

The effect of random perturbations on the performance of tank electrolyzers

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The effect of stationary random perturbations in the electric current on the fluctuation of active ion concentration in the electrolyte in electrolyzers amenable to CSTER-based analysis is examined via the theory of stochastic differential equations. The approach to random fluctuations in other inputs can be readily extended.

Notation			
c	active ion concentration; c_0 its initial value in a batch electrolyser; c_i its inlet value in a CSTER	V_R	active electrolyser volume
F	Faraday's constant	X	random input signal
I	electric current	Y	random output signal; conversion
i	imaginary operator; $i \equiv \sqrt{-1}$	z	valency
K_{VW}	(cross) covariance function of random variables V and W	α	lumped parameter defined under Equation 2
K_{WW}	auto-covariance function of random variable W	β	lumped parameter defined under Equation 2
Q	volumetric flow rate	γ	parameter in the Markovian and quasi-Markovian input autocovariance function
$S_W(\omega)$	power spectrum of random variable W	η	current efficiency
t	time; t_1 and t_2 arbitrary time instants; t_m mean residence time; t_R rise time	μ_W	mean (or expectation) of random variable W
		σ_W^2	variance of random variable W
		ω	frequency

1. Introduction

The behaviour of tank electrolyzers in the presence of random perturbations occurring in their operating variables (inflow electrolyte concentration, current, etc.) has been considered only to a limited extent in terms of signal recognition techniques [1] and Markov-chain theory [2], with the primary purpose of parameter identification, and the finding of equivalent analytical models. A complementary – and equally important – problem is the prediction of electrolyser behaviour when a random perturbation in an input variable has certain stochastic characteristics: under appropriate conditions the stochastic characteristics of the output variables (e.g. outlet electrolyte concentration) can be predicted by means of an analytical model. The purpose of the current paper is to present such an approach by considering specifically how stochastic perturbations in the electric current affect the conversion of an active ion in the electrolyte to the product of an electrode reaction. Three systems – a batch electrolyser, a continuous-flow electrolyser, and a two-element continuous-flow electrolyser cascade – serve as working examples. It is assumed that the CSTER concept [3–6] can be employed at least approximately as the mathematical tool for the modelling of

electrolyser behaviour in order to arrive at relatively unencumbered analytical results. The findings are expected to be useful for the rational design of tank electrolyzers.

2. Theoretical background

The structures of interest are ordinary linear differential equations

$$\frac{dY}{dt} = X(t) \quad \text{for } t \geq 0 \quad (1)$$

$$\frac{dY}{dt} + \alpha Y = X(t) \quad \text{for } t \geq 0 \quad (2)$$

with initial condition $Y(0) = 0$. As discussed in the related literature (for example, [7] and [8]), if $X(t)$ is random, solutions of Equations 1 and 2 exist in the mean-square sense. In this manner, μ_y and μ_x can replace Y and X , respectively, and the covariance functions (see Appendix 1) $K_{XY}(t_1, t_2)$ and $K_{YY}(t_1, t_2)$ are obtained by solving differential equations isomorphic to Equations 1 and 2, specifically,

$$\frac{\partial K_{XY}(t_1, t_2)}{\partial t_2} = K_{XX}(t_1, t_2) \quad K_{XY}(t_1, 0) = 0 \quad (3a)$$

$$\frac{\partial K_{YY}(t_1, t_2)}{\partial t_1} = K_{XY}(t_1, t_2) \quad K_{YY}(0, t_2) = 0 \quad (3b)$$

for a system governed by Equation 1, and

$$\frac{\partial K_{XY}(t_1, t_2)}{\partial t_2} + \alpha K_{XY}(t_1, t_2) = K_{XX}(t_1, t_2) \quad K_{XY}(t_1, 0) = 0 \quad (4a)$$

$$\frac{\partial K_{YY}(t_1, t_2)}{\partial t_1} + \alpha K_{YY}(t_1, t_2) = K_{XY}(t_1, t_2) \quad K_{YY}(0, t_2) = 0 \quad (4b)$$

for a system governed by Equation 2. Complete solution involves the splitting of the integration procedure in two steps, covering the time domains $0 \leq t_2 \leq t_1$; $t_2 \geq t_1$ for Equations 3a and 4a, and $0 \leq t_1 \leq t_2$; $t_1 \geq t_2$ for Equations 3b and 4b. The concomitant initial conditions are, therefore, the values of $K_{XY}(t_1, t_1)$ and $K_{YY}(t_2, t_2)$ for the second domains. The long-term behaviour of the autocovariance function is directly related to the standard deviation of the output $Y(t)$ around its mean value of steady-state conditions:

$$\sigma_Y = [K_{YY}(\tau)]_{\tau=0}^{1/2} \quad (5)$$

where

$$\lim_{\substack{t_1 \rightarrow \infty \\ t_2 \rightarrow \infty}} (t_1, t_2) = K_{YY}(\tau) \quad (6)$$

In this manner, the autocovariance function represents a measure of dispersion of the system response to a stationary random perturbation and it is a prime characteristic of stochastic system performance. A

selected number of autocovariance functions have been assembled in Table 1: for the system described by Equation 2, K_{YY} is an extremely complex function of time instants t_1 and t_2 , in the case of Markovian and other input-covariance functions K_{XX} , hence it is omitted. On the other hand, the long-time behaviour in these cases may be established at relative ease by replacing the path via Equation 6 with the path involving the power spectrum, i.e. the Fourier integral of the covariance function, as shown in Appendix 2. The results for the systems discussed in this paper are summarized in Table 2. Equation 1, as a pure integrator, does not reach a steady state and is excluded from the Table; conversely, the equation

$$\frac{d^2 Y}{dt^2} + a_1 \frac{dY}{dt} + a_2 Y = b_0 \frac{dX}{dt} + b_1 X \quad (7)$$

is included on account of its governance of the behaviour of the two-element CSTER cascade. In contrast to the analytical expressions for $K_{YY}(0)$ in the case of a first order system, the output standard deviation has to be obtained by a numerical solution of the $K_{YY}(0)$ integrals (the solution for a quasi-Markovian input shows an interesting application of sine and cosine integrals).

3. Application to electrolyser behaviour

3.1. Batch electrolyser

Under isothermal conditions, the mole balance in terms of the active ion concentration

$$-V_R \frac{dc}{dt} = \eta \frac{I}{zF} \quad (8)$$

Table 1. Autocovariance functions of output $Y(t)$ for selected forms of random inputs $X(t)$

System	$K_{XX}(\tau)$	$K_{YY}(t_1, t_2)$
Equation 1	$\sigma_x^2 \delta(\tau)$ (white noise)	$\sigma_x^2 t_1$; $0 \leq t_1 \leq t_2$ $\sigma_x^2 t_2$; $t_1 \geq t_2$
	$\sigma_x^2 e^{-\gamma \tau }$ (Markovian)	$\frac{\sigma_x^2}{\gamma} \left[2t_1 + \frac{1}{\gamma} \{e^{-\gamma t_1} + e^{-\gamma t_2}(1 - e^{-\gamma t_1}) - 1\} \right]$ $0 \leq t_1 \leq t_2$ $\frac{\sigma_x^2}{\gamma} \left[2t_2 - \frac{1}{\gamma} (e^{-\gamma t_1} + e^{-\gamma t_2} - e^{-\gamma(t_1-t_2)}) - 1 \right]$ $t_1 \geq t_2$
	$\sigma_x^2 e^{-\gamma \tau^2}$ (Quasi-Markovian)	$\frac{\sigma_x^2 \sqrt{\pi}}{2\gamma} \left[t_1 \operatorname{erf}(t_1 \sqrt{\gamma}) - \frac{1}{\gamma \sqrt{\pi}} (1 - e^{-\gamma t_1^2}) \right]$ $0 \leq t_1 \leq t_2$ $\frac{\sigma_x^2 \sqrt{\pi}}{2\gamma} [t_1 \operatorname{erf}(t_1 \sqrt{\gamma}) - (t_1 - t_2) \operatorname{erf}(t_1 - t_2) \sqrt{\gamma}]$ $+ \frac{1}{\gamma \sqrt{\pi}} (e^{-\gamma t_1^2} - e^{-\gamma(t_1-t_2)^2})$; $t_1 \geq t_2$
Equation 2	$\sigma_x^2 \delta(\tau)$	$\frac{\sigma_x^2}{\alpha} e^{-\alpha t_2} \sinh(\alpha t_1)$; $0 \leq t_1 \leq t_2$ $\frac{\sigma_x^2}{2\alpha} [1 - e^{-2\alpha t_2}] e^{-\alpha(t_1-t_2)}$; $t_1 \geq t_2$

Table 2. Long-time behaviour of the output autocovariance function for selected forms of random inputs

System	$K_{XX}(\tau)$	$K_{YY}(\tau)$	$K_{YY}(0)$
Equation 2	$\sigma_x^2 \delta(\tau)$	$\frac{\sigma_x^2}{2a} e^{-a \tau }$	$\frac{\sigma_x^2}{2a}$
	$\sigma_x^2 e^{-\gamma \tau }$	$\frac{\sigma_x^2}{\alpha} \frac{e^{-\gamma \tau } - \gamma e^{-a \tau }}{\alpha^2 - \gamma^2}$	$\frac{\sigma_x^2}{\alpha(\gamma + \alpha)}$
	$\sigma_x^2 e^{-\gamma\tau^2}$	$\frac{\sigma_x^2}{\sqrt{\pi\gamma}} \int_0^\infty \frac{e^{\omega^2/4\gamma} \cos \tau\omega}{\alpha^2 + \omega^2} d\omega$	$\frac{\sigma_x^2}{2\alpha} \frac{\sqrt{(\pi)}}{\gamma} e^{\alpha^2/4\gamma} \operatorname{erf} c \left(\frac{\alpha}{2\sqrt{(\gamma)}} \right)^*$
Equation 7	$\sigma_x^2 \delta(\tau)$	$2\sigma_x^2 \int_0^\infty \frac{b_0^2 \omega^2 + b_1^2}{(a_2 - \omega^2)^2 + a_1^2 \omega^2} e^{i\tau\omega} d\omega$	
	$\sigma_x^2 e^{-\gamma \tau }$	$\frac{2\sigma_x^2 \gamma}{\pi} \int_0^\infty \frac{(b_0^2 \omega^2 + b_1^2) e^{i\tau\omega} d\omega}{[a_2 - \omega^2]^2 + a_1^2 \omega^2} [\gamma^2 + \omega^2]$	To obtain $K_{YY}(0)$ set $\tau = 0$ in all three integrals
	$\sigma_x^2 e^{-\gamma\tau^2}$	$\frac{\sigma_x^2}{\sqrt{(\pi\gamma)}} \int_0^\infty \frac{e^{\omega^2/4\gamma} (b_0^2 \omega^2 + b_1^2)}{(a_2 - \omega^2)^2 + a_1^2 \omega^2} e^{i\omega\tau} d\omega$	

$$*\operatorname{erf} c(x) \equiv 1 - \frac{2}{\sqrt{(\pi)}} \int_0^x e^{-t^2} dt \quad [9]$$

may be rewritten for conversion Y as $dY/dt = X(t)$, as given in Equation 1, where $Y \equiv (c_0 - c)/c_0$ and $X \equiv \eta I/zFc_0 V_R$. As indicated in the previous section, Equation 1 has a solution in the mean-square sense, i.e. the mean value of the conversion is obtained by solving the isomorphic equation

$$\frac{d\mu_Y}{dt} = \mu_X = \eta\mu_I/zFc_0 V_R \quad (9)$$

to

$$\mu_Y = \mu_X t \quad (10)$$

Further, since

$$\sigma_X^2 = \eta^2 \sigma_I^2 / (zFc_0 V_R)^2 \quad (11)$$

the forms of the autocovariance function given in Table 1 apply equally to current and the variables X . Typical numerical values shown in Table 3 depict the evolution of the conversion autocovariance function with increasing time whose square-root is the standard deviation of the conversion around its mean

Table 3. The evolution of the conversion autocovariance function in a typical batch electrolyser ($c_i = 3 \text{ mol m}^{-3}$; $z = 2$, $V_R = 0.16 \text{ m}^3$; $\mu_I = 1 \text{ kA}$; $\sigma_I^2 = 25 \text{ A}^2$) employed for trace metal recovery; $t_1 = t_2 = t$

Time (min)	$\sqrt{K_{YY}(t)}$		
	White noise	Markovian ($\gamma = 0.02 \text{ min}^{-1}$)	Quasi-Markovian ($\gamma = 0.02 \text{ min}^{-1}$)
0.1	0.001025	0.0003239	0.0004528
1.0	0.003240	0.0035288	0.004528
10.0	0.010247	0.031359	0.040000
60.0	0.025100	0.238960	0.125658
100.0	0.032404	0.244130	0.162234

Asymptotic tendency: $K_{YY}(t) \rightarrow 0.00105t - 0.0525$ (Markovian)
 $K_{YY}(t) \rightarrow 0.00026319t - 28.21$ (Quasi-Markovian)

(Equation 10) at any given time. The largest increase in dispersion with time is exhibited in the case of the Markovian current indicating the necessity of terminating batch electrolysis in such a case after a short time, in order to avoid relatively large fluctuations of performance from batch to batch.

3.2. Continuous flow electrolyser (CSTER)

Under isothermal conditions, the mole balance

$$V_R \frac{dc}{dt} = Qc_i - Qc - \frac{\eta I}{zF} \quad (12)$$

may be rewritten, in a fashion similar to the batch electrolyser, as

$$\frac{dY}{dt} + \alpha Y = X = \beta I \quad (2)$$

where $\alpha \equiv Q/V_R$ and $\beta \equiv \eta/zFc_i V_R$; α is recognized as the reciprocal of the mean residence time of fluid elements in the tank. Due to the mathematical complexity of the evolution equations for $K_{YY}(t_1, t_2)$, Table 4 contains only white noise-excited behaviour, as a function of the electrolyte volumetric throughput rate. The evolution relationship in the case of white-noise fluctuations in the current is given by

$$K_{YY}(t, t) = \frac{\sigma_X^2}{2\alpha} (1 - e^{-2\alpha t}) \quad (13)$$

The times corresponding to the rise time $t_R = 1/2\alpha$ and mean residence time $t_m = 1/\alpha$ serve as convenient points of reference, where

$$K_{YY}(t_R, t_R) \simeq 0.632 t_m \sigma_X^2 \quad (14a)$$

and

$$K_{YY}(t_m, t_m) \simeq 0.432 t_m \sigma_X^2 \quad (14b)$$

demonstrate the effect of flow rate: as Q increases, α

Table 4. Parameters of the transient behaviour of a CSTER under white-noise current fluctuation: $c_1 = 3 \text{ mol m}^{-3}$; $z = 2$; $V_R = 0.16 \text{ m}^3$; $\mu_I = 1 \text{ kA}$; $\sigma_I^2 = 25 \text{ A}^2$

Q ($\text{m}^3 \text{ h}^{-1}$)	α (min^{-1})	t_R (min)	t_m (min)	$\sqrt{K_{YY}(t_R)}$	$\sqrt{K_{YY}(t_m)}$
7	0.729	0.686	1.372	0.002135	0.002493
8	0.833	0.600	1.200	0.001995	0.002334
9	0.937	0.534	1.067	0.001881	0.002200
10	1.041	0.480	0.961	0.001784	0.002087

becomes larger, the mean residence time and rise time become smaller, the evolution becomes faster, resulting also in a smaller fluctuation of the conversion about its mean. This result shows clearly that relatively long transient dynamics in a CSTER are deleterious from the point of view of performance.

The long-term behaviour of the CSTER performance is shown in Table 5, where the conversion fluctuation range defined as $\mu_Y \pm \sqrt{K_{YY}(0)}$ is tabulated as a function of flow rate and the reciprocal of the mean residence time. The mean conversion is obtained by straightforward integration of Equation 2 (in the mean-square sense) as

$$\mu_Y = \frac{\mu_X}{\alpha} (1 - \varepsilon^{-\alpha}) \tag{15a}$$

and it follows that after a sufficiently long time lapse,

$$\mu_Y \longrightarrow \frac{\mu_X}{\alpha} \tag{15b}$$

3.3. Two-element CSTER cascade

The isothermal mole balance equations

$$V_1 \frac{dc_1}{dt} = Qc_1 - Qc_1 - \frac{\eta_1 I_1}{zF} \tag{16a}$$

$$V_2 \frac{dc_2}{dt} = Qc_1 - Qc_2 - \frac{\eta_2 I_2}{zF} \tag{16b}$$

govern the behaviour of the system. In terms of elements conversions Y_1 and Y_2 and related lumped parameters $\alpha_1 \equiv Q/V_1$; $\alpha_2 \equiv Q/V_2$; $\beta_1 \equiv \eta_1/zFc_1V_1$ and $\beta_2 \equiv \eta_2/zFc_1V_2$, the relationship

$$\begin{aligned} \frac{d^2 Y_2}{dt^2} + (\alpha_1 + \alpha_2) \frac{dY_2}{dt} + \alpha_1 \alpha_2 Y_2 \\ = \alpha_2 \beta_1 I_1 + \alpha_1 \beta_2 I_2 + \beta_2 \frac{dI_2}{dt} \end{aligned} \tag{17}$$

relates conversion from the second CSTER to time. In the practical case of $I_1 = I_2$ and in terms of the additional lumped parameters $a_1 \equiv \alpha_1 + \alpha_2$; $a_2 \equiv \alpha_1 \alpha_2$, $b_0 \equiv \beta_2$ and $b_1 \equiv \alpha_2 \beta_1 + \alpha_1 \beta_2$, the conversion autocovariance function for the second CSTER is obtained at sufficiently long times by taking the inverse Fourier transform of its power spectrum:

$$K_{Y_2 Y_2}(\tau) = \int_{-\infty}^{+\infty} \frac{(b_0^2 \omega^2 + b_1^2) S_I(\omega)}{(a_2 - \omega^2)^2 + a_1^2 \omega^2} e^{i\tau\omega} d\omega \tag{18}$$

where $S_I(\omega)$ is the power spectrum of the current; in Table 2 specific forms of the integral in Equation 18 are shown for the random current perturbations considered. The effect on overall cascade performance is illustrated in Table 6; the analysis of first element performance follows closely the single CSTER case and is not repeated here.

4. Discussion

The numerical illustrations demonstrate the strategy of estimating the effect of random perturbations in the current on the performance of certain tank electrolyser systems. The approach can be readily extended to various configurations: in the specific case of an N-element CSTER cascade for example, the integral expression in Equation 18 carries an N-th order polynomial of ω in the denominator and an (N - 1)st order polynomial of ω in the numerator. The numerical handling of integration would pose little additional encumbrance for a microcomputer, unless N is very large (an impractical case).

The numerical similarity of the oscillation ranges shown in Tables 5 and 6 is fortuitous inasmuch as the range is determined by the magnitude of the parameters of the current autocovariance functions. Depending on these numerical values, the oscillation ranges may be widely different: a systematic numerical simulation of performance, beyond the scope of this paper, would enable the designer to identify random input functions against which appropriate control action is required to suppress unacceptably large fluctuations of the exit conversion.

The method of attack can be applied in a similar fashion to random perturbations in other inputs, for example, the inlet electrolyte concentration in a flow electrolyser. If the current remains constant, the

Table 5. Long-term performance of the CSTER*

Q ($\text{m}^3 \text{ h}^{-1}$)	α (min^{-1})	μ_Y	Range of conversion fluctuation		
			White-noise	Markovian $\gamma = 0.02 \text{ min}^{-1}$	Quasi-Markovian $\gamma = 0.02 \text{ min}^{-1}$
7	0.729	0.889	0.886-0.892	0.885-0.893	0.886-0.892
8	0.833	0.778	0.775-0.780	0.774-0.782	0.776-0.780
9	0.937	0.692	0.690-0.694	0.688-0.696	0.690-0.694
10	1.041	0.622	0.620-0.624	0.619-0.625	0.620-0.624

*Numerical parameters as in Table 4.

Table 6. Long-term performance of a two-element CSTER cascade. ($c_i = 3 \text{ mol m}^{-3}$; $z = 2$; $V_1 = V_2 = 0.16 \text{ m}^3$; $\mu_I = 500 \text{ A}$; $\sigma_I^2 = 6.25 \text{ A}^2$)

Q ($\text{m}^3 \text{ h}^{-1}$)	α (min^{-1})	μ_{Y_1}	μ_{Y_2}	Range of conversion fluctuation*		
				White-noise	Markovian $\gamma = 0.02 \text{ min}^{-1}$	Quasi-Markovian $\gamma = 0.02 \text{ min}^{-1}$
7	0.729	0.444	0.888	0.886–0.890	0.884–0.892	0.882–0.894
8	0.833	0.389	0.777	0.775–0.779	0.773–0.781	0.772–0.782
9	0.937	0.346	0.691	0.689–0.693	0.687–0.694	0.686–0.696
10	1.041	0.311	0.622	0.620–0.624	0.619–0.625	0.617–0.627

*The integrals shown in Table 2 were evaluated by a Romberg quadrature method [10] to the upper limit of $\omega = 200$, ensuring a minimum of four decimal accuracy.

forcing function $X(t)$ in Equation 2 comprises the constant term $\eta I / VzF$ and the randomly fluctuating term αc_i . The solution of Equation 2, for initial condition $t = 0$ $c = c_i$, in terms of mean quantities:

$$\mu_c = \mu_{c_i} - \frac{\eta I}{zFV_R \alpha} (1 - e^{-\alpha t}) \quad (19)$$

is obtained via conventional techniques of integrating first order linear differential equations. If both c_i and I are random inputs with means μ_{c_i} and μ_I , respectively, the solution of Equation 2 may be written as

$$\mu_c = \mu_{c_i} - \frac{\eta \mu_I}{zFV_R \alpha} (1 - e^{-\alpha t}) \quad (20)$$

The treatment of the associated covariance functions follows similarly.

Finally, it is worth noting that although $X(t)$ is stipulated to be a stationary random process (strictly speaking, wide-sense stationary (WSS), i.e. its stationarity involves only its means and covariance functions), the zero initial conditions appearing in the solution of Equations 2–4 create transient nonstationarities, which vanish, however, at large times if the system reaches a steady state [8]. This can be seen also by the output autocovariance function reaching a time-independent form $K_{YY}(\tau)$, indicating that tank electrolyzers subjected to stationary random perturbations are asymptotically WSS. Batch electrolyzers do not belong to this class since they do not operate under steady-state conditions.

5. Conclusions

The foregoing analysis portrays the application of an elementary theory of stochastic processes to performance studies of electrolyzers. The full scope of this approach, relatively new to electrochemical engineering, will require a thorough exploration in future investigations.

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Appendix 1

The covariance function of real random variables X and Y with means μ_X and μ_Y , respectively, is defined as

$$K_{XY}(t_1, t_2) = E[(X(t_1) - \mu_X(t_1))(Y(t_2) - \mu_Y(t_2))] \quad (A.1)$$

where E is the expectation operator. The definition of the autocovariance function of a real random variable X follows directly:

$$K_{XX}(t_1, t_2) = E[(X(t_1) - \mu_X(t_1))(X(t_2) - \mu_X(t_2))] \quad (A.2)$$

If the process involving the random variable X is stationary [i.e., $X(t + \tau)$ has the same probability distribution function for all τ], Equation A.2 may be simplified to

$$K_{XX} = E(X(t + \tau)X(t)) - \mu_X^2 \quad (A.3)$$

where the expectation term is known as the correlation function of random variable X . The important corollary for the variance:

$$\sigma_X^2 = K_{XX}(0) = E(X^2) - \mu_X^2 \quad (A.4)$$

is a fundamental theorem in probability theory.

Appendix 2

Let a stationary linear system be represented by the differential equation with constant coefficients.

$$\sum_{k=0}^N a_k \frac{d^k Y}{dt^k} = \sum_{j=0}^M b_j \frac{d^j X}{dt^j} \quad (A.5)$$

and let the power spectrum of the input be $S_X(\omega)$. Then, the power spectrum of the output may be expressed [9] as

$$S_Y(\omega) = \frac{\left| \sum_{j=0}^M b_j (i\omega)^j \right|^2}{\left| \sum_{k=0}^N a_k (i\omega)^k \right|^2} S_X(\omega) \quad (A.6)$$

It follows that the asymptotic form of the output autocovariance function, a measure of long-term behaviour, may be obtained from the known input autocovariance function in two steps. First, $S_X(\omega)$ is

obtained by Fourier transformation:

$$S_X(\omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} K_{XX}(\tau) e^{-i\omega\tau} d\tau \quad (\text{A.7})$$

then $S_Y(\omega)$ is obtained by Equation (A.6), whose inverse Fourier transform yields

$$K_{YY}(\tau) = \int_{-\infty}^{+\infty} S_Y(\omega) e^{i\tau\omega} d\omega \quad (\text{A.8})$$

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