Polarization model

The output voltage of a stack at a given current density J^4 :

$$V_{st}(J) = n \cdot \{ \frac{E - r}{V} \cdot J - A \cdot ln(\frac{J}{j_0}) - m_1 \cdot exp(m_2 \cdot J) \}$$

 $V_{st}: \text{stack voltage} \\ n: \text{number of cells in the stack} \\ E: \text{open circuit voltage} \\ r: \text{internal resistance} \\ J: \text{operating current density} \\ A: Tafe coefficient \\ j_0: \text{exchange current density} \\ \end{cases}$

m1 and m2: mass-transfer constants

Time impact on model parameters



⁴J. Larminie and A. Dicks, *Fuel cell systems explained*. Wiley, 2003.
Polarization model

The output voltage of a stack at a given current density J^4 :



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- Vst: stack voltage
- n: number of cells in the stack
- E: open circuit voltage
- r: internal resistance
- J: operating current density
- A: Tafel coefficient
- j₀: exchange current density

 m_1 and m_2 : mass-transfer constants

Time impact on model parameters

Parameters estimation results

Time (h)	E(V)	$r~(\Omega~{ m cm}^2)$
0	1,055	0,190
48	1,054	0,193
185	1,052	0,202
348	1,051	0,204
515	1,051	0,206
658	1,047	0,211
823	1,043	0,219
991	1,044	0,208

^{*}J. Larminie and A. Dicks, *Fuel cell systems explained*. Wiley, 2003.

SOH degradation model



SOH degradation model $\gamma(t)$ of a PEM fuel cell ⁵

- Definition: degradation rate of E(t) and r(t)
- ► Assumption: availability of a procedure which returns an estimation of *γ*(*t*) from polarization measurements

⁹Bressel et al.,"Remaining Useful Life Prediction and Uncertainty Quantification of Proton Exchange Membrane Fuel Cell Under Variable Load," *IEEE Trans. Ind. Electron.*, vol.63, no.4, pp.2569-2577, 2016.

RUL aggregation

- SOH degradation $\gamma(t)$
- Voltage degradation $V_{st}(t)$ driven by $\gamma(t)$



$$\widehat{RUL}_t = \sum_{i=1}^{M} W_t^i \cdot \widehat{RUL}_t^i$$
$$p(\widehat{RUL}_t) = \sum_{i=1}^{M} W_t^i \cdot p(\widehat{RUL}_t^i)$$

 $\begin{array}{l} \widehat{RUL}_t \text{: the ensemble outcome for the RUL prediction} \\ RUL_t^i \text{: RUL prediction by each } i^{t\,h} \text{ individual model} \\ M \text{: the number of models} \end{array}$

 W_t^i : local weights (non-negative and sum to 1).

Local weight W_t determination

$$\widehat{RUL}_t = W_t^{(1)} \cdot \widehat{RUL}_t^{(1)} + W_t^{(2)} \cdot \widehat{RUL}_t^{(2)}$$

Why: to benefit from both measurements

- "good" RUL quality \rightarrow large weight
- "poor" RUL quality \rightarrow small weight

How

- Evaluated locally on a similar training set (historical data)
- Weights are recomputed at each time step



Numerical Experiment









Numerical experiment

Simulation objective

Produce realistic signals:

- signal variability and randomness from stack to stack
- dependency between two types of measurement

Simulated signals synthesis

- Two types of measurement: $\gamma(t)$ and $V_{st}(t)$
- Ad hoc simulations based on dependent stochastic processes (Gamma process)

Simulated signals

• SOH degradation $\gamma(t)$

• SOH degradation $\gamma(t)$



- SOH degradation $\gamma(t)$
- Voltage degradation $V_{st}(t)$ driven by $\gamma(t)$

$$V_{st}(t) = f(E(t), r(t), J(t)) \begin{cases} E(t) = E_0 \cdot (1 - \gamma(t)) \\ r(t) = r_0 \cdot (1 + \gamma(t)) \end{cases}$$

- SOH degradation $\gamma(t)$
- Voltage degradation $V_{st}(t)$ driven by $\gamma(t)$

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Degradation Models

Ensemble-based Approach

Numerical Experiment

Summary o

RUL prediction results

Estimation of $\gamma(t)$



RUL predictions based on $\gamma(t)$



Estimation of $V_{st}(t)$



RUL predictions based on $V_{st}(t)$



RUL aggregation

Individual RUL



RUL aggregation



Local error *LE*



RUL aggregation



Local error LE



Local weight w



RUL aggregation



Local error *LE*



Local weight w



RUL aggregation



Local error LE



Local weight w



Performance evaluation

Average performance on 50 test trajectories:

Performance	Model 1	Model 2	Ensemble	
			Point value	PDF
Acc	0.52	0.12	0.55	0.56
αAc	0.31	0.24	0.54	0.49
Std	0.16	0.14	0.07	0.07

- * Prediction quality is improved with different configurations
 - different process variances
 - different signals' dependencies

Summary

- RUL prediction quality has been evaluated on historical data
- Weights have been locally associated to individual predictions from two different measurement signals
- Aggregated results provided better quality than individual ones
- Ready for the application in PEM fuel cell when data requirement is met

Part V

Conclusion & Perspectives

- Developed a PF-based approach for RUL prediction of PEM fuel cell using deeper characterization information
- Proposed an online inspection of degradation covariate at different level to adapt RUL prediction
- Proposed an Ensemble-based approach using degradation information at different levels to improve the RUL prediction quality

- Developed a PF-based approach for RUL prediction of PEM fuel cell using deeper characterization information
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Perspectives

Short term

- Data involves varying usage profile for both development and validation
- ► Inspection schemes based on simple cost function → consideration of more impacts, *e.g.* timeliness

Long term

- Validation of developed approaches with real-life data should be envisaged to become commercially attractive
- Not limited to the framework of PEM fuel cell

 adapt and extend to other applications

Thank you !

Publications

- Zhang D, Cadet C, Yousfi-Steiner N and Bérenguer C (2017) PEMFC RUL Prognostics Considering Degradation Recovery Phenomena. *Journal of Risk and Reliability*, revised.
- Zhang D, Cadet C, Yousfi-Steiner N and Bérenguer C (2017) A Study of Online Inspection for Multi-level Prognostics. In: IFAC-PapersOnLine, 50(1), pp.13716-13721. DOI: 10.1016/j.ifacol.2017.08.2549.
- Zhang D, Baraldi P, Cadet C, Yousfi-Steiner N, Bérenguer C and Zio E (2017) A Study of Local Aggregation of An Ensemble of Models for RUL Prediction. In: *The 10th International Conference on Mathematical Methods in Reliability (MMR 2017)*, July 3-6, 2017, Grenoble, France.
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- Zhang D, Cadet C, Yousfi-Steiner N and Bérenguer C (2016) Some Improvements of Particle Filtering Based Prognosis for PEM Fuel Cells. In: *IFAC-PapersOnLine*, volume 49. pp.162-167. DOI: 0.1016/j.ifacol.2016.11.028.
- Zhang D, Cadet C, Yousfi-Steiner N and Bérenguer C (2015) PHM-oriented degradation indicators for PEM fuel cells:What can be learnt from battery State of Charge estimation . In: Proc. 6th International Conference on Fundamentals & Development of Fuel Cells, Paper 190. Feb 2015, Toulouse, France.

Part VI

Appendix

RUL uncertainty



Proposition 1

Two indicators $V_{st}(t)$ and $\gamma(t)$ are generated from the same realization of a Gamma degradation process, with different additive noises.

$$\overline{\gamma}(t) = \overline{\gamma}(t-1) + \Gamma(\overline{\alpha}\Delta t, \overline{\beta})$$
$$\gamma^{i}(t) = \gamma^{i}(t-1) + \Gamma(\alpha^{i}\Delta t, \beta^{i})$$

$$\begin{cases} \gamma_1^i(t) = \gamma^i(t) + \mathcal{N}(0, \sigma_1^2(t)) \\ \gamma_2^i(t) = \gamma^i(t) + \mathcal{N}(0, \sigma_2^2(t)) \end{cases} \rightarrow \begin{cases} \gamma_{meas}^i(t) = \gamma^i(t) + \mathcal{N}(0, \sigma_{\omega_1}^2(t)) \\ V_{st\,meas}^i(t) = V_{st}^i(t) + \mathcal{N}(0, \sigma_{\omega_2}^2(t)) \end{cases}$$
Proposition 2

Two indicators $V_{st}(t)$ and $\gamma(t)$ are simulated from two different degradation processes, yet the two processes are dependent processes by construction.

$$\begin{array}{l}
G_1(\alpha_1,\beta)\\
G_2(\alpha_2,\beta)
\end{array} \rightarrow \begin{cases}
a_1 = \alpha_1 - \rho \sqrt{\alpha_1 \alpha_2} \quad \rightarrow g_1^i(a_1,\beta)\\
a_2 = \alpha_2 - \rho \sqrt{\alpha_1 \alpha_2} \quad \rightarrow g_2^i(a_2,\beta)\\
a_1 = \rho \sqrt{\alpha_1 \alpha_2} \quad \rightarrow g_1^i(a_3,\beta)
\end{array} \rightarrow \begin{array}{l}
G_1^i = g_1^i + g_3^i\\
G_2^i = g_2^i + g_3^i
\end{array}$$

$$\begin{cases} \gamma_1^i(t) = \gamma_1^i(t-1) + G_1^i \\ \gamma_2^i(t) = \gamma_2^i(t-1) + G_2^i \end{cases} \rightarrow \begin{cases} \gamma_{meas}^i(t) = \gamma^i(t) + \mathcal{N}(0, \sigma_{\omega_1}^2(t)) \\ V_{st\,meas}^i(t) = V_{st}^i(t) + \mathcal{N}(0, \sigma_{\omega_2}^2(t)) \end{cases}$$