

Prediction of steel corrosion in magnesium cement concrete based on two dimensional Copula function

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Abstract. In order to solve the life prediction problem of damaged coating steel bar in magnesium cement concrete, this study tries to establish the marginal distribution function by using the corrosion current density as a single degradation factor. Representing the degree of steel corrosion, the corrosion current density were tested in electrochemical workstation. Then based on the Copula function, the joint distribution function of the damaged coating was established. Therefore, it is indicated that the corrosion current density of the bare steel and coated steel bar can be used as the boundary element to establish the marginal distribution function. By using the Frank-Copula function of Copula Archimedean function family, the joint distribution function of the damaged coating steel bar was successfully established. Finally, the life of the damaged coating steel bar has been lost in 7320d. As a new method for the corrosion of steel bar under the multi-dimensional factors, the two-dimensional Copula function has certain practical significance by putting forward some new ideas.

Keywords: corrosion current density; marginal distribution function; joint distribution function; Frank-Copula function; life prediction

1. Introduction

Most regions of western China belong to saline soil and salt lake areas (Xu 1993), where has large temperature difference between day and night and windy and dusty weather, therefore, the ordinary concrete infrastructure is difficult to meet the design requirements to achieve the scheduled working life. One of the most important factors is that the saline soil and salt lake area contain a lot of chloride, sulfate and other harmful salts which plays a determined role in the service life of concrete (Wen 2010). To be specific, chloride accelerates the hydration rate of silicate and destroys the passive film of steel in ordinary concrete as a strong depassivator, thus accelerating steel corrosion and concrete cracking, then losing its working performance prematurely. For ordinary concrete, chloride corrosion accounts for 33%, while internal chloride salt (from the raw materials of the concrete) corrosion accounts for 5% (Department of civil engineering and architecture, Chinese Academy of Engineering 2004). Magnesium

oxychloride cement is a kind of magnesia cementitious material composed by $\text{MgO-MgCl}_2\text{-H}_2\text{O}$ system, which has a good performance of bittern-resistance without modification (Cao 2004, Wen 2013, Zhang 2014). However, due to this characteristic, the low corrosion to steel bar is the main reason to make its popularization and application greatly limited (Ma 2015). The coating can solve the corrosion problem of steel bars in magnesium cement concrete very well.

At present, the prediction model of chloride includes the model that critical chloride ion concentration has been used as the main parameter to reflect the corrosion situation and the probabilistic model of corrosion time based on probabilistic statistic method. Qiao (2017) used the Weibull distribution and the lognormal distribution to assess the durability of concrete structures. Anoop (2016) used the Brunswikian theory and probabilistic mental models to evaluate the residual life of reinforced concrete bridge subjected to chloride ion. Tian (2016) used the method of probabilistic and finite element to evaluate and predict the lifetime performance of reinforced concrete (RC) bridges undergoing various maintenance actions. The prediction of initiation time of corrosion in RC is studied by Yao (2015). Zheng (2009) studied and established the probability model of the initial corrosion time of steel bar based on the Fick's second law. Based on probability analysis and statistics, the pitting corrosion of steel bar surface was investigated and studied, and the concerned model of corrosion rate was obtained by Wang (2012). The model of the above-mentioned steel bar corrosion prediction is based on the bare steel bars, which can not be applied to the corrosion

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Table 1 Mixing proportion of magnesium oxychloride cement concrete (kg/m³)

MgO (kg/m ³)	water reducer (kg/m ³)	water repellent (kg/m ³)	sand (kg/m ³)	cobblestone (kg/m ³)	MgCL ₂ (kg/m ³)	water (kg/m ³)	slum (mm)
388.96	2.288	1.6016	625	1162	147.811	135.586	120

prediction of coated steel bars.

The coating provides protection for the steel bar by sacrificing anode to provide cathodic protection. The zinc flake scale in the coating produces a corrosion material (Zn₅(OH)₆(CO₃)₂) during the protection process, which in turn makes the coating recover. Therefore, there is a fundamental difference between the degenerative process of the durability about coated steel bar and that of bare steel bar. Although the coating can protect steel bar from corrosion well, in the practical engineering, the coating can be damaged to a certain degree by the interference of external factors (coated steel bar produces horizontal and vertical cracks in the process of tension or compression). So the corrosion prediction of damaged coated steel bars, plays an important role in the popularization and application of magnesium cement coated reinforced concrete. As a joint distribution function, Copula function can be combined with multivariate marginal distribution function to construct multi-dimensional joint distribution. Therefore, the joint distribution function of the damaged coating steel bar can be obtained by using the bare steel and coated steel as the marginal distribution functions, and finally the corrosion prediction of the damaged coated steel bar has been realized.

Aiming to solve the steel bar corrosion problem of reinforced magnesium cement concrete, this paper tested the corrosion current density of steel corrosion by using the electrochemistry principle. Then, based on the Copula function, the prediction model of damaged coated steel bar corrosion in magnesium cement concrete was established. Finally, the corrosion of steel bars in magnesium cement concrete was predicted.

2. Experiment

2.1 Experimental materials

The raw materials of magnesium cement reinforced concrete are mainly composed of light-burned magnesium oxide (MgO), magnesium chloride (MgCL₂), water reducing agent, water repellent agent, fly ash, sand, gravel and steel bars. Magnesium oxide (MgO) is light-burned. MgO and MgCL₂ are both produced by Qarhan Salt Lake Magnesium chloride plant, Golmud City, Qinghai Province. River sand comes from Shuifu, Lanzhou, which is well graded, belonging to the medium level. Gravel is provided by Gansu Hualong Concrete Co. Ltd., which belongs to the continuous gradation, performance indicators qualified. The I grade fly ash used in this study is produced by a steel mill in Lanzhou (in order to improve the durability of concrete). Water proofing agent is phosphoric acid from Tianjin Baishi Chemical Co., Ltd., and the content of H₃PO₄

Table 2 Chemical constituents of fly ash

MgO	CaO	Fe ₂ O ₃	Al ₂ O ₃	SO ₃	Ignition loss	SiO ₂	Other
1.19	5.30	9.43	20.93	0.41	3.26	54.32	5.16

Table 3 The physical indicators of water reducing agent

species	Density value /g/ml	PH	Alkali content /%	Water reduction Rate/%	Bleeding rate/%	Recommended dosage
PCA(I)	0.0003	8.08	≤3.88	34	0	0.02

Table 4 The corresponding relationship of corrosion current density and corrosive degree of rebars

$i_{cor}/(\mu A \cdot cm^{-2})$	$i_{cor} < 0.1$	$0.1 < i_{cor} < 0.5$	$0.5 < i_{cor} < 1$	$i_{cor} > 1$
Corrosion situation	No corrosion	Low corrosion	Moderate corrosion	Severe corrosion

is not less than 85%; the Hazen Unit is less than 25. The water reducing agent belongs to KD naphthalene series with high efficiency. The tap water is in accordance with the industry standard and requirements of "Standard Specification for concrete mixing water" JGJ63-2006. HPB300 steel bar is employed and its yield strength is $f_y=300$ N/mm². The Japan's GEOMET coating (a kind of zinc coating that includes a large number of ultrafine zinc-aluminum flake) is provided by Ningbo Metal Surface Treatment Co. Ltd.. The mixing ratio of magnesium cement concrete is shown in Table 1. The Chemical constituents of fly ash are shown in Table 2. The physical indicators of water reducing agent is shown in Table 3.

2.2 Experimental program

In this experiment, the length of HPB300 steel bar is 105mm and the diameter is 8mm. The steel bars were divided into two groups according to the presence of coating: Group A (bare steel bars), group B (coated steel bars). Then, according to the concrete protective layer of 25 mm, the two groups of steel bars were placed in a magnesium cement concrete test pieces whose size is 100 mm×100 mm×100 mm referring to the mixing proportion of Table 1. The test blocks were put into the magnesium oxychloride solution with chloride ion concentration of 1.5 mol/L, and the height of the solution was 2/3 of block. Electrochemical experiments were carried out by CS350 electrochemical work station. Bare and coated steel is the working electrode, whose electrode area covers is 25.12 cm². Thin stainless steel plate works as an auxiliary electrode, whose electrode area is 30 cm², greater than the working electrode area. Saturated KCL electrode is acted as reference electrode. AC impedance measurement frequency range is 0.01 Hz-100000 Hz, and AC positive rotation excitation signal amplitude is 10 mV, meanwhile, the AC impedance test uses metal shielding for electromagnetic shielding. The test was carried out every 90 days and focused on its polarization curve and AC impedance parameters. The relationship between corrosion current density and corrosion of steel bar is shown in Table 4 (Luo 2002). The experimental flow chart is shown in Fig. 1.

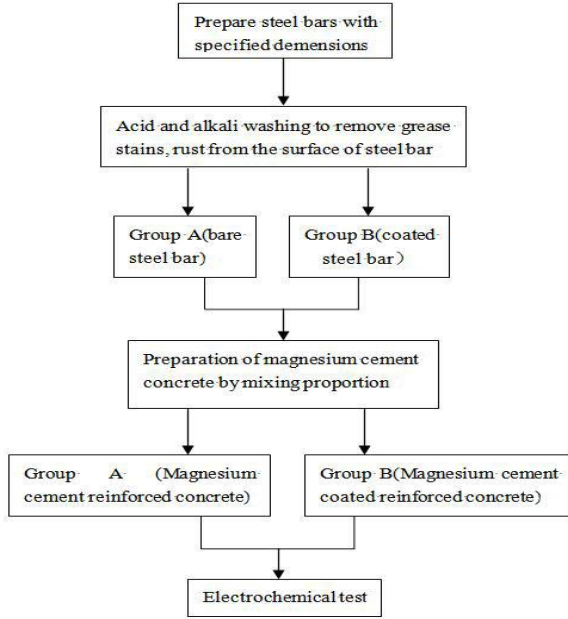


Fig. 1 Experimental flow chart

3. Relevant content of Copula function

In 1959, Sklar pointed out that a joint distribution can be decomposed into n marginal distributions and a Copula function, which describes the correlation between n variables. Thus the Copula function can be viewed as a function that associates the joint distribution with its n marginal distributions. In some models of life prediction, it is often assumed that the joint distribution is subject to multiple logarithms, multivariate exponential distributions or multivariate normal distributions in order to facilitate the calculation. However, these assumptions can't describe the correlation between several variables very well. It is unreasonable to use hypotheses to describe them. The Copula function can well describe the correlation between marginal distribution and joint distribution.

3.1 Definition of Copula function and related properties

N -dimensional function $C: I^n=[0,1]^n \rightarrow [0,1]=I$ can be called copula function, if it meets the followings (Yi 2011).

- (1) For $\forall u \in I^n$, as long as there is a value of 0 in the u component, there is $C(u)=0$
- (2) For $\forall i \in \{1,2,3,\dots,n\}$, $u_i \in [0,1]$, $C(1,\dots,1,u_i,1,\dots,1)=u_i$;
- (3) For $\forall u_i$, $C(u_1,\dots,u_n)$ increases;
- (4) For $\forall 0 \leq a_i \leq b_i \leq 1$, there exist $\Delta_{a_i}^{b_i} \Delta_{a_{i-1}}^{b_{i-1}} \dots \Delta_{a_1}^{b_1} C(u_1,\dots,u_n) \geq 0$, and $\Delta_{a_i}^{b_i} C(u_1,\dots,u_n) = C(u_1,\dots,u_{i-1},b_i,u_{i+1},\dots,u_n) - C(u_1,\dots,u_{i-1},a_i,u_{i+1},\dots,u_n)$

: Supposing the marginal distribution functions of random vectors (X_1, X_2, \dots, X_n) respectively are $F_1(x_1), F_2(x_2), \dots, F_n(x_n)$ and the joint distribution functions are $F(x_1, x_2, \dots, x_n)$. Then there exists a Copula function $C: F(x_1, \dots, x_n) = C(F(x_1), \dots, F(x_n))$ (Wang 2011).

Table 5 Electrochemical parameters of A steel bar

parameter	0d	90d	180d	270d	360d	450d	540d	630d	720d	810d	900d	990d
$i_{cor} / (\mu A \cdot cm^{-2})$	9.5	5.8	5.5	5.9	5.4	5.1	4.96	5.3	5.3	5.1	5.9	6.4

Table 6 Electrochemical parameters of B steel bar

parameter	0d	90d	180d	270d	360d	450d	540d	630d	720d	810d	900d	990d
$i_{cor} / (\mu A \cdot cm^{-2})$	1.12×10^{-3}	5.66×10^{-2}	4.42×10^{-2}	3.78×10^{-2}	4.06×10^{-2}	3.59×10^{-2}	4.43×10^{-2}	4.85×10^{-2}	4.05×10^{-2}	3.52×10^{-2}	4.10×10^{-2}	4.50×10^{-2}

3.2 Steps of predicting steel corrosion based on Copula function

Step 1; Firstly, the electrochemical parameters of steel bar corrosion were selected, and then the distribution of electrochemical parameters were checked. Finally, The $S(t)$ graph of each marginal distribution was calculated by Matlab.

Step 2; According to the characteristics of electrochemical parameters, the appropriate Copula function type was selected.

Step 3; The two-step estimation was used to estimate the unknown parameters of Frank-Copula function.

Step 4; The density function $c_\delta(u, v)$ and distribution function $C_\delta(u, v)$ of Frank-Copula function were calculated and plotted by Matlab.

Step 5; The $S(t)$ graph of the Frank-Copula function was plotted by Matlab and analyzed with the marginal distribution $S(t)$ graph. And finally the corrosion prediction time of the damaged coated steel bar was obtained.

4. Determination of marginal distribution function

The electrochemical parameters of magnesium cement concrete were obtained by electrochemical experiments, as shown in Table 5 and Table 6. Based on the data characteristics of the table, the P-P diagram is plotted to verify whether the data is in accordance with the specified distribution (Yang 2007).

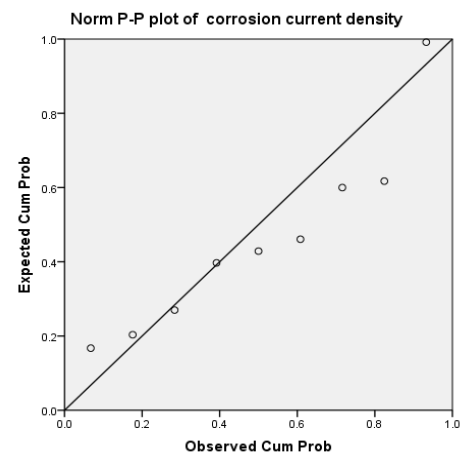


Fig. 2 Corrosion current density PP plot of steel bar A

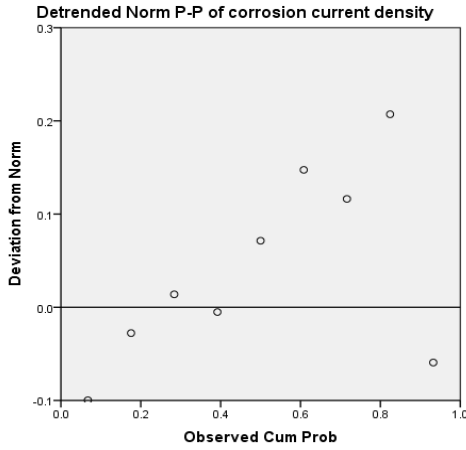


Fig. 3 Detrended corrosion current density PP plot of steel bar A

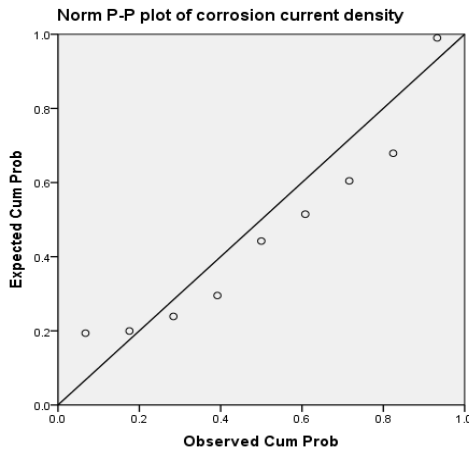


Fig. 4 Corrosion current density PP plot of steel bar B

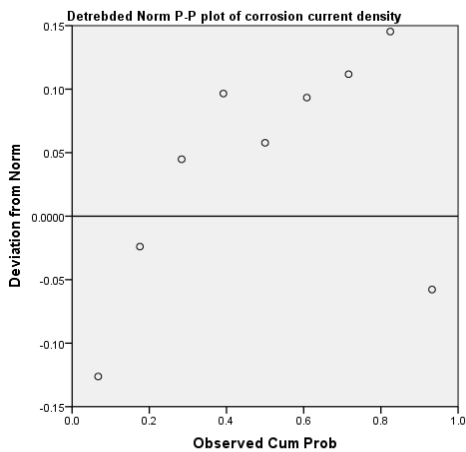


Fig. 5 Detrended corrosion current density PP plot of steel bar B

Figs. 2 to 5 are P-P plots and detrended P-P plots of steel bars in group A and B, which are drawn by their corrosion current density that acts as marginal distribution function and follows the normal distribution. It can be seen from the figure that the data points in the P-P diagram are in the right diagonal position, while it is discretely distributed in the

detrended P-P plots. Therefore, the normal distribution can be used as the marginal distribution of the single factor, moreover, it is reasonable. Its probability density function is shown below.

$$f(x) = \frac{1}{\sqrt{2\pi}\sigma} e^{-\frac{(x-\mu)^2}{2\sigma^2}}, \quad x \in (-\infty, +\infty) \quad (1)$$

As the function of steel bar corrosion in the magnesium cement concrete is complex and difficult to determine, the regression analysis of the measured corrosion current density is used to describe the corrosion of the bare steel bars and the coated steel bars. The following function is obtained.

$$y_i(t) = \theta_i + \varsigma_i t \quad (2)$$

Where the $i=1$ represents bare steel bar, $i=2$ represents coated steel bar, θ, ς is relevant parameter, t is time, and the mean value of the corrosion current density is $\hat{\mu}_i$, variance is $\hat{\sigma}_i$.

So the distribution function based on the corrosion current density is $F_i(t)$

$$F_i(t) = P(y_i(t) \leq \phi) = \frac{1}{\sqrt{2\pi}(\phi_1 - \phi_2)} \int_{\phi_1}^{\phi_2} \int_{-\infty}^{\bar{x}} e^{-\frac{x^2}{2}} dx d\phi \quad (3)$$

Where $P(y_i(t) \leq \theta)$ indicates corrosion current density that is less than the corrosion current density value of critical corrosion state. X is the generalized integral term in mathematics. ϕ represents corrosion current density and

$$\bar{x} = \frac{y_{i(t)} - \hat{\mu}_i}{\hat{\sigma}_i}.$$

When the reinforced concrete is in service, the residual life of the steel bar in concrete is 1, and as the reinforced concrete starts to work, the steel bar corrosion occurs continuously, then the remaining life of steel bar is decreased continuously. Therefore, the remaining life function is $S(t)=1-F_i(t)$ and the marginal distribution function based on corrosion current density of steel bars is

$$\begin{aligned} S(t) &= 1 - F(t) = 1 - P(y(t) \leq Y) = 1 - P(y_i(t) \leq \phi) \\ &= 1 - \frac{1}{\sqrt{2\pi}(\phi_2 - \phi_1)} \int_{\phi_1}^{\phi_2} \int_{-\infty}^{\bar{x}} e^{-\frac{x^2}{2}} dx d\phi \end{aligned} \quad (4)$$

ϕ_1 and ϕ_2 can be determined by the relationship between corrosion current density and corrosion degree of steel bars in Table 4. In this paper let $\phi_1=0.1$, $\phi_2=1$, $\phi_x \in [0.1, 1]$. According to the corrosion current density of bare steel bar in Table 5, the probability life graph of bare steel bar can be obtained and the corresponding parameters are $\hat{\mu}_1=5.134$, $\hat{\sigma}_1=1.382$, $\theta_x=0.9$.

From the picture, it can be seen that the bare steel bar is corroded at the beginning, and the corrosion becomes more and more serious as time goes on. In 1800d the life probability of bare steel bar is 0.

According to the corrosion current density of coated steel bar in Table 6, the probability life graph of coated steel bar can be obtained and the corresponding parameters are $\hat{\mu}_2=0.0392$, $\hat{\sigma}_2=0.0133$, $\theta_x=0.9$.

From Fig. 7, it can be seen that the coated steel bar is corroded at the beginning and its life curve approximates to

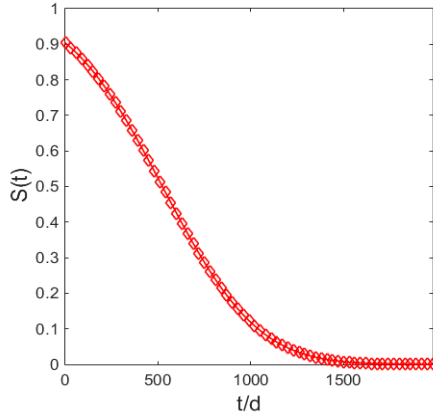


Fig. 6 Residual life prediction curve of corrosion current density of steel A

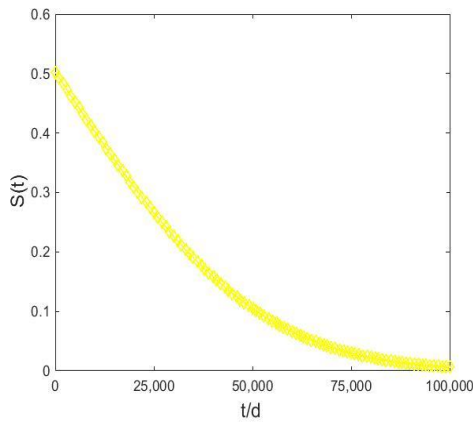


Fig. 7 Residual life prediction curve of corrosion current density of steel B

a straight line. But its decay rate is very small, its life probability is changed by 1% in 2000d. In 100000d the life probability of coated steel bar is 0.

5. Selection and correlation analysis of Copula function

5.1 Types of Copula function

The common Copula functions are the family of elliptic Copula functions and the Archimedean Copula functions. The elliptic Copula function family mainly consists of multivariate normal Copula function and multivariate t-Copula function. The Archimedean Copula function family mainly includes Clayton Copula, Gumbel Copula and Frank Copula (Hu 2013). Its function expression is as follows;

5.1.2 Tail dependence coefficient

Supposing that the random variables X and Y respectively follow the continuous distribution functions F and G , then the upper and lower tail dependence coefficients of X and Y can be defined as (Li 2012)

$$\lambda_U = \lim_{n \rightarrow 1^-} P\{Y > G^{-1}(u) | X > F^{-1}(u)\} \quad (5a)$$

Table 7 Common forms of Copula function

Function type	Function expression	Definition domain
Gumbel Copula	$C(u_1, \dots, u_n; \theta) = \exp \left\{ - \left[\sum_{i=1}^n (\ln u_i)^{\frac{1}{\theta}} \right]^{\theta} \right\}$	$\theta \in [1, \infty]$
Clayton Copula	$C(u_1, \dots, u_n; \theta) = \max \left(\left[u_1^{-\theta} + u_2^{-\theta} + \dots + u_n^{-\theta} - n + 1 \right]^{\frac{1}{\theta}}, 0 \right)$	$\theta \in [1, \infty] \setminus \{0\}$
Frank Copula	$C(u_1, \dots, u_n; \theta) = -\frac{1}{\theta} \ln \left[1 + \frac{(e^{-\theta u_1} - 1)(e^{-\theta u_2} - 1) \dots (e^{-\theta u_n} - 1)}{(e^{-\theta} - 1)^{n-1}} \right]$	$\theta \in [-\infty, \infty] \setminus \{0\}$

$$\lambda_L = \lim_{u \rightarrow 0^+} P\{Y \leq G^{-1}(u) | X \leq F^{-1}(u)\} \quad (5b)$$

If $\lambda_U \in (0, 1]$, then it is called X and Y on the upper tail correlation, if $\lambda_U = 0$, X and Y on the upper tail independence. On the contrary, if $\lambda_L \in (0, 1]$ then called X and Y on the lower tail correlation, if the $\lambda_L = 0$, X and Y on the lower tail independence. They can be also expressed as follows

$$\lambda_U = \lim_{u \rightarrow 1^-} \frac{1 - 2u + C(u, v)}{1 - u} \quad \lambda_L = \lim_{u \rightarrow 0^+} \frac{C(u, v)}{u} \quad (5c)$$

5.2 Selection of Copula function and parameter estimation

In the calculation process, the Gumbel-Copula, Clayton-Copula and Frank-Copula are used as the connecting function. However, the probability of Gumbel-Copula and Clayton-Copula's lifetime prediction probability curve may be greater than one or less than zero. It can be seen from Fig. 8 and Fig. 9 that the fore-aft symmetry of joint frequencies and frequency histograms of steel bars in A and B are not obvious, but it is in line with the characteristics of Frank-Copula function, so the Frank-Copula function can be used as the joint function. Its parameters are estimated as follows;

The Log Likelihood Function of Copula Function

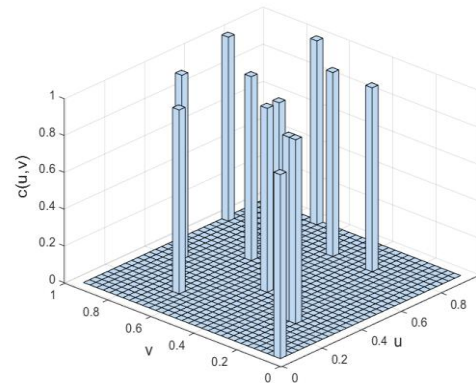


Fig. 8 Binary frequency histogram

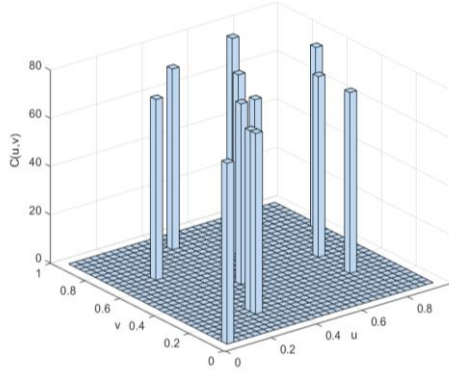
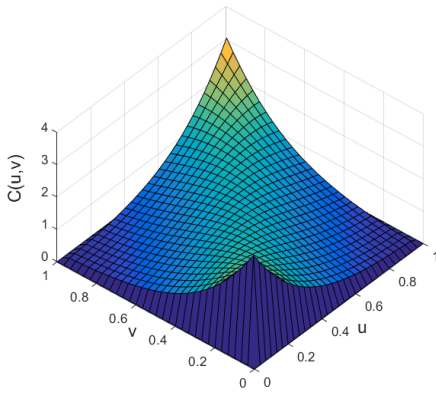
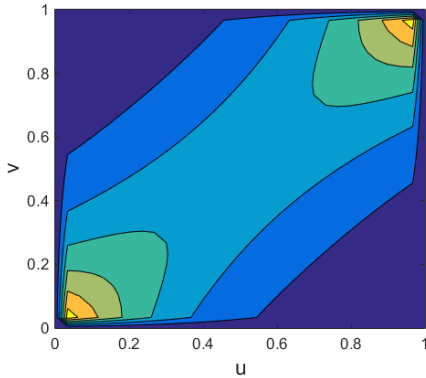


Fig. 9 Binary rate histogram



(a) Density function graph



(b) Contour map

Fig. 10 Frank-Copula function density function graph and contour map

$$l(\theta) = \sum_{i=1}^T \ln(c(F_1(x_1, \theta_1), F_2(x_2, \theta_2); \alpha)) + \sum_{i=1}^T \sum_{j=1}^2 \ln f_j(x_i; \theta_j) \quad (6)$$

After calculated

$$\hat{\alpha} = \arg \max \sum_{i=1}^T \ln(c(F_1(x_1, \hat{\theta}_1), F_2(x_2, \hat{\theta}_2); \alpha)) \quad (7)$$

5.3 Correlation analysis

The density function graph and distribution function graph of Frank-Copula function can be obtained after

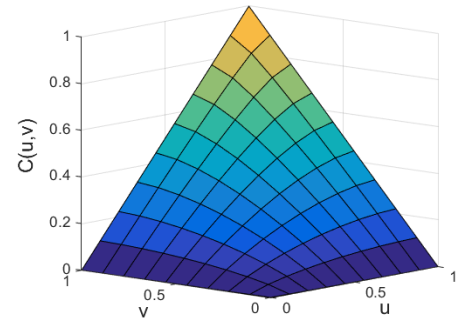


Fig. 11 Frank-Copula function distribution function graph

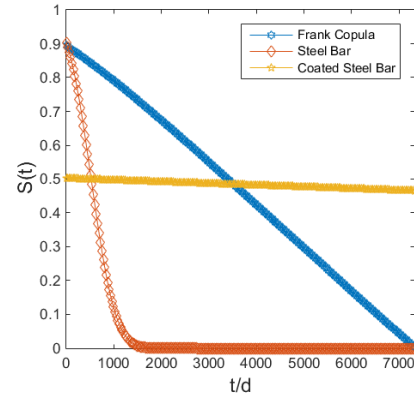


Fig.12 Different life prediction curve

analysis and calculation, as shown in Fig. 10, Fig. 11.

As the upper and lower tail correlation coefficient $\lambda_{C_F}^{up}$, $\lambda_{C_F}^{lo}$ of Frank-Copula function are equal to zero (She 2012), it indicates that variables in the Frank-Copula distribution tail are independent. That is to say, when one observing variable reaches its extremum, the probability of the other reaching its extremum is zero. It can be seen from the figure that the density function of the Frank-Copula function is “U”-shaped, symmetrical, and can not capture the asymmetric relationship between the variables.

It can be seen from Fig. 12 that the corrosion of the damaged coated steel bar and the bare steel bar begins almost simultaneously, but the corrosion rate of damaged coated steel bar is much less than that of the bare steel bar. At about 7320d, the life of the damaged steel bars is zero. The main reason is that the natural corrosion potential of Zn is -0.762 V, the natural corrosion potential of Fe is -0.440 V, and zinc acts as the sacrificial anode to provide cathodic protection for the substrate. When the coating is partially damaged, the exposed substrate and zinc will form the primary battery and zinc will become anode corrosion, whose product, hydroxide, will be attached to the damaged place; then the incompatible zinc carbonate or basic zinc carbonate will formed by reacting with carbon dioxide, providing protection for the substrate to restore the barrier.

Therefore, it can be concluded that the Frank-Copula function can well predict the remaining life of the damaged coated steel bar, and the prediction is more suitable for engineering practice, which provides a new method for predicting the life of coated steel bar.

6. Conclusions

- The corrosion current density, tested by electrochemical workstation, can be regarded as a single degradation factor to establish the marginal distribution function.
- Frank-Copula function can be more reasonable to predict the corrosion of the damaged steel bars.
- The Copula function provides a good life prediction model for the prediction of steel corrosion under the influence of multidimensional factors in the future, having a good applicability.

Acknowledgements

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