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Transreac: a model for the calculation of combined chemical reactions and transport processes and its extension to a probabilistic model

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Abstract

Transreac is a computer model for the simulation of the chemical attack to which mineral building materials are exposed. For reliability analysis, Transreac is used in conjunction with the Monte Carlo simulation method. The probabilistic model was evaluated for cement mortar corroded as a result of sulfate attack. The experimentally derived corrosion effects and their scatter are in good agreement with simulation results. In addition, the probability of failure was predicted and a sensitivity analysis was performed.

Keywords: Modelling; Monte Carlo simulation; Durability; Sulfate attack

1. Introduction

Modern building codes make use of a reliability concept based on a probabilistic approach. These approaches increasingly expand into durability design [1–5]. For example, Corr et al. [2,6,7] presented an empirical reliability analysis for sulfate attack on concrete.

A deterministic model (Transreac) to simulate corrosion effects in building materials caused by the attack of chemical solutions was developed by Schmidt-Döhl [8–10]. This model combines the calculation of chemical reactions with the calculation of transport processes within the structure. The calculation of chemical reactions is based on a repeated determination of the stable phase assemblage (Gibbs energy minimization, Pitzer model, consideration of chemical kinetics). The transport module covers a large number of different transport processes in porous materials. In addition, modules for the calculation of corrosion effects were implemented. These corrosion effects are the expansion caused by the formation of ettringite or the loss of material strength and the loss of mass caused by phase dissolution. Additional improvement has been made since the papers [8–10] were published. The accuracy and speed of the thermodynamic algorithm has been increased, and the simulation of redox-reactions has been incorporated. All transport processes can be calculated by means of 2-D processes. The heat and moisture transport can be calculated on the basis of the Künzel model [11]. It is also possible to calculate flow as a result of seepage pressure, and the model for transport processes caused by the diffusion potential has been improved.

With this highly flexible model many different simulations were performed in the past. The materials examined include cement mortar, concrete (also with fly ash and blast furnace slag), sandstone and Sorel cement. The aggressive solutions include sulfate solutions, chloride solutions, acid, seepage water from a landfill, saline solutions characteristic for salt mines, and solutions with different amounts of aggressive carbon dioxide (see for example Refs. [8,10,12]).

Transreac has been extended to a probabilistic model. For this purpose, the deterministic model has been incorporated into a Monte Carlo simulation. The Monte Carlo method is a

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stochastic method, which means that it is based on the use of random numbers and probability statistics to investigate problems (see Ref. [13]). This numerical approach was used to maintain all features and the full flexibility of the deterministic model.

One objective of this study is to compare the experimentally derived and simulated scatter of corrosion effects. For this purpose, an experiment that considered expansion caused by sulfate attack was used. The second intention of this study is to investigate the reliability of structures made from mortar used in the experiment mentioned. Here, calculation of failure probability and some sensitivity factors are demonstrated. These parameters are one of the results of a Monte Carlo simulation.

2. Methods

2.1. Implementation of Transreac in a Monte Carlo simulation

The implementation of Transreac in a Monte Carlo simulation is in principle a multiple calling of the main program of Transreac (see Fig. 1). With every calling, the initial values are changed corresponding to their experimentally derived pattern of distribution. Then the results of the Transreac algorithm produced for changed initial values are recorded. A random generator produces the changed input parameter. The number of callings (referred to as "trials" below) has an effect on the system response. This must be examined, following which a suitable number of trials must be determined (see Section 5.2). At the end there is a statistic analysis of the response. The program offers the possibility to split the calculation on different computers.

2.2. Corrosion experiment

The validation of the probabilistic model will be demonstrated by a corrosion experiment. The experiment is described in Refs. [8,10]. Mortar specimens with a length of 100 mm were in contact with sodium sulfate solution (44 g/L). Then the formation of ettringite and the induced corrosion effects were investigated. During the first phase of the experiments, capillary suction was the main transport process. After saturation of the specimens, the diffusion of dissolved species was the only transport process. The experimental input parameter for Transreac (mean value and standard deviation) are listed in Ref. [8]. All these parameters were derived from non-corroded material. Due to the lack of experimental data it is assumed that the initial values have a normal distribution.

2.3. Validation of the probabilistic model

In this section, the experimentally derived and the simulated results were compared, with a special focus on the formation of ettringite and the expansion of the specimens. Fig. 2 shows the ettringite profile of the mortar after 154 and 303 days of contact with the sulfate solution. The measured profiles show an additional formation of ettringite caused by the attack of the sulfate solution. After 303 days it was found that gypsum had also formed on the corroded side of the specimen.

The calculated results are consistent with the experimental data (see Fig. 2). The experimentally observed formation of gypsum was also simulated by Transreac. The error bars are the result of the Monte Carlo simulation. They show the standard deviation of the simulated ettringite profile. The Monte Carlo simulation shows an increased standard deviation beginning at 75 mm from the non-corroded end

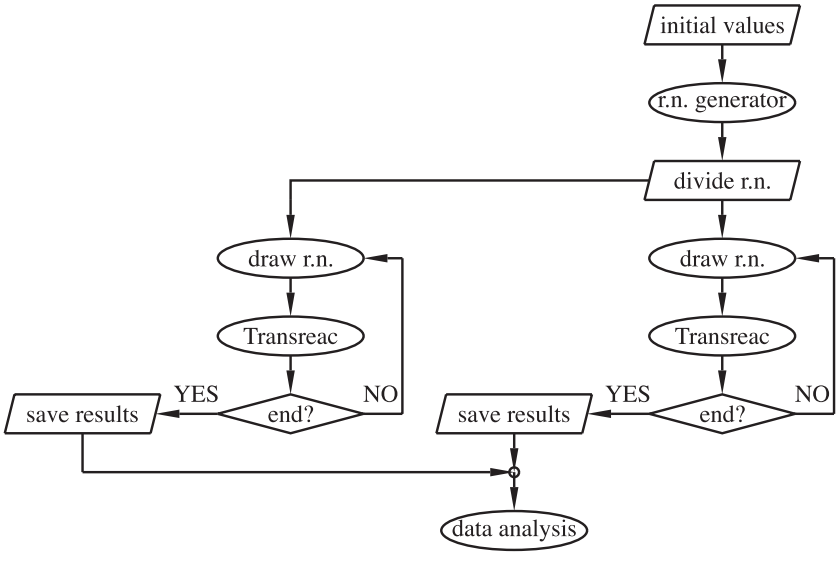

Fig. 1. Structure of the probabilistic algorithm, split for two computers (r.n.=random number).

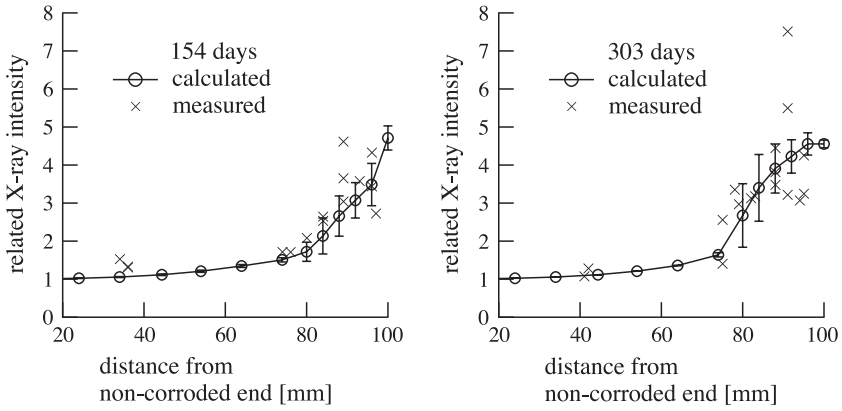

Fig. 2. Ettringite profile of corroded specimen after 154 days (left) and after 303 days (right) of contact with Na_2SO_4-solution (see 12). The X-ray intensity of ettringite is related to the X-ray intensity of the non-corroded material.

of the specimen. This can also be found in the experimental results. The calculated maximum coefficients of variation are 22% (154 days) and 31% (303 days). The experimental measurements show a maximum variance of 24% (154 days) and 40% (303 days). Most of the experimentally derived scatter of the ettringite concentration can be predicted by the Monte Carlo simulation. The simulated scatter at 90 mm from the non-corroded end of the specimen is too small.

The evaluation of the expansion caused by the formation of ettringite is shown in Fig. 3. The experimentally derived data and their scatter match the simulated data.

3. Reliability analysis

3.1. Sensitivity of initial values

Sensitivity factors provide information on the importance a number of initial values have on the simulated corrosion effect. To make a rough estimate, the ettringite concentration was calculated for changed initial parameters that were changed one by one. Each parameter is varied between $\mu-\sigma$ and $\mu+\sigma$ (where μ: mean value and σ: standard deviation). Examples are shown in Fig. 4. It can be seen that with greater total open porosity the amount of newly formed ettringite is increased. This is expected, because the transport of corrosive solution increases with increasing porosity. Another example shows the influence of a water vapour diffusion coefficient on the ettringite concentration. It is found that the ettringite concentration is not influenced. The water vapour diffusion coefficient is not very important for the corrosion experiment presented here, because after a short time the specimens are saturated with water.

In what follows, the sensitivity factors of the total open porosity, the porosity derived by mercury intrusion porosimetry (MIP), the bulk density, the water absorption coefficient, the ultimate tensile strain and the diffusion coefficient of SO_4^{2-}, Na^+, OH^- are examined. The calculation method is described in Section 5.3. The ultimate tensile strain is used to indicate the onset of cracks, with an additional diffusion process trough the cracks. Fig. 5 shows the normalized sensitivity factors relating to the overall linear strain. This diagram illustrates the significance the input parameters have on the predicted linear strain. The greatest influence has to be attributed to parameters that are needed for modifying the transport parameter during the corrosion process. As corrosion proceeds, the diffusion coefficients and the water absorption coefficient are changed. This is considered in Transreac (see Ref. [9]).

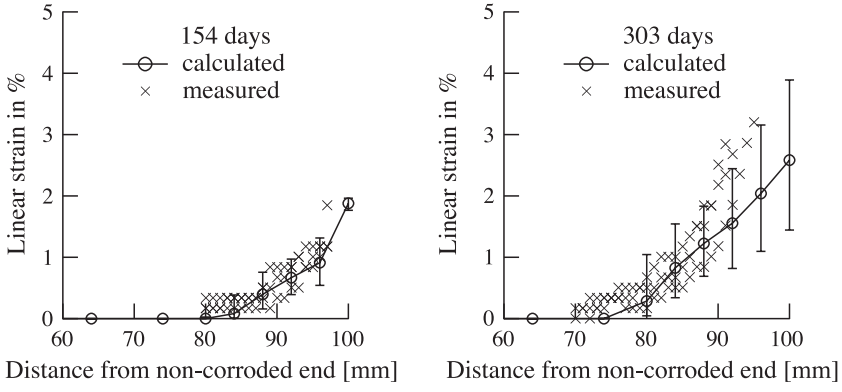

Fig. 3. Expansion of corroded specimen after 154 (left) and 303 (right) days of contact with Na_2SO_4-solution (see 12).

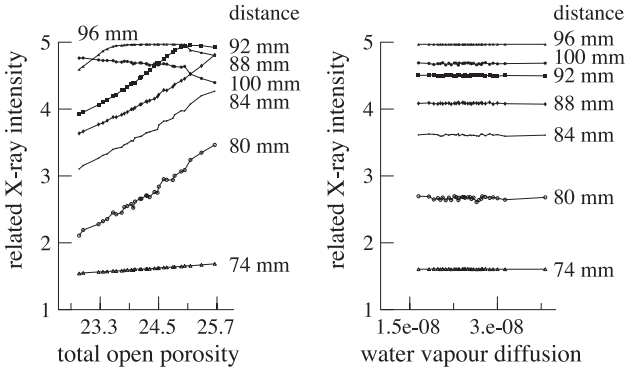

Fig. 4. Influence of total open porosity (left) and water vapour diffusion coefficient (80% r.h.) (right) on ettringite formation. The X-ray intensity of ettringite is related to the X-ray intensity of the non-corroded material. Distance means the distance from the non-corroded end of the specimen.

For modification, the following parameters are required: the total open porosity, the porosity derived by MIP, the density and the water absorption coefficient. The ultimate tensile strain shows little influence on the simulated linear strain. This observation can be used as a design basis for new corrosion experiments. It is important to carefully derive the experimental values for the total open porosity, the porosity derived by MIP, the density, and the water absorption coefficient. Experimental examination of the ultimate tensile strain and the diffusion coefficient may be estimated roughly. Fortunately, examination of porosity, density and absorption coefficient is a much less time consuming process than diffusion coefficient derivation.

3.2. Limit state and probability of failure

Structures must be reliable with respect to their life time. In the mathematical sense, reliability is the probability of the structures to resist environmental influences. The probability of failure, which is the complementary value to the probability of reliability, is the value that is commonly used. The acceptability of low residual risk and low probability of failure allow economic aspects to be considered as part of building design.

The eurocode 1 (EC 1) defines maximum allowable failure probabilities for different limit states under consideration of different economic aspects and different consequences of a failure. A distinction is drawn between two limit states: serviceability limit state and ultimate limit state.

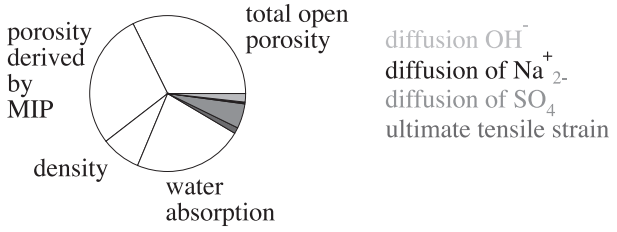

Fig. 5. Normalized sensitivity factor of input parameter from non-corroded material.

The ultimate limit state is associated with the collapse of a building. The serviceability limit state corresponds to the capacity of a building to perform the service function for which it is designed and used. Because we do not focused focus on a special building, assembly, component or structure we considered the serviceability limit state as an example. For this kind of limit state a maximum allowable failure probability of 2.275×10^{-2} is suggested if noticeable consequences of a durability failure are expected. Here, high costs for manufacturing are present, but only low costs occur to maintain the reliability of the building (safety class I). The safety class I is chosen because the aggressive solution in the following example is highly concentrated.

The probability of failure of the mortar described in Refs. [8,10] was examined as an example. For calculation of the probability of failure, see Section 5.4. As a failure criterion, 0.5% expansion was used, which is defined by U.S. Bureau of Reclamation (USBR) [2,6,7]. During the long-term testing program of the USBR, the corrosion induced by sulfate in several concrete mixtures was examined. The specimens were submerged in 2.1% Na_2SO_4 solution at room temperature for up to 40 years. These experimental conditions were used to simulate corrosion effects by means of Transreac. Fig. 6 shows the simulated linear strain induced by the formation of ettringite as measured for the USBR specimens. Error bars indicate the results of the Monte Carlo simulation. They show a maximum coefficient of variation of 36%. The limit state of 0.5% strain is already exceeded after 10 years. An empirical model for the sulfate-induced expansion is described in Ref. [6]. It is based on the results of the long-term testing program of USBR. Unfortunately, this model does not include mixtures with a C_3A content of 9% and w/c ratios of 0.6, which are the mixture properties that are regarded here. One model relates to the expansion of cements with low C_3A (<8%) content and the other model deals with cements with high C_3A (>10%) content. Fig. 6 shows both models that are calculated for the w/c ratio and the C_3A content mentioned above. As expected, the results of the simulation of Transreac were between the above models. Fig. 6 shows the probability of failure versus the exposure time. 2.275×10^{-2} (see Ref. [14]) for the maximum allowable

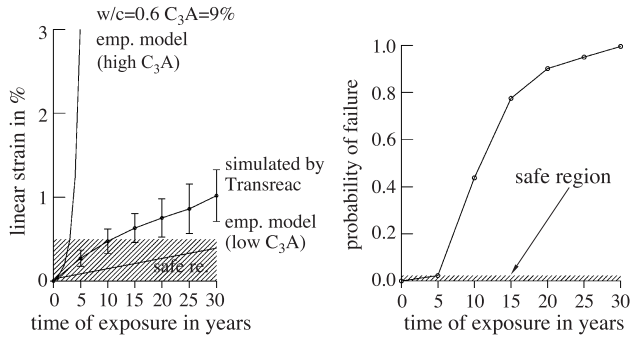

Fig. 6. Linear strain (left) and probability of failure (right) as a function of time of exposure.

probability of failure is marked in Fig. 6. The results of the Monte Carlo simulation show that the specimens are in the safe region for up to 5 years. This is in agreement with practical guidelines according to which type of cement is not suitable for the production of concrete or mortar with high durability against sulfate attack.

4. Conclusions

The transport reaction model Transreac was extended to a probabilistic model. To predict the scatter of corrosion effects, the Monte Carlo simulation method is used in conjunction with Transreac. The probabilistic model is validated by means of corrosion experiments. Here, an experiment that deals with a sulfate solution attack on mortar is used. Comparison of the corrosion experiments and the computer simulation shows that the chemical reactions were predicted correctly. The changes of the phase assemblage and the damage were calculated quantitatively. In addition, the scatter of the experimentally derived data can be calculated by means of the Monte Carlo simulation. The experiment mentioned is used to present the determination of sensitivity factors and the calculation of the probability of failure. The results produced are consistent with practical experience made for concrete durability against sulfate attack. These results show that it is possible to use Transreac in conjunction with a Monte Carlo simulation in connection with a reliability concept for durability.

5. Details of the probabilistic simulations

5.1. Production of random number

For the production of random numbers, the Latin Hypercube sampling rule was applied. This variance reduction method allowed the necessary number of trials to be reduced (see Ref. [13]). For this procedure, 50 trials are enough to obtain an error of less than 0.01% between the estimated and the calculated standard deviation or mean value. The appropriate number of trials must be a minimum of 50 trials (see Section 5.2).

5.2. Appropriate number of trials

The appropriate number of trials for the Monte Carlo simulation depends on the necessary precision of the result of the simulation, the necessary precision by generation of random number (see Section 5.1) and on the computing time. To predict a suitable number of trials, the influence on the response of the Transreac calculation was examined. The number of trials is expected to be between 50 (see Section 5.1) and 1000. Up to a distance of 64 mm from the non-corroded end of the specimen there is no significant influence on the mean value or on the 95% confidence interval of the mean value. Fig. 7 shows the dependence of the mean value of ettringite concentration on the number of trials. For clarity's sake, only the results for distances between 74 and 100 mm from the non-corroded end of the specimen are shown. The error bar shows the 95% confidence intervals for the mean value of the ettringite concentration. It is found that the confidence interval narrows with an increasing number of trials. For trial numbers between 250 and 1000, the mean values do not show any dependence on the number of trials. Fig. 7, in addition, shows the experimental error produced when examining the ettringite concentration determined by XRD. It can be seen that the experimental error is greater than the error produced when reducing the number of trials to 250. In Fig. 7, the influence of the linear strain on the number of trials is also shown. However, as described for the ettringite concentration, the experimental error is greater than the error that results from a reduction of the number of trials to 250 trials.

For the validation of the probabilistic model an experiment with a duration time of 303 days is used (see Section

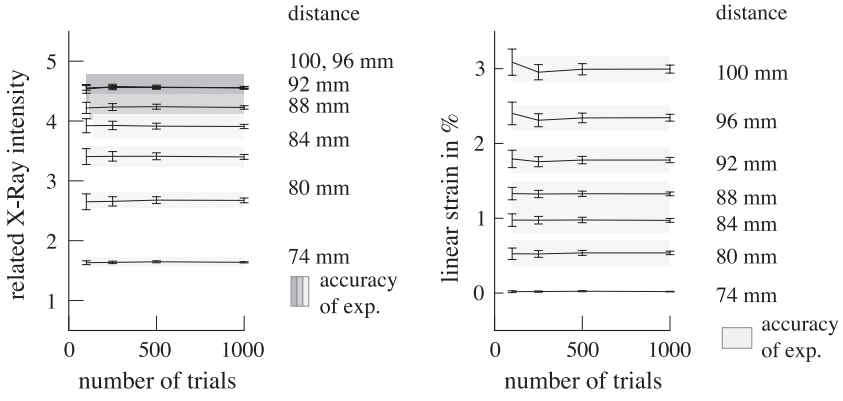

Fig. 7. Mean value, 95% confidence interval (error bar), and experimental accuracy (shaded) plotted against number of trials, shown for ettringite concentration (left), and linear strain (right). The X-ray intensity of ettringite is related to the X-ray intensity of the non-corroded material. Distance means the distance from the non-corroded end of the specimen.

2.2). The calculation time for one of these experiments is about 10 min. Since the Monte Carlo simulation can be split up on different computers, 1000 trials are used to validate the model. To estimate the sensitivity of one input parameter on the formation of ettringite and the expansion (see Section 3.1) only 250 trials were made for the Monte Carlo simulations, which means that for this routine a total of 4250 trials is needed (see Section 5.3). The prediction of the probability of failure (see Section 3.2) is simulated for 30 years. Because of the increased calculation time, 250 trials for the Monte Carlo simulation were used in this case.

5.3. Calculation of sensitivity factor

The sensitivity factor (on an average) is calculated by means of the following equation:

$$e_i = \frac{\frac{\delta G(\bar{x}_1,\ldots,\bar{x}_{i-1},\bar{x}_i,\cdots,\bar{x}_N)}{\delta(x_i)}}{\bar{G}}\bar{x}_i \quad (1)$$

where $\bar{x}_i$ is the mean value of a basic variable i, and $\bar{G}$ is the limit state equation. The derivation of $G\left(\frac{\delta G(\bar{x}_1,\ldots,\bar{x}_{i-1},\bar{x}_i,\cdots,\bar{x}_N)}{\delta(x_i)}\right)$ was determinated by linear interpolation to three points (P1, P2, P3), each spaced with standard deviation σ:

P1 $\bar{x}_1, \ldots, \bar{x}_{i-1}, \bar{x}_i, \ldots, \bar{x}_N$
P2 $\bar{x}_1, \ldots, \bar{x}_{i-1}, \bar{x}_i+\sigma_i, \ldots, \bar{x}_N$
P3 $\bar{x}_1,\ldots, \bar{x}_{i-1}, \bar{x}_i-\sigma_i,\ldots, \bar{x}_N$

For every interpolation point, at least 250 trials are necessary (altogether 4250 trials for eight parameters, see Section 5.2).

5.4. Calculation of probability of failure

The probability of failure P_f is defined by means of the following equation:

$$P_f = \int_{\{x|g(\rightarrow x)<0\}} f_x(\vec{x})\mathrm{d}\vec{x} = F(g(\vec{x}) = 0) \quad (2)$$

According to the theory of reliability, the probability of failure contains the area where the limit state is exceeded. This is equal to the function value of the cumulative distribution function F at the limit state. Usually, the limit state is characterized by a limit state equation less than zero ($g(\rightarrow x)<0$). The probability of density f_x is a function of different basic variables x_i, e.g. transport parameters, porosity, etc. One possibility to solve the integral of Eq. (2) is the Monte Carlo method, which is used here. With every trial it must be checked whether the system response is within the failure area. Thus, the failure probability is calculated from the number of trials in the failure area n_f, and the overall number of trials n by Eq. (3).

$$P_f = \frac{n_f}{n} \quad (3)$$

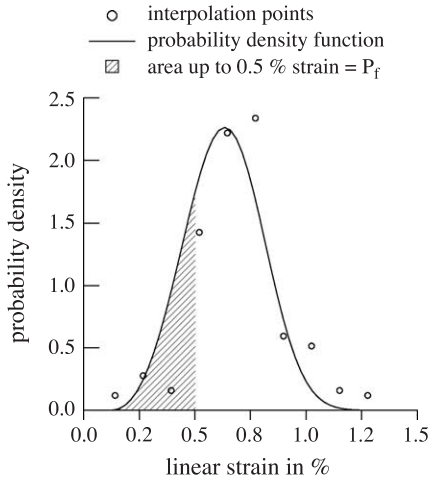

Fig. 8. Definition of probability of failure for 0.5% strain as limit state.

Generally, the failure probability is very low (10^{-4}). To calculate such a low failure probability, 100×10^4 Monte Carlo simulation trials are necessary as a minimum [15]. To reduce the number of trials, the following procedure is applied, which is suggested in Ref. [15]. One result of a Monte Carlo simulation is a histogram, which shows the distribution of the limit state equation. The histogram is approximated by fitting a known probability density function to the data (see Fig. 8). The probability of failure is then calculated by using the known or numerically derived cumulative density function of the selected probability density function (see Eq. (2)). For this purpose, the program dataplot is used [16]. The number of trials is thus reduced to 250 (see Section 5.2).

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