Effect of Correlation Factors on the Measurement Uncertainty of Physical Inventory in Bulk Handling Facilities

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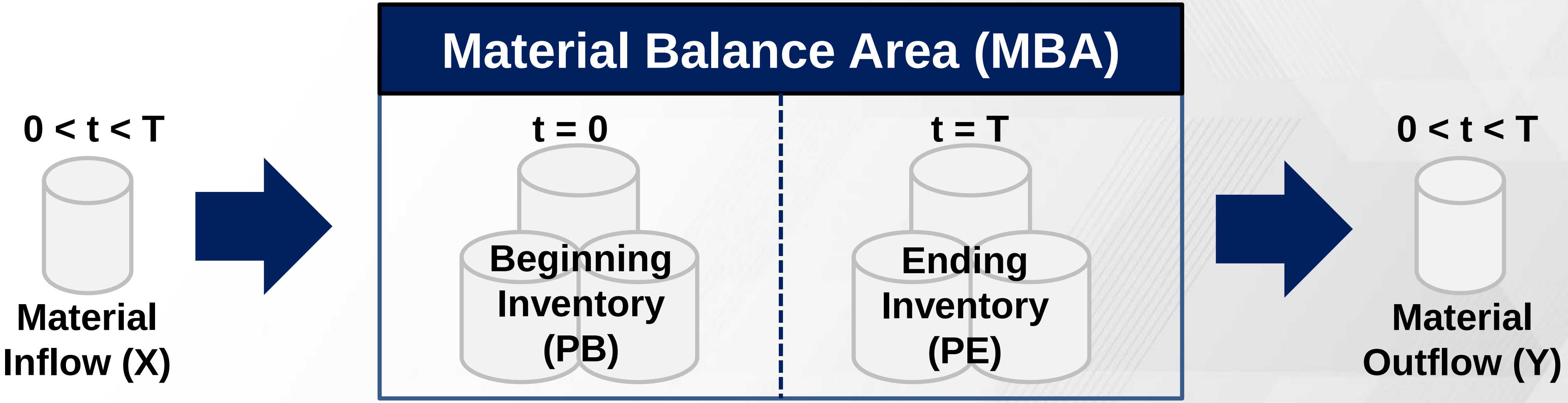
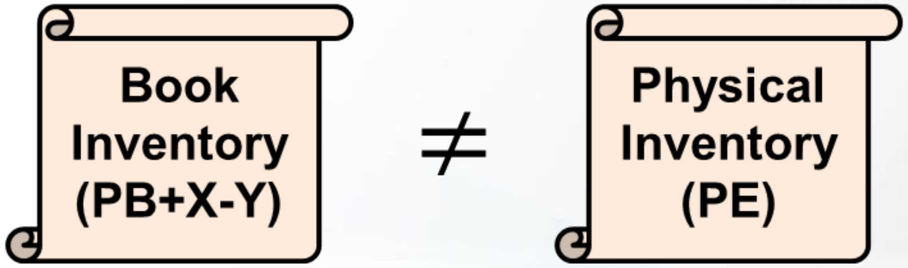
Contents

- **Material Balance Evaluation (MBE)**



In bulk handling facilities (BHFs)



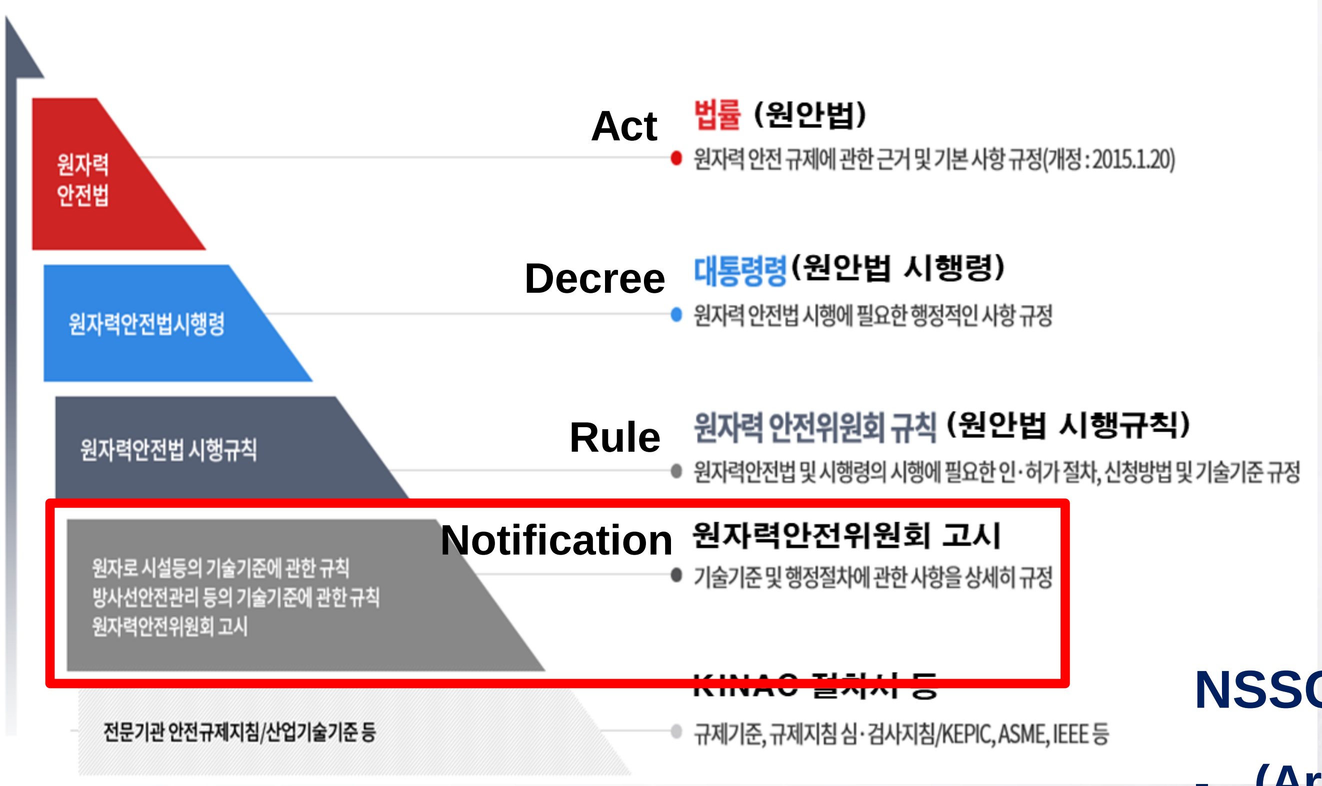
MBE process

1) Calculate MUF

$$MUF = PB + X - Y - PE$$

2) Calculate measurement uncertainty of MUF (σ_{MUF})
3) Evaluate the MUF by comparing MUF and σ_{MUF}

• Legal basis of MBE※



※ nsic.mssp.go.kr/nsic.do?nsicKey=300101

NSSC notification No. 2017-83

- **(Article 4) National safeguards inspection includes**
  - **(Subparagraph 8) verification of** uncertainties in material accounting including **MUF**

- **MBE in national inspection**

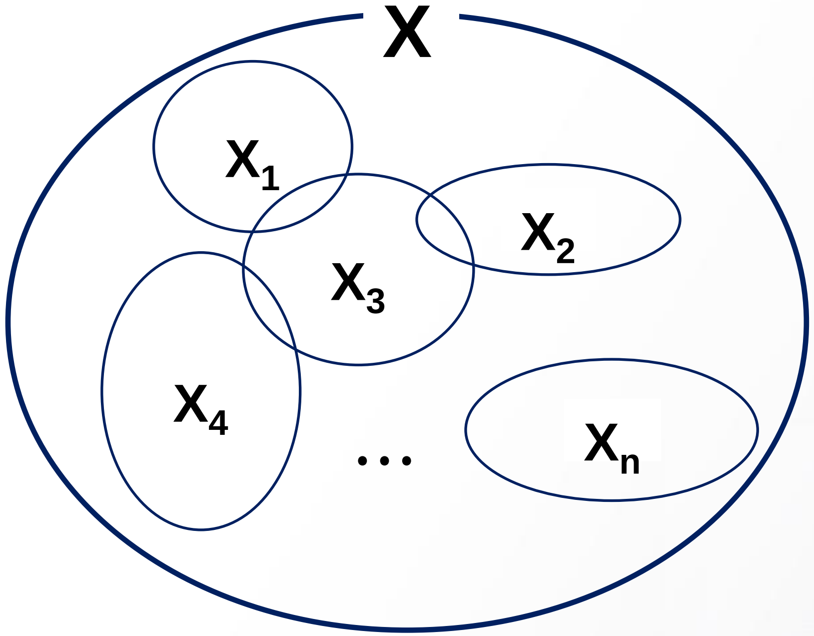
As-Is (IAEA Inspection)	To-Be (National Inspection)
• Adopt the results of IAEA results • Purpose of IAEA inspection: - Diversion detection	• Independent MBE in national inspection • Purpose of national inspection - Diversion detection - Evaluation of facilities' accounting system

Develop an **optimized MBE method** for national inspection,
which **satisfies diversion detection** & **accounting system evaluation**

- **Uncertainty expression methods in MBE**

	Top-down approach (IAEA)	GUM (National Inspection)
Statistics	Pair-wise difference (ANOVA & Grubbs' method)	Bayesian inference
Individual Components	Random & systematic error	Type A & B uncertainty
Advantages	• Minimized facility information • Wide applicability	• High traceability & repeatability • Detailed uncertainty management
Disadvantages	• Low traceability & repeatability • Limited application for detailed uncertainty management	• Detailed information on accounting system • Limited applicability

• Correlation factors in GUM



$$u(x_i) = \begin{cases} \frac{s(x_i)}{\sqrt{n}} & (Type\ A) \\ Depends & (Type\ B) \end{cases}$$

- Combined standard uncertainty of mesurand X with n variables

Equation for the mesurand X

$$X = f(x_1, x_2, \dots, x_n)$$

Apply Taylor series

$$X = X_o + \sum_{i=1}^{n} \frac{\partial X}{\partial x_i}(x_i - x_{i0}) + \sum_{k=1}^{n} \sum_{j=1}^{n} \frac{1}{2!} \frac{\partial^2 X}{\partial x_j x_k}(x_j - x_{j0})(x_k - x_{k0}) + \cdots$$

Approximation

$$X \cong X_o + \sum_{i=1}^{n} \frac{\partial X}{\partial x_i}(x_i - x_{i0})$$

Estimate the variance of the mesurand X

$$E[(X - X_o)^2] = \mathrm{E}\left[\left(\sum_{i=1}^{n} \frac{\partial X}{\partial x_i}(x_i - x_{i0})\right)^2\right]$$

$$u_c(X)^2 = \sum_{i=1}^{n} \left(\frac{\partial X}{\partial x_i}\right)^2 \mathrm{u}(\mathrm{x_i})^2 + \sum_{j=k+1}^{n} \sum_{k=1}^{n-1} \frac{\partial^2 X}{\partial x_j \partial x_k} r(\mathrm{x_j}, \mathrm{x_k}) \mathrm{u}(\mathrm{x_i}) \mathrm{u}(\mathrm{x_j})$$

Independent factors (Random components)

Correlation factors (Systematic components)

Usually neglected for single item measurement

- **Correlation** factors were **usually neglected due to the low contribution** to propagate the combined uncertainty of a mesurand
- However, **the contribution significantly increases** if;
  - The number of variables (**n**) significantly **increases**
  - The correlation between variables are strong **($\boldsymbol{r(x_j, x_k) = 1}$)**
- Combined MUF uncertainty (σ_{MUF}) satisfies both criteria
  - $N \gg 1{,}000,\ r(\mathrm{x_j}, \mathrm{x_k}) = \pm 1$

Purpose of the research

Compare the $\boldsymbol{\sigma_{MUF}}$ with correlation and without correlation for accounting systems in a benchmark bulk handling facility

02 Methods

- **Governing equation for PE**

$$M(PE) = \sum_{i=1}^{I} \sum_{j=1}^{J} (m_{ij} f_{U,ij} w_{235,ij})$$

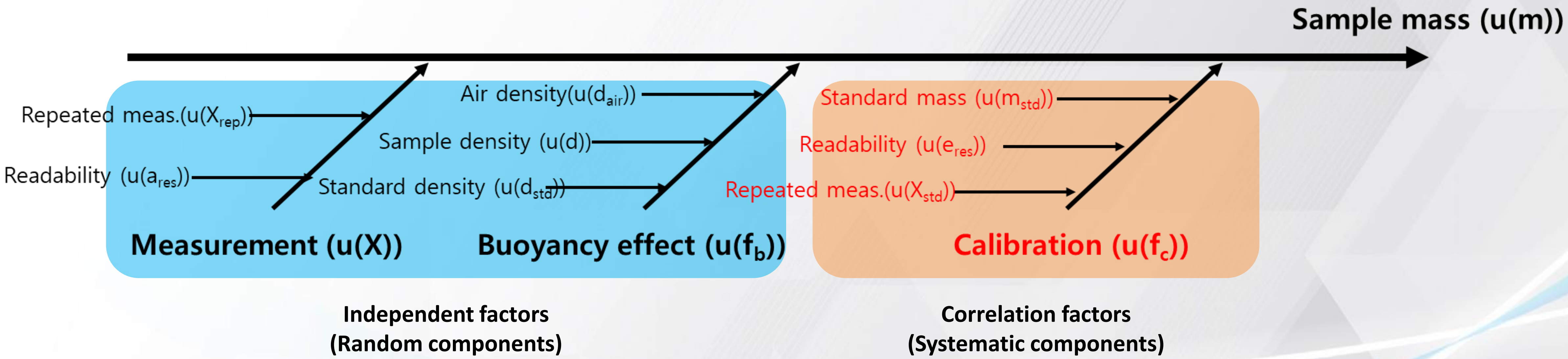
m: Net mass, f: Uranium concentration,
w_{235}: ^{235}U enrichment,
i: number of stratum ($i = 1,2,\dots,I$)
j: number of lot (process) (j $= 1,2,\dots,J$)

- **Measurement (uncertainty) components in inventory taking**
  - Weighing (m) with EBAL
    - EBAL indicator (X), buoyancy factor (f_b), and calibration factor (f_c)
  - U concentration analysis (f_U) with GRAV
    - Oxygen to Uranium ratio (O/U)
    - ➢ Mass ratio (m_i/m_f), impurity concentration (w_I, w_F), and ^{235}U enrichment (w_{235})
  - ^{235}U enrichment (w_{235}) with TIMS
    - Isotopic ratio (R_{234}/R_{238}, R_{235}/R_{238}, R_{236}/R_{238})
    - ➢ Sample meas. ratio ($RS(m)_{23x/238}$), cert. meas. ratio ($RC(m)_{23x/238}$), certificate ratio ($RC(c)_{23x/238}$), BKG ($RB_{23x/238}$), det. eff. ($\delta(Y), \delta(L), \delta(F)$)

- **Measurement uncertainty for weighing ($u(m)$)**

$$m = Xf_bf_c \qquad \begin{aligned} X &= X_m + \varepsilon \\ f_b &= (1 - d_a/d_s)/(1 - d_a/d) \\ f_c &= m_s/(X_{sm} + \varepsilon) \end{aligned}$$

  - Assumptions:
    - Standard measurement procedure
    - Single calibration for an EBAL between each physical inventory taking (PIT)

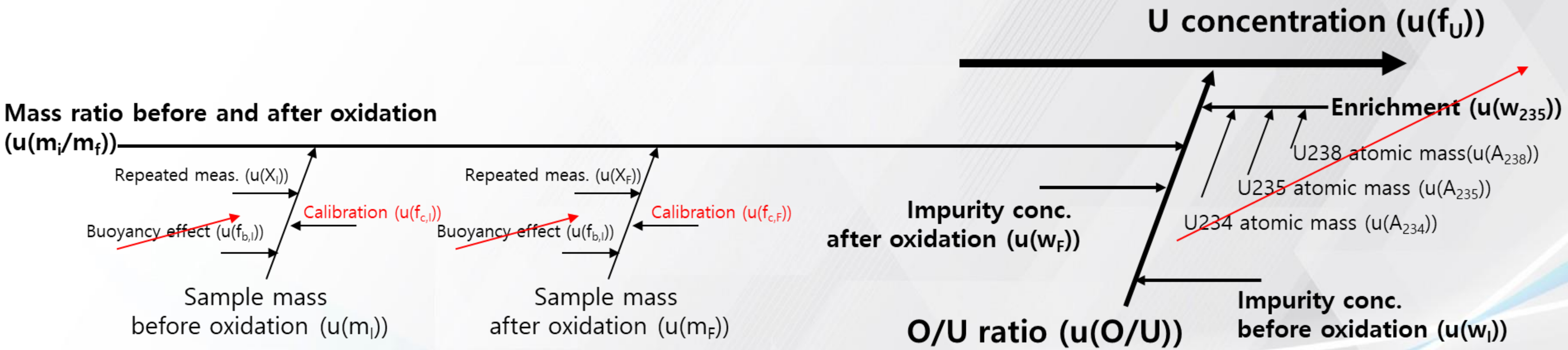


02 Methods

- **Measurement uncertainty for U conc. analysis ($u(f_U)$)**

$$f_U = \frac{A_U}{A_U + A_O(O/U)} \qquad O/U = \frac{[(1-w_I)(m_i/m_f) - F_S(1-w_F)]A_U}{F_S(1-w_F)A_O}$$

  - Assumptions:
    - Standard measurement procedure
    - Neglected uncertainty components with small contribution (< 0.01%)



02 Methods

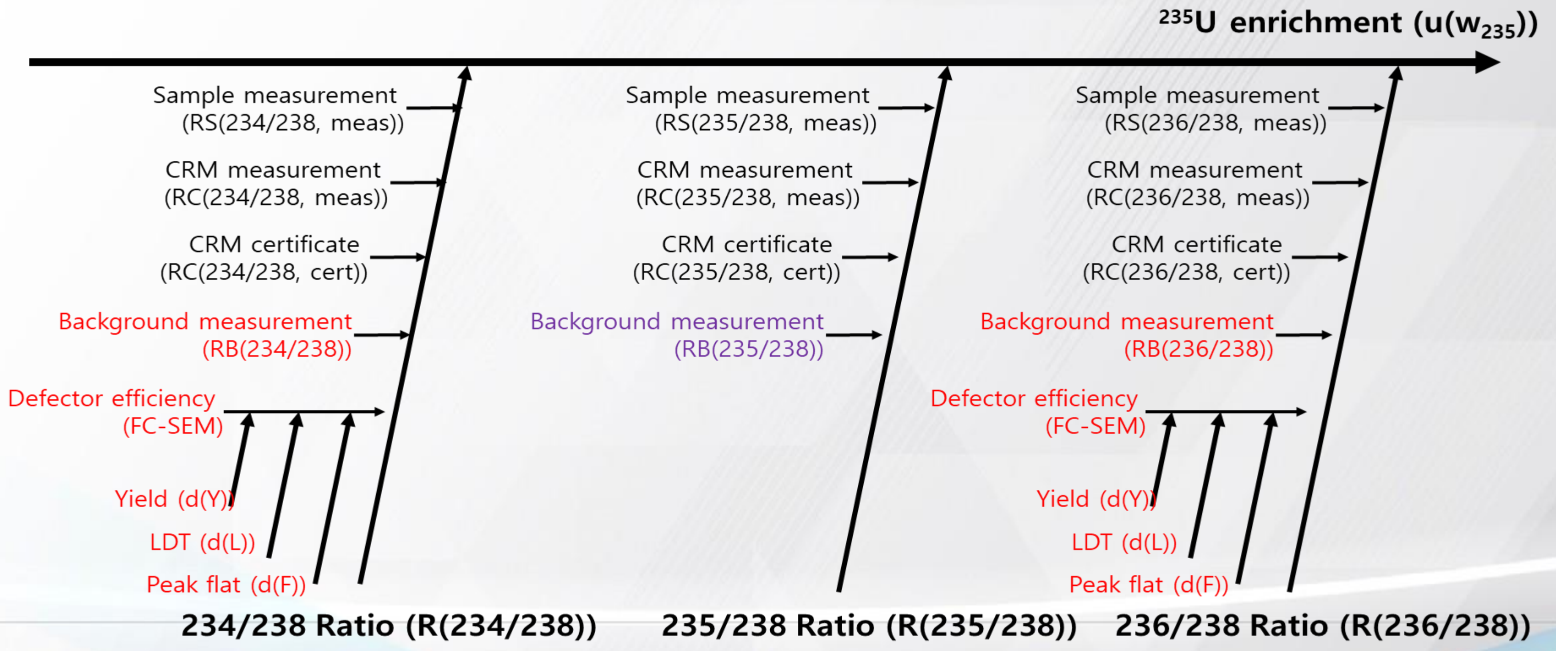
- **Measurement uncertainty for ^{235}U enrichment analysis ($u(w_{235})$)**

$$w_{235} = \frac{A_{235}R_{235/238}}{A_{234}R_{234/238}+A_{235}R_{235/238}+A_{236}R_{236/238}+A_{238}}$$

$$R_{235/238} = \left(\frac{RS_{\frac{235}{238}}(m)RC_{\frac{235}{238}}(c)}{RC_{\frac{235}{238}}(m)}\right) - RB_{\frac{235}{238}}$$

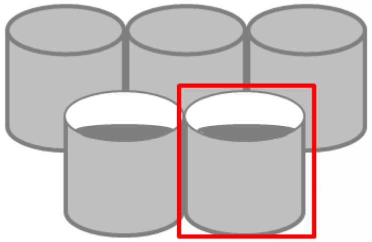
$$R_{23x/238} = \left(\frac{RS_{\frac{23x}{238}}(m)RC_{\frac{23x}{238}}(c)\delta(Y)\delta(L)\delta(F)}{RC_{\frac{23x}{238}}(m)}\right) - RB_{\frac{23x}{238}} \quad (x = 4, 6)$$

  – Assumptions:
    - Standard measurement procedure
    - Different detector types for isotopes (FC: ^{235}U, ^{238}U, SEM: ^{234}U, ^{236}U)



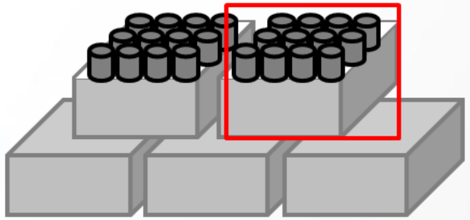
- **Benchmark case setup for evaluating correlation factors**
  - Compared $u(M(PE))$ **contribution between "with correlation"** and **"without correlation"**
    - KINAC's DA results were applied to quantify relative total, rand., sys. uncertainty $(\delta, \delta_r, \delta_s)$

UO_2 Powder



$N: 100,\ \bar{m}: 500\ kg,$
$f_u: 88.145\%, \overline{w_{235}}: 4.5\ \%$

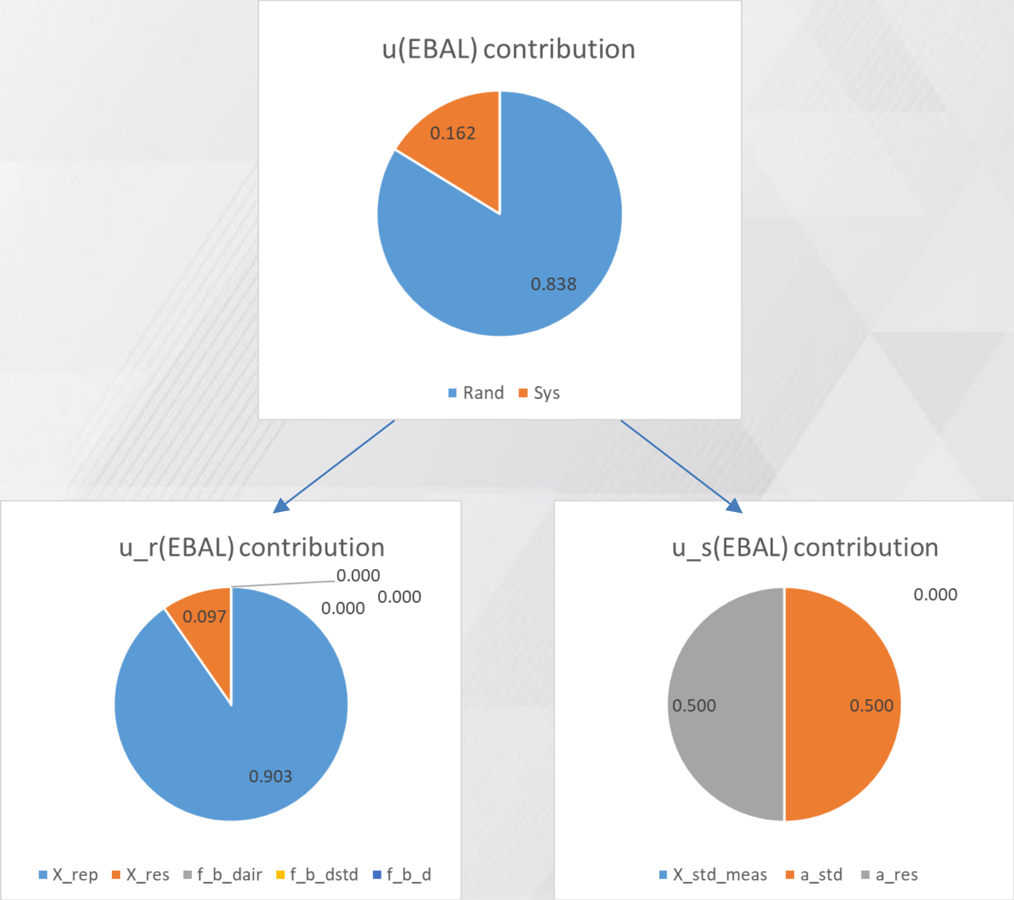
UO_2 Pellet



$N: 1{,}000,\ \bar{m}: 50\ kg,$
$f_u: 88.145\%, \overline{w_{235}}: 4.5\ \%$

	Combined Standard Uncertainty for Physical Inventory
With Correlation	$u(M) = \sqrt{\begin{array}{l}(\sum_{PD}^{N_{PD}} M_{PD}^2)(\delta_{rm(PD)}^2 + \delta_{rf_U(PD)}^2 + \delta_{rw_{235}(PD)}^2) \\ +(\sum_{PL}^{N_{PL}} M_{PL}^2)(\delta_{rm(PL)}^2 + \delta_{rf_U(PL)}^2 + \delta_{rw_{235}(PL)}^2) \\ +(\sum_{PD=1}^{N_{PD}} M_{PD})^2 \delta_{sm(PD)}^2 + (\sum_{PD=1}^{N_{PD}} M_{PD})^2 \delta_{sf_U(PD)}^2 \\ +(\sum_{PD=1}^{N_{PD}} M_{PD})^2 \delta_{sw_{235}(PD)}^2 + (\sum_{PL=1}^{N_{PL}} M_{PL})^2 \delta_{sm(PL)}^2 \\ +(\sum_{PL=1}^{N_{PL}} M_{PL})^2 \delta_{sf_U(PL)}^2 + (\sum_{PL=1}^{N_{PL}} M_{PL})^2 \delta_{sw_{235}(PL)}^2\end{array}}$
Without Correlation	$u(M) = \sqrt{\begin{array}{l}(\sum_{PD=1}^{N_{PD}} M_{PD}^2) \times (\delta_{m(PD)}^2 + \delta_{f_U(PD)}^2 + \delta_{w_{235}(PD)}^2) \\ +(\sum_{PL=1}^{N_{PL}} M_{PL}^2) \times (\delta_{m(PL)}^2 + \delta_{f_U(PL)}^2 + \delta_{w_{235}(PL)}^2)\end{array}}$

- **Relative uncertainty of weighing using EBAL**
  - Repeated measurement for 1 g sample
  - Constant temperature and pressure
  - Calibration using 1 g standard mass

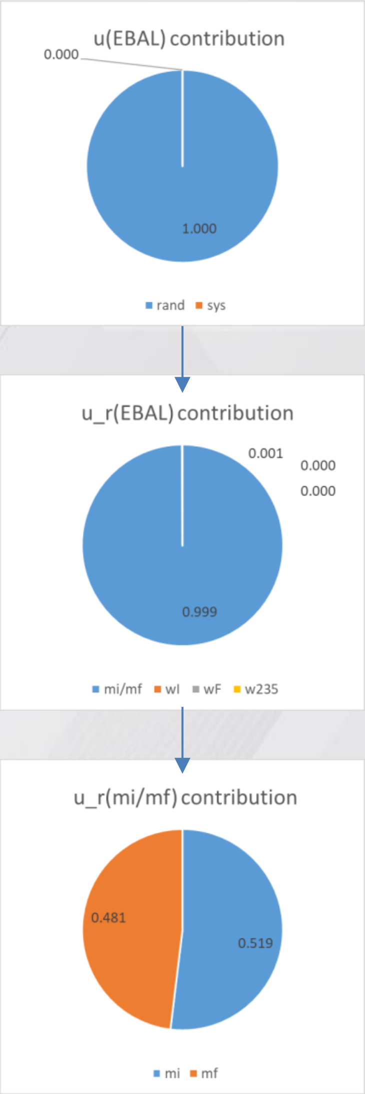


δ(m)	**u(m)**	**m**	X (rand)	f_b (rand)	f_c (sys)
1.014E-04	1.014E-04	1.0000	1.0001	1.0000	1.0000
δ_r(m)	**u_r(m)**	**nu(m)**	u(X)	u(f_b)	u(f_c)
9.279E-05	9.279E-05	3.4909	9.280E-05	8.476E-08	4.082E-05
δ_s(m)	**u_s(m)**		c(X)	c(f_b)	c(f_c)
4.082E-05	4.083E-05		1.0000	1.000	1.000
			cont(X)	cont(f_b)	cont(f_c)
			8.378E-01	6.992E-07	1.622E-01

03 Results

- **Relative uncertainty of weighing using GRAV**
  - Oxidation of 1.4 g pure UO_2 sample
  - External weighing of initial sample mass
  - Measurement of Δm before & after oxidation

δ(f_U)	**u(f_U)**	**f_U**	u_r(mi/mf)	u(w_I)	u(w_F)	u(w_235)
6.939E-04	6.116E-02	8.814E+01	6.672E-04	2.125E-05	5.952E-06	6.300E-05
δ_r(O/U)	**u_r(f_U)**	**nu(f_U)**	c(mi/mf)	c(w_I)	c(w_F)	c(w_235)
6.939E-04	6.116E-02	1.5280E+01	17.5308	-16.8669	16.8664	0.0072
			cont_r(mi/mf)	cont(w_I)	cont(w_F)	cont(w_235)
			9.990E-01	9.382E-04	7.359E-05	1.511E-09
δ_s(O/U)	**u_s(f_U)**		u_s(mi/mf)			
1.101E-06	9.707E-05		1.060E-06			
			c(mi/mf)			
			17.5308			
			cont_s(mi/mf)			
			2.519E-06			



03 Results

- **Relative uncertainty of weighing using TIMS**
  - Measuring 3.4 wt% UO_2 sample
  - Measurement of sample
  - Adjustment of isotopic ratio using a CRM
  - FC for $^{235,238}U$, SEM for $^{234,236}U$

δ(w_235)	**u(w_235)**	**w_235**	u_r(R234/238)	u_r(R235/238)	u_r(R236/238)
5.154E-04	1.747E-05	3.389E-02	6.866E-06	1.879E-05	1.849E-08
δ_r(w_235)	**u_r(w_235)**	**nu(w_235)**	c(R234/238)	c(R235/238)	c(R236/238)
5.109E-04	1.731E-05	5.458E+01	-0.03218	-0.92128	-0.03246
			cont(R234/238)	cont(R235/238)	cont(R236/238)
			1.600E-04	9.823E-01	1.180E-09
δ_s(w_235)	**u_s(w_235)**		u_s(R234/238)	u_s(R235/238)	u_s(R236/238)
6.826E-05	2.314E-06		2.959E-07	2.500E-06	2.508E-08
			c(R234/238)	c(R235/238)	c(R236/238)
			-0.03218	-0.92128	-0.03246
			cont(R236/238)		
			1.754E-02		

03 Results

- **Effect of correlation factors for MBE**
  - Comparing $u(M)$ between "with" and "without correlation"
    - Significant difference(~15%) between total uncertainty of M(^{235}U) in a BHF

u(M)						
0.214						
	u_r(M)	Contribution				
	0.180	0.707				
			u_r(PD)	Contribution		
			0.172	0.909		
					u_r(PD(m))	Contribution
					0.018	0.011
					u_r(PD(f_u))	Contribution
					0.138	0.641
					u_r(PD(w_235))	Contribution
					0.101	0.348
			u_r(PL)	Contribution		
			0.054	0.091		
					u_r(PL(m))	Contribution
					0.006	0.011
					u_r(PL(f_u))	Contribution
					0.044	0.641
					u_r(PL(w_235))	Contribution
					0.032	0.348
	u_s(M)	Contribution				
	0.116	0.293				
					u_s(EBAL_PD)	Contribution
					0.081	0.202
					u_s(GRAV_PD)	Contribution
					0.002	0.000
					u_s(TIMS_PD)	Contribution
					0.014	0.006
					u_s(EBAL_PL)	Contribution
					0.081	0.202
					u_s(GRAV_PL)	Contribution
					0.002	0.000
					u_s(TIMS_PL)	Contribution
					0.014	0.006

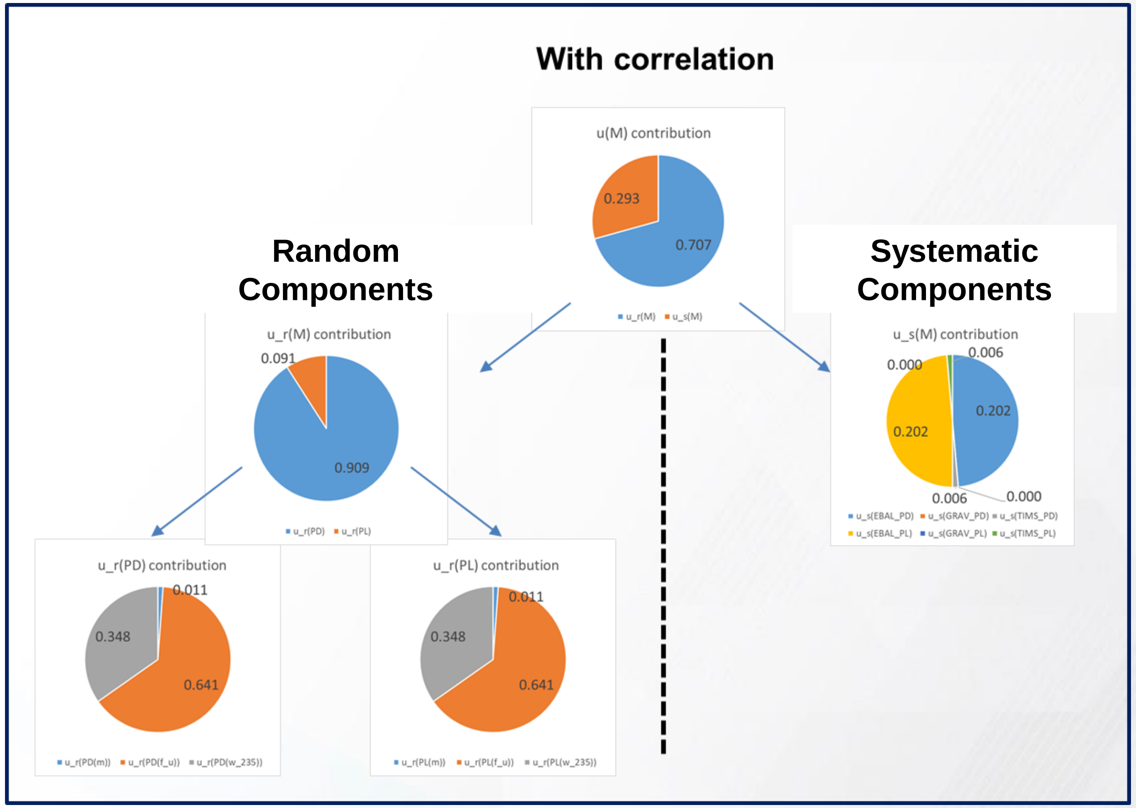
With correlation

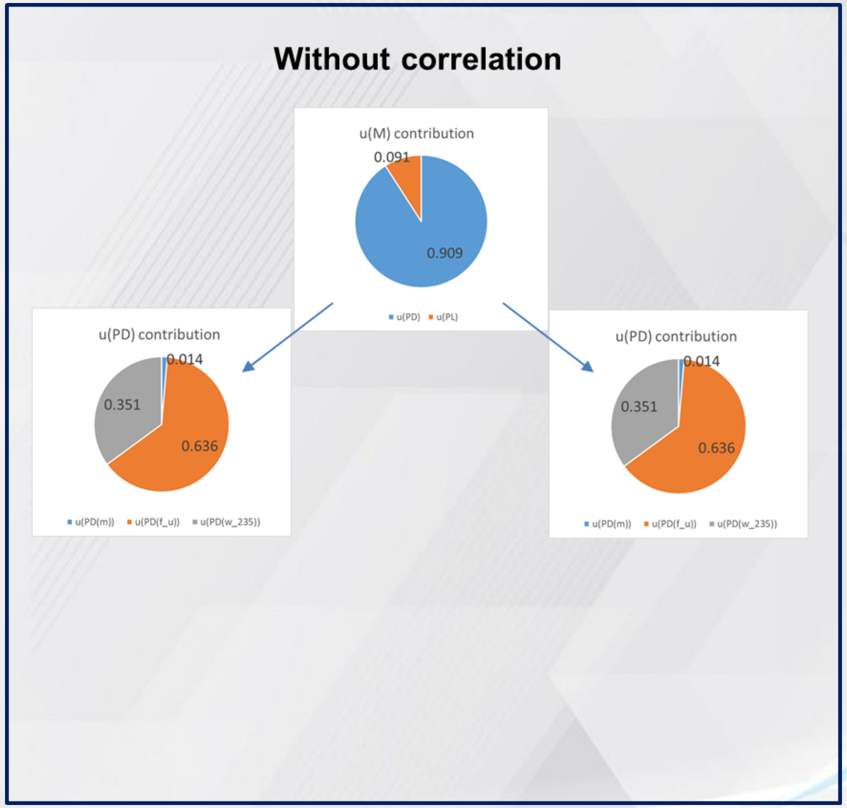
vs.

u(M)				
0.181				
	u(PD)	Contribution		
	0.173	0.909		
			u(PD(m))	Contribution
			0.020	0.014
			u(PD(f_u))	Contribution
			0.138	0.636
			u(PD(w_235))	Contribution
			0.102	0.351
	u(PL)	Contribution		
	0.055	0.091		
			u(PD(m))	Contribution
			0.006	0.014
			u(PD(f_u))	Contribution
			0.044	0.636
			u(PD(w_235))	Contribution
			0.032	0.351

Without correlation

- **Effect of correlation factors for MBE**
  - Contribution of uncertainty components between “with” and “without correlation”





- **Effect of correlation factors for MBE**
  - Contribution of uncertainty components between “item level” and “facility level”
    - **The contribution of correlation factors becomes significant in facility level**

	Uncertainty contribution (%)			
	Single item		Entire BHF	
	Rand.	Sys.	Rand.	Sys.
EBAL	83.78	16.22	4.91	95.09
GRAV	99.99	0.01	99.97	0.03
TIMS	98.25	1.75	98.25	1.75
Total	99.77	**0.23**	70.68	**29.32**

04 Conclusion

- The **propagation of uncertainty** in GUM **contains correlation factors**
- **It** has **usually** been **neglected in lab-scale** analytical procedures due to its small contribution to total uncertainty
- However, the **effect of correlation factors becomes significant if** the **large number of items are accumulated** and highly correlated
- Therefore, the **effect of correlation has to be considered** in the measurement of material balance **in a large bulk handling facility**
- Future works will demonstrate the effect of the entire material balance evaluation

Q&A

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