FLUID MECHANICS AND TRANSPORT PHENOMENA

Theoretical and Experimental Study of Vapor Deposition onto a Dressed Body Part

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When air-permeable clothing is worn, vapor will penetrate to the inside of the clothing. The vapor can deposit onto the skin, thus, forming a potential health threat. In a previous article a model was presented, which describes the airflow around body parts, covered with clothing. This airflow profile is used to calculate the vapor deposition onto the skin. A test setup was developed to validate the deposition model. Cylinders are used as a representative for human body parts. They are covered by a layer of protective clothing and exposed to vapor of methylsalicylate. The amount of vapor which deposites onto the surface of the cylinder is determined by using charcoal cloth. The influence of the clothing air permeability, the wind speed, the diameter of the cylinder, and the distance between clothing and cylinder surface was investigated. The experimental results show reasonable to good agreement with the model. *AIChE J,* 54: 844–849, 2008
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Introduction

Air permeable clothing with a layer of carbon is used to protect persons against toxic agents, especially chemical warfare agents. The basis of the protection is adsorption of the toxic agent onto the carbon. The use of air permeable materials in the clothing reduces the thermal load offered by the clothing, by allowing air to flow through the material. At the same time the air flow can transport the toxic agents to the inside of the clothing, if the agent is not fully adsorbed by the carbon.

The purpose of the study described here is to use this airflow profile as a base to investigate the vapor transport through the clothing material and the resulting mass transfer of the chemical agent to the skin. A one-dimensional (1-D) model, describing the concentration profile, is proposed here, together with experiments used for validation of the model. In this study a body part is represented by a cylinder. This cylinder is dressed with clothing at a constant distance from the surface, and is put in a fixed position with respect to the wind direction (Figure 1). Wind flows around the body part, through the clothing, underneath it and at the backside out again. In a previous article a 1-D theoretical description of the wind-flow profile around a body part, covered with air permeable clothing, was presented.[1]

Parameters like the air velocity profile, the concentration profile, and the temperature profile were analyzed earlier by using computational fluid dynamics (CFD).[2–4] Since these calculations are quite time-consuming, simplified models have been derived and are presented here, which do not need CFD.

The mechanism of skin exposure of a dressed person to vapor consists of the following sequence of steps:

- penetration of the chemical agent through the clothing material;
- mass transfer across the air gap separating the protective clothing from the skin;
- adsorption *onto* the skin;
- absorption of the agent *into* the skin.

The first three steps have been described in the theoretical model presented here. The model has been experimentally validated by measuring the mass deposition onto the skin, since that is the endpoint of the deposition process. Intermediate steps like the air velocities through and under the clothing have been studied earlier.[1]

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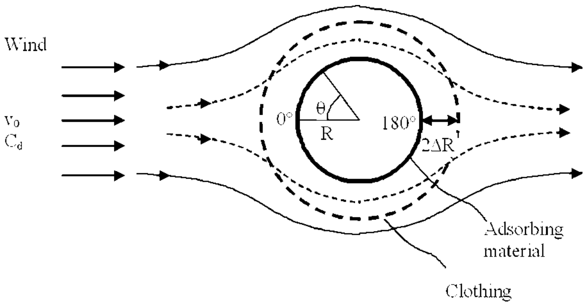

Figure 1. Representation of a body part, dressed with clothing, with airflow around and underneath the clothing.

Theory

If the airflow profile around, through and underneath the clothing is known, a concentration profile can be derived. To model the concentration profile underneath the clothing, and the local mass deposition on the cylindrical surface, it was assumed that:

- all the mass of the chemical agent which reaches the cylinder surface is adsorbed;
- the mass transport to the surface is described by the penetration theory[5,6];
- the clothing does not adsorb any agent.

The first assumption implies that the concentration on the surface of the cylinder is always zero. The third assumption is done to simplify the current study. Previously, a model was proposed, which predicts the amount of agent which can be adsorbed by NBC-protective clothing.[7] However, since the deposition process itself is not influenced by this adsorption process, this aspect needs not to be taken into account here.

A continuity equation (a mass balance for a slice of air underneath the clothing) describes the vapor concentration underneath the clothing as a function of the angle θ with respect to the wind direction around the cylinder (Figure 1). The airflow profile is used as input for the deposition model. A representation of the mass balance is shown in Figure 2.

The continuity equation will contain both convection and diffusion terms, and is described by a partial differential equation (Eq. 1)

$$\frac{dC}{dt} = D\frac{d^2C}{dx^2} - v_{\text{air}}\frac{dC}{dx} + \frac{v_{\text{mat}}}{2\Delta R}(C_{\text{d}} - C) + \frac{\varepsilon D}{2\Delta R d_{\text{mat}}}(C_{\text{d}} - C) - \frac{k_{\text{g}}}{2\Delta R}C \quad (1)$$

Equation 1 describes the concentration between two parallel plates. This relation can be modified for the flow of air between clothing and a cylinder by assuming that the radius of the cylinder is large in comparison to the size of the air gap. In that case an approximation can be used for dx

$$dx \approx R d\theta \quad (2)$$

Equation 1 then changes into Eq. 3

$$\frac{dC}{dt} = \frac{D}{R^2}\frac{d^2C}{d\theta^2} - \frac{v_{\text{a}}}{R}\frac{dC}{d\theta} + \frac{v_{\text{mat}}}{2\Delta R}(C_{\text{d}} - C) + \frac{\varepsilon D}{2\Delta R d_{\text{mat}}}(C_{\text{d}} - C) - \frac{k_{\text{g}}}{2\Delta R}C \quad (3)$$

The first term at the right represents the diffusion underneath the clothing, the second term is the convection underneath the clothing, the third is the convection through the clothing, the fourth is the diffusion through the clothing, and the fifth is the deposition onto the surface of the cylinder. Equation 3 can be rearranged into Eq. 4

$$\frac{dC}{dt} = \frac{D}{R^2}\frac{d^2C}{d\theta^2} - \frac{v_{\text{a}}}{R}\frac{dC}{d\theta} - \left(\frac{v_{\text{mat}}}{2\Delta R} + \frac{\varepsilon D}{2\Delta R d_{\text{mat}}} + \frac{k_{\text{g}}}{2\Delta R}\right)C + \left(\frac{v_{\text{mat}}}{2\Delta R} + \frac{\varepsilon D}{2\Delta R d_{\text{mat}}}\right)C_d \quad (4)$$

The boundary conditions of this equation are given in Eq. 5

$$\left.\frac{dC}{d\theta}\right|_{\theta=0} = 0 \qquad \left.\frac{dC}{d\theta}\right|_{\theta=\pi} = 0 \qquad C|_{t=0} = 0 \qquad C|_{t=\infty} = C_d \quad (5)$$

A boundary layer will be created near the surface in the air. The concentration profile in this boundary layer is supposed to be linear.

Usually the thickness of the boundary layer δ, is unknown. It is customary to combine δ with the diffusion coefficient D to calculate k_{g}, Eq. 6[5]

$$k_{\text{g}} = \frac{D}{\delta} \quad (6)$$

The mass-transfer coefficient k_{g}, is unknown too. The thickness δ is a function of the air velocity underneath the clothing, the covered distance along the cylinder surface x, and the thickness of the air layer between the clothing and the cylinder surface $2\Delta R$. Thus, the thickness of the air gap determines the maximum boundary thickness. It is customary to write these dependencies in the dimensionless numbers as

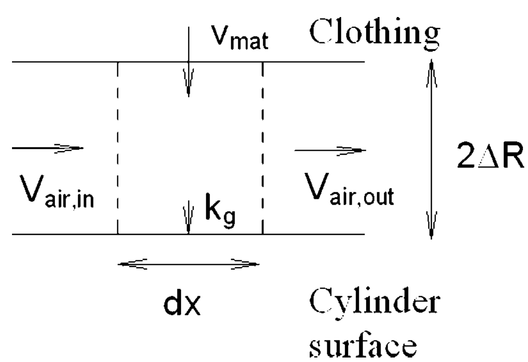

Figure 2. Volume element of air between the clothing and the surface of the cylinder.

in *Sh* (Sherwood), *Re* (Reynolds), and *Sc* (Schmidt). For different situations, different relationships between these parameters are found. Several different relationships were tried (for instance, the penetration theory,[6] and an empirical relationship for parabolic velocity profiles in tubes[5]). Although the difference in calculated values for the mass-transfer coefficient was not much, it was decided to use the empirical relationship for parabolic velocity profiles, since it was shown by Sobera et al.[8] that the tangential velocity profile in the air gap has a parabolic shape. The used relationship for parabolic profiles is[5]

$$Sh = 1.08\ \mathrm{Re}^{\frac{1}{3}} Sc^{\frac{1}{3}} \left(\frac{2\Delta R}{x}\right)^{\frac{1}{3}} \tag{7}$$

or

$$Sh = 1.08\ \mathrm{Re}^{\frac{1}{3}} Sc^{\frac{1}{3}} \left(\frac{2\Delta R}{R\theta}\right)^{\frac{1}{3}} \tag{8}$$

with

$$Sh = \frac{2k_g \Delta R}{D} \qquad \mathrm{Re} = \frac{2\rho\ v_{\mathrm{air}} \Delta R}{\eta} \qquad Sc = \frac{\eta}{\rho D} \tag{9}$$

The mass-transfer coefficient changes, because the thickness of the boundary layer changes as a function of the distance covered by the air flow. This thickness δ, will increase until it reaches the size of the air gap underneath the clothing, after which it becomes constant. Effectively this means that the Sherwood number will not become smaller than 1. Thus, for large distances

$$\mathrm{Sh} = 1 \tag{10}$$

The relations 8 and 10 give the mass-transfer coefficient (the Sherwood number) as a function of the air velocity underneath the clothing (the Reynolds number). These equations, together with the flow profile model as described earlier,[1] can be used to calculate the concentration profile underneath the clothing. From this concentration profile, both a dosage and a deposition profile (the total amount of deposited mass per unit area) can be derived (Eqs. 11 and 12)

$$Dosage = Ct = \int_0^t C dt \tag{11}$$

$$Deposition = M_{\mathrm{d}} = \int_0^t k_g C dt \tag{12}$$

The vapor deposition process is quantitatively expressed by the deposition velocity v_d, which is the velocity wherewith the vapor molecules are transferred to the skin (Eq. 13). It should be noted that this is a mass-transfer velocity and not an air velocity.

$$v_{\mathrm{d}} = \frac{M_{\mathrm{d}}}{Ct} \tag{13}$$

where v_d represents the vapor deposition velocity M_d, the mass deposited per unit area and *Ct* the dosage of exposure of the chemical agent.

If the mass-transfer coefficient k_g, is independent of time, it will be equal to the deposition velocity v_d. This deposition velocity can be measured as a function of the angle around the cylinder, and can be compared with the theoretical mass-transfer coefficient for validation.

Experiments

Figure 1 gives an overview of the test setup, shown from the top. A body part is represented by a stainless steel cylinder (outer diam. 10 cm, length 75 cm).

The test cylinder was placed in a 19 m^3 test chamber in which the air was circulated at an average linear velocity of 1.8 ± 0.1 m/s or 5.1 ± 0.4 m/s over the full length of the cylinder, measured in separate experiments with an anemometer (TSI, model 8455-300). The cylinder was exposed to methylsalicylate vapor, as a simulant for mustard gas. The methylsalicylate concentration in the test chamber was continuously monitored by GC-FID (Chrompack, CP 9001). The average concentration was 77 ± 4 mg m^{-3}, and the duration of a test run was 45 min, giving an average dosage *Ct* around 3,500 mg min m^{-3}. Temperatures and relative humidities were measured with a thermohygrometer (Hanna Instruments HI 9161C). Temperatures varied between 29 and 37°C. The relative humidity was below 40%.

Two types of clothing material were used in the experiments, material A with a high-air permeability, and material B with a low-air permeability. Air flow resistances were determined by an inhouse method using a Thommen HM 28 manometer (accuracy <0.5 Pa). Both clothing types were outer layers of air permeable NBC protective suits. These outer layers are responsible for the major part of the air permeability of the material. The adsorptive carbon layers of the suits were not used. Therefore, it was assumed that the dosage inside the clothing was the same as the dosage outside the clothing, which made it possible to use a relatively convenient test setup: the concentration under the clothing materials could be constant and high. In preliminary experiments it was found that the vapor deposition velocity did not depend on the dosage of exposure for dosages up to at least 20,000 mg min/m^3.

The two clothing materials A and B were tested simultaneously. By using four spacer rings, two 10 cm bands of clothing material were wrapped around the cylinder. The bands were 8 cm apart. Due to a difference in stiffness material A was positioned at an average distance of 32 mm from the cylinder surface, and material B at an average distance of 36 mm (accuracy around 1 mm).

The cylinder surface was covered by an adsorbing material, carbon containing filter paper (Schleicher and Schuell GmbH, 508), which was used to analyze the mass deposition onto the surface. After the vapor exposure 18 mm diam. circular samples were taken at different angles from the stagnation point, the point at which the wind collides perpendicularly with the cylinder (angle = 0°). The mass deposition M_d, was determined by extraction of these samples with carbon disulfide, followed by GC-FID analysis. The extraction efficiency has been determined separately to be 94 ± 3% for mass depositions between 76 and 4,645 mg/m^2. The measured

Table 1. Observed Experimental Conditions

	Material		
Parameter	A	B	Unit
Γ	5.66	0.265	mm/(Pa s)
vw	5.1	1.8	m/s
Ct	3598	3325	mg min/m^3
t	45	45	min
2ΔR	3.2	3.6	mm
R	5	5	cm
d_{mat}	0.5	0.5	mm

deposition velocity was calculated with Eq. 13, using the observed experimental conditions as given in Table 1.

The experiments have been carried out with carbon containing paper as the adsorbing material. This material has a much higher adsorptive capacity than the human skin. Therefore, the amount of vapors adsorbed onto this carbon paper will be much higher than the amount, which will be adsorbed onto a similar area of human skin. However, it is expected that the vapor deposition on the skin will show similar wind speed effects as found in this study. The permeability of a polyethylene membrane toward methylsalicylate has been found to be in the same order of magnitude as the permeability of pigskin.[9] Pigskin itself has been shown to be a good simulant for human skin.[10] Therefore, some preliminary experiments were performed, where the carbon paper was covered by a polyethylene membrane, thickness around 15 micrometer.

Calculations were performed with the same set of conditions as used in the experiments (Table 1). The diffusion coefficient of methylsalicylate is $5.7*10^{-6}$ m^2/s. The porosity of the clothing is not known. A "worst case scenario" is assumed by using a porosity of $\varepsilon = 1$. Standard values, like the density of air were taken as described previously,[1] or were taken from literature.[11]

Results and Discussion

Environmental conditions

By using the model described previously,[1] the air velocity profiles through and underneath the clothing were calculated. These profiles are shown in Figures 3 and 4.

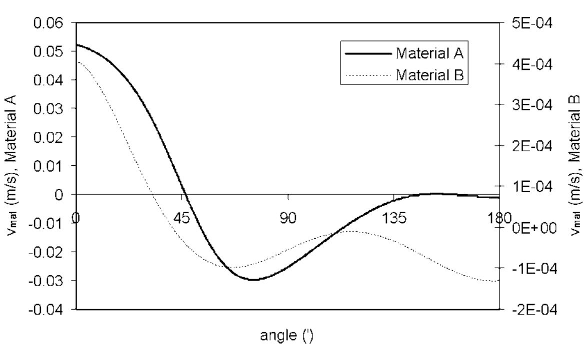

Figure 3. Calculated velocity through clothing around a body part as a function of the angle.

Negative values indicate flow leaving the air gap between clothing and cylinder through the clothing.

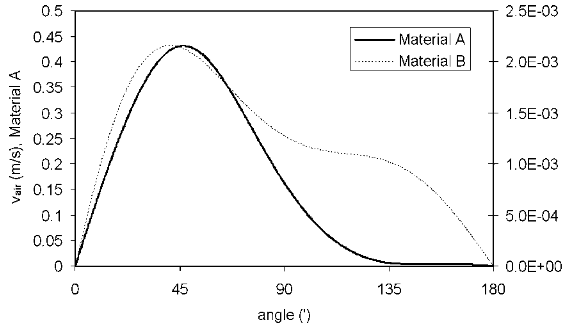

Figure 4. Air velocity underneath the clothing around a body part as a function of the angle.

The calculated velocity profiles through the clothing are similar to those calculated before,[1] and furthermore, they have the same shape as the radial velocity profile, calculated by means of CFD.[2,3,8] Note that the velocity at the back (above 150°) for this situation is almost equal to zero. The calculated velocity underneath the clothing is also similar to those published earlier,[1] and has the same shape as the tangential velocity profile calculated by means of CFD.[2,3,8] The velocity through the clothing at the back is very low for this situation. Therefore, the velocity underneath the clothing is also very low (even backflow is possible for some situations).

Vapor concentration

The velocity profiles, presented in the previous paragraph, were used to calculate the concentration profile and the dosage profile underneath the clothing. These profiles are shown in the Figures 5 and 6.

Since the concentration is not time-dependent, the concentration profile and the dosage profile are similar (Figures 5 and 6). At the back of the cylinder a decrease is observed. This is due to the fact that the velocity underneath the clothing is almost zero for this situation. Thus, contaminated air does not reach this side of the cylinder in this situation.

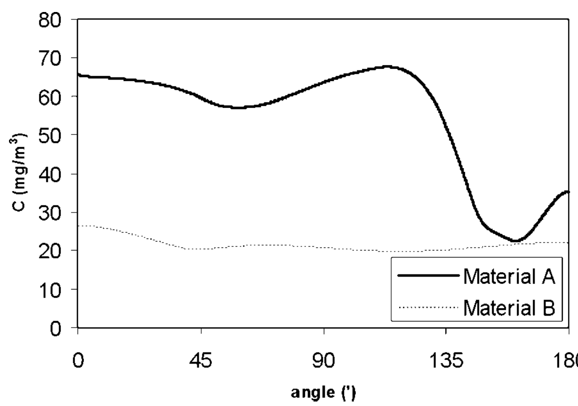

Figure 5. Concentration profile of vapor underneath clothing around a body part as a function of the angle.

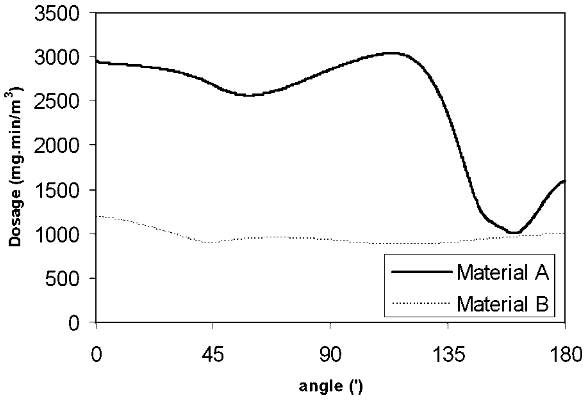

Figure 6. Dosage profile of vapor underneath clothing around a body part as a function of the angle.

Deposition velocity

The measured deposition velocities and the calculated mass-transfer coefficients are shown in Figure 7.

The agreement between the measurements and the calculations is reasonably well. Especially the material with high-air permeability shows good agreement. Furthermore, the shape of the deposition velocity profile shows very good agreement with the shape of the Sherwood number, calculated by means of CFD[3].

In the calculations a constant distance between the clothing and the cylinder wall is assumed, which does not necessarily has to be the case in reality. To calculate the deposition velocity underneath the clothing from the experiments, it was assumed that the dosage underneath the clothing was equal to the dosage outside the clothing. In reality, this dosage will be somewhat lower (Figure 6), which will result in a higher measured deposition velocity (Eq. 13), and will give an even better match between calculations and experimental values.

The concentration profile and the calculated mass-transfer coefficients (see next paragraph) were used to calculate the mass deposition on the surface (Figure 8). The calculated mass deposition decreases gradually as a function of the angle.

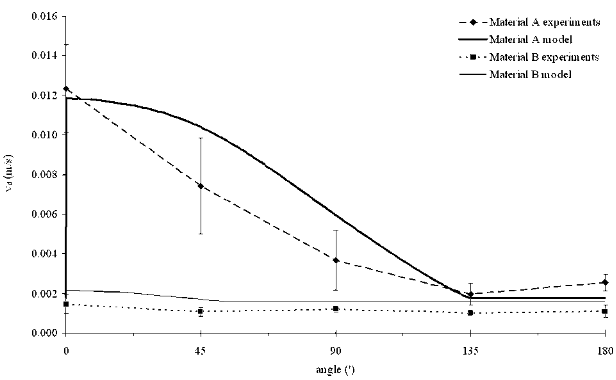

Figure 7. Deposition velocities as a function of the angle around the cylinder.

Measurements (interrupted line) calculations (uninterrupted line). The total length of the error bars is two times the standard deviation.

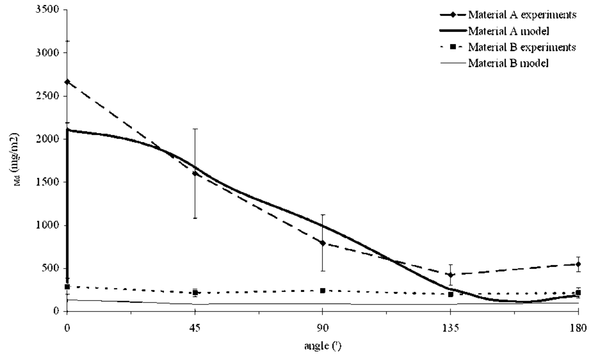

Figure 8. Mass deposition onto the surface of the cylinder as a function of the angle.

The deposition velocity is expected to be influenced by the velocity under the clothing (Eq. 8). However, at an angle of 0°, this velocity equals zero, but at the same time the velocity through the clothing is at its maximum. To analyze this effect of the wind velocity on the deposition velocity, the deposition velocity is plotted as a function of the wind velocity through (angle = 0°) and under (angle = 45°) the clothing (Figure 9). The measured deposition velocity values were fitted by a square root function of the wind velocity. As can be seen, the vapor deposition velocity can be well described as a function of the square root of the wind velocity, which can be expected according to the model (Eq. 8).

As simulant for human skin the combination of a polyethylene membrane on top of carbon paper was used. Figure 10 illustrates the measured deposition velocity for this polyethylene – carbon combination. Applying the membrane not only reduces the absolute magnitude of the vapor deposition, but also reduces the influence of the wind speed. This indicates that the permeation through the membrane is a significant rate determining step in the deposition process.

Conclusions

A model, which describes the mass-transfer process under the protective clothing, was developed and validated. Deposition profiles of vapor onto the surface of a cylinder underneath clothing around a cylinder were measured and calcu-

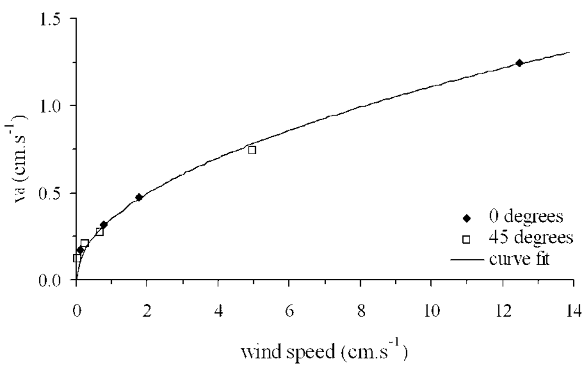

Figure 9. Vapor deposition velocity as a function of the wind speed through 0°, and under 45° the clothing material.

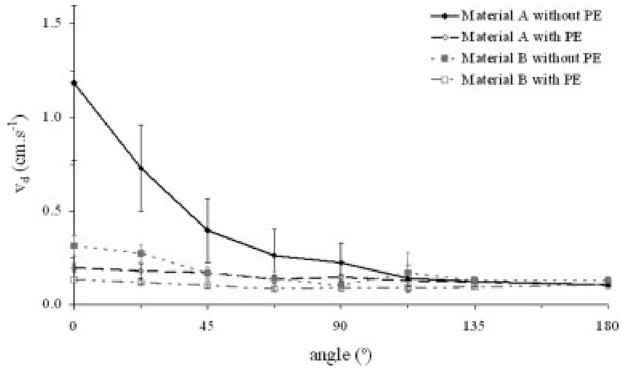

Figure 10. Influence of applying a polyethylene membrane as a dosimeter on the experimental vapor deposition at wind speed 5.1 m/s.

lated. The agreement between the measurements and the calculations is reasonably well.

The deposition velocity and the mass deposition are at its maximum at the front of the cylinder compared to the wind direction.

Higher wind speeds and higher air permeabilities of the clothing will lead to higher depositions. The best NBC-protective clothing will be a compromise between comfort and protection. Thus, a clothing type which has a medium air permeability will probably be the best.

Vapor deposition on real skin was simulated by using a polyethylene membrane on top of carbon paper as surrogate skin. The measured vapor depositions on this surrogate skin were lower than the worst case vapor depositions as found by measurement with single carbon paper.

Further studies will be done for the evaluation of other parameters, which may influence the vapor deposition velocity, such as the diameter of the cylinder, and the distance between the clothing and the cylinder.

Moreover, the translation of these vapor deposition velocities to toxic effects on the skin will be investigated.

Acknowledgments

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Notation

C = concentration, kg/m^3
C_d = outside concentration, kg/m^3
D = diffusion coefficient, m^2/s
d_{mat} = thickness clothing, m
g = gravity constant, m/s^2
k_g = mass-transfer coefficient, m/s
M_d = mass deposition, kg/m^2
R = radius of cylinder, m
ΔR = half size of air gap, m
R_{cloth} = radius of clothing cylinder, m
Re = reynolds number
Sc = schmidt number
Sh = sherwood number
t = time, s
v_0 = wind velocity, m/s
v_{air} = air velocity in air gap, m/s
v_d = (mass) deposition velocity, m/s
v_{mat} = air velocity through clothing, m/s
x = distance along surface of cylinder, m

Greek letters

δ = boundary thickness, m
ε = porosity of clothing
η = dynamic viscosity of air Pa s
Γ = air permeability of clothing, m/(Pa s)
ρ_l = density of air, kg/m^3
ρ_w = density of water, kg/m^3
θ = angle around cylinder, °

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