

# Constitutive model of filled elastomers capable of capturing Mullins effect, hysteresis, induced anisotropy and permanent set – Part I. Model theory & Implementation

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## Abstract

In this contribution, a finite element implementation of a micromechanically based constitutive model describing several inelastic effects of filled rubbers in multiaxial deformation states is presented. The model describes the elastic and inelastic effects of filled rubbers and is based on the network decomposition concept. Accordingly, the rubber network is decomposed into an isotropic elastic network E, responsible for the polymer matrix, and two anisotropic permanent damage networks (M and H) which are responsible for the filler-polymer interaction. The anisotropic damage networks M and H are capable of capturing the Mullins effect, hysteresis, permanent set and induced anisotropic stress softening.

This model is implemented into LS-DYNA<sup>®</sup> by means of a subroutine within *\*MAT\_USER\_DEFINED\_MATERIAL\_MODELS (UMAT)*. The user can easily switch between different combinations of elastic and inelastic models by activating and deactivating each network. The user can also select the number of directions to be considered depending on the complexity of the loading history. The model with appropriate material constants demonstrates good agreement with experimental data.

## 1 Introduction

Elastomers are characterized by their ability to undergo large elastic deformations. Modeling of rubber-like materials have been widely addressed in recent years due to the increasing number of industrial applications. A reliable prediction of material behavior of elastomer is still very challenging due to the variety of complex mechanical properties. In particular, the mechanical response of elastomers is highly non-linear and exhibits hysteresis and stress softening (Mullins effect) upon cyclic loading. There is also some amount of permanent set upon removal of the applied load.

Rubber-like materials are widely used in the automotive industry in variety of parts, such as seals, hoses, tires, brakes, engine and transmission mounts. The simulation of such rubber materials is becoming increasingly important in particular in automotive crashworthiness simulations. Nowadays, the design of these highly technical parts demands the use of a FE software for the numerical analysis. The implementation of an appropriate constitutive model of an elastomeric material in FE solver is a necessary prerequisite for a good numerical prediction. Although highly sophisticated material laws are available, no constitutive models capturing Mullins effect, induced anisotropy and permanent set are currently available in LS-DYNA.

In LS-DYNA there exist several constitutive models for rubber, but many of them are incapable of modeling all the inelastic phenomena of interest - Mullins effect, induced anisotropy and permanent set. For example, *\*MAT\_077* (Ogden or general hyperelastic model) and *\*MAT\_181* (simplified rubber model) are two rubber models which are extensively used in industries for various applications.

*\*MAT\_OGDEN\_RUBBER* & *\*MAT\_HYPERELASTIC\_RUBBER* material models reproduce the quasi-static uniaxial tension test results based on parameters resulting from the fit to the corresponding experimental data. The hysteresis effect is captured by providing additionally unloading curves for some specific values of the deviatoric strain energy density.

*\*MAT\_SIMPLIFIED\_RUBBER* material model provides a rubber model defined by single uniaxial load curve. It reproduces the quasi-static uniaxial tension and compression tests exactly without fitting. The

hysteresis is reproduced by defining a single uniaxial unloading curve or two-parameter formulation (HU & shape).

Although these models provide an accurate or even exact fit to experimental curves, they fail to capture anisotropic stress softening under cyclic loading (the Mullins effect) and a permanent set (residual deformation after unloading). In order to take these effect into account, a micromechanically based user-defined material model was implemented based on the analytical network averaging concept developed in [1]. The model is implemented in LS-DYNA using the user defined material subroutine (UMAT), which captures all the phenomena of interest. The model was further validated in comparison to experimental data from the uniaxial cyclic tension [2]. In this contribution, the implementation of the material model and its numerical features are discussed.

## 2 Implementation of user defined material subroutine

### 2.1 Constitutive model

The network averaging model describes the elastic and inelastic behavior of filled elastomers in multiaxial states of deformation based on a network decomposition concept. Accordingly, the rubber network is decomposed into a purely elastic rubber network (E) containing polymer subnetworks with crosslinks at both ends, and two anisotropic damage networks (M and H) containing polymer subnetworks attached to two filler particles [1].

The strain energy function (per unit referential volume) of the entire network is formulated as,

$$\psi = \psi_E + \psi_M + \psi_H, \quad (1)$$

$$\psi = \underbrace{\frac{\mu_c \psi_c(n, \bar{\lambda})}{A}} + \underbrace{\frac{\mu_t \bar{v}}{B}} + \underbrace{\sum_{i=1}^{21} [\mu_{Mi} \psi_c(n, \lambda_{Mi})]}_C + \underbrace{\sum_{i=1}^{21} [\mu_{Hi} \psi_c(n, \lambda_{Hi})]}_D + \underbrace{\frac{\kappa}{4} [(J-1)^2 + (\ln J)^2]}_U, \quad (2)$$

where  $\kappa$  is the bulk modulus,  $J = \det \mathbf{F}$  is the relative volume change and  $\mathbf{F}$  is the deformation gradient. Terms A and B are responsible for the linear elastic behavior at low strains ( $\psi_E$ ). Term C controls the stress softening under cyclic loading condition ( $\psi_M$ ). Term D captures the hysteresis loops due to unloading at large strains ( $\psi_H$ ) and the last term U enforces the incompressibility condition.

The other terms of the strain energy function are expressed as follows,

$$\mu_{Mi} = \frac{\mu_c \delta}{m} \frac{C_0}{(1-C_0)} \phi(\Lambda_i^{max}), \quad \mu_{Hi} = \frac{\mu_c (1-\delta)}{m} \frac{C_0}{(1-C_0)} \phi(\Lambda_i^{max}), \quad (3)$$

$$\psi_c(n, \bar{\lambda}) = n \ln \frac{\pi \cdot x}{\sqrt{n} \sin(\frac{\pi \cdot x}{\sqrt{n}})}, \quad \bar{\lambda} = \left( \sqrt{\frac{I_1}{3}} \right)^q, \quad \bar{v} = \sqrt{\frac{I_2}{3}}, \quad (4)$$

$$I_1 = \text{tr} \bar{\mathbf{C}}, \quad I_2 = \frac{1}{2} [(\text{tr} \bar{\mathbf{C}})^2 - \text{tr} \bar{\mathbf{C}}^2], \quad (5)$$

where  $I_1$  and  $I_2$  are the principle invariants of the isochoric right Cauchy-Green tensor  $\bar{\mathbf{C}}$ ,

$$\bar{\mathbf{C}} = J^{-\frac{2}{3}} \mathbf{C}, \quad \mathbf{C} = \mathbf{F}^T \mathbf{F}, \quad (6)$$

while

$$\lambda_{Mi} = \left( \sqrt{\bar{\mathbf{C}} : \mathbf{E}_i \otimes \mathbf{E}_i} \right)^{q_m(\Lambda_i^{max})}, \quad \lambda_{Hi} = \left( \sqrt{\bar{\mathbf{C}} : \mathbf{E}_i \otimes \mathbf{E}_i} \right)^{q_m(\Lambda_i^{cmax})}, \quad (7)$$

$$q_M(x) = \frac{q}{1 - [C_0 \phi(x)]^{\frac{1}{3}}}, \quad \phi(x) = \exp \left( \frac{\bar{\alpha}}{4\pi} \left[ n \sin \frac{2\pi x L}{n} - 2\pi x L \right] \right). \quad (8)$$

The internal variables at time  $t$  are defined as,

$$\Lambda_i^{max} = \max_{\tau \in (-\infty, t]} \Lambda_i(\tau), \quad \Lambda_i^c{}^{max} = \max_{\tau \in [t_i^c, t]} \Lambda_i(\tau), \quad \Lambda_i = \sqrt{\mathbf{C} : \mathbf{E}_i \otimes \mathbf{E}_i} \quad (10)$$

where the time at the end of the previous cycle  $t_i^c$  is defined through the next local minimum value of  $\Lambda_i$ .

$\mathbf{E}_i$  ( $i = 1, 2, \dots, m$ ) denote unit vectors specifying preferred directions, where  $m$  is the number of directions considered for induced anisotropy. For the M and H networks the energy function is obtained by summation over  $m=21$  directions of anisotropy (Eq. 2). The coordinates of 21 points on the unit sphere specifying these directions are given in Appendix.

| Parameters     | Physical meaning   |
|----------------|--|
| $\mu_c$        | Effective phantom shear modulus of pure rubber network [MPa] |
| $n$            | Average number of chain segments                             |
| $q$            | Stretch amplification exponent                               |
| $\mu_t$        | Topological shear modulus of the pure rubber network [MPa]   |
| $\bar{\alpha}$ | Functionality of adsorption area                             |
| $\bar{L}$      | Average referential distance of subnetworks                  |
| $\delta$       | Fraction of broken chains                                    |
| $C_o$          | Volume fraction of fillers                                   |
| $\kappa$       | Bulk modulus   |

Table 1: Physical meaning of material constants

The response of the network averaging model depends on 9 material constants, whose physical meanings are described in Table 1.

## 2.2 Constitutive relations

In this section, the derivative of the strain energy function is presented in order to obtain the second Piola-Kirchhoff stress tensor  $\mathbf{S}$ . The Cauchy stress  $\boldsymbol{\sigma}$  is the output of the material subroutine at each time step, which can be derived as follows.

The strain energy function (Eq. 2) can be split into an isochoric (iso) and a volumetric (vol) part as follows,

$$\psi(\mathbf{C}) = \underbrace{\bar{\psi}(\bar{\mathbf{C}})}_{iso} + \underbrace{U(J)}_{vol}. \quad (11)$$

In Eq. 2, the terms A, B, C and D are represented by  $\bar{\psi}(\bar{\mathbf{C}})$  and the volumetric part of energy function is written as  $U(J)$ . Thus, from Eq.11 we get,

$$\mathbf{S} = \underbrace{2\bar{\psi}(\bar{\mathbf{C}})_{,c}}_{S_{iso}} + \underbrace{2U(J)_{,c}}_{S_{vol}}, \quad (12)$$

$$S_{iso} = \underbrace{2\bar{\psi}(\bar{\mathbf{C}})_{,c}}_{\bar{\mathbf{S}}} : \underbrace{\bar{\mathbf{C}}_{,c}}_{\mathcal{P}_{iso}} \quad (13)$$

where  $\mathcal{P}_{iso}$  denotes the isochoric projection tensor [3].

$$\mathbf{S} = \left( 2\mu_c \frac{\partial \bar{\psi}}{\partial \lambda} \frac{\partial \bar{\lambda}}{\partial I_1} \frac{\partial I_1}{\partial \mathbf{C}} + 2\mu_t \frac{\partial \bar{\psi}}{\partial I_2} \frac{\partial I_2}{\partial \mathbf{C}} + \sum_{i=1}^{21} 2\mu_{Mi} \frac{\partial \bar{\psi}}{\partial \lambda_{Mi}} \frac{\partial \lambda_{Mi}}{\partial \mathbf{C}} + \sum_{i=1}^{21} 2\mu_{Hi} \frac{\partial \bar{\psi}}{\partial \lambda_{Hi}} \frac{\partial \lambda_{Hi}}{\partial \mathbf{C}} \right) : \frac{\partial \bar{\mathbf{C}}}{\partial \mathbf{C}} + 2 \frac{\partial U}{\partial \mathbf{C}}, \quad (14)$$

$$\boldsymbol{\sigma} = \frac{1}{J} \mathbf{F} \mathbf{S} \mathbf{F}^T. \quad (15)$$

The Cauchy stress is therefore calculated by the material subroutine for every element at each time step based on the Eq. 14-15.

### 2.3 User defined material

In this section, the definition for different variables of the user defined material card is presented. The constitutive model described in Section 2.1 was implemented in the LS-DYNA material subroutine. The keyword input shown in Fig. 1 is used to define the material properties for the subroutine. The user is able to activate or deactivate the elastic and inelastic networks and as well as to switch on/off the hysteresis and Mullins effects by using an additional material parameter.

| Card 1   | 1   | 2  | 3  | 4   | 5   | 6                | 7     | 8  |
|----------|-----|----|----|-----|-----|------------------|-------|----|
| Variable | MID | R0 | MT | LMC | NHV | IORTHO/<br>ISPOT | IBULK | IG |
| Type     | A8  | F  | I  | I   | I   | I                | I     | I  |

| Card 2   | 1     | 2     | 3      | 4      | 5    | 6    | 7 | 8 |
|----------|-------|-------|--------|--------|------|------|---|---|
| Variable | IVECT | IFAIL | ITHERM | IHYPER | IEOS | LMCA |   |   |
| Type     | I     | I     | I      | I      | I    | I    |   |   |

Fig.1: Material card of \*MAT\_USER\_DEFINED\_MATERIAL\_MODELS

The number of material parameters defined in the user subroutine is specified in the material card under the variable LMC (Length of Material Constant) as shown in Fig. 1. As discussed in Section 2.1, totally 9 material constants are responsible for the behavior of network averaging model of rubber material. In addition, a switch parameter also helps the user to switch between different networks of the model. Therefore, totally 10 material parameters are defined in LMC. The LMC material parameters are defined in the card as shown in Fig. 2 with 8 parameters per card.

| Card     | 1  | 2  | 3  | 4  | 5  | 6  | 7  | 8  |
|----------|----|----|----|----|----|----|----|----|
| Variable | P1 | P2 | P3 | P4 | P5 | P6 | P7 | P8 |
| Type     | F  | F  | F  | F  | F  | F  | F  | F  |

Fig.2: LMC material parameters definition card

User defined history variables are used when the preceding value in previous time step have to be used in the current time step. Since the value of internal variables, such as  $\Lambda_i$ ,  $\Lambda_i^{max}$  and  $\Lambda_i^{c max}$  at current time step depend on the value of the previous time step, they are considered as history variables (NHV). These internal variables are also directionally dependent and their update at the beginning of each loading cycle is important for the prediction of the directional sensitivity of stress softening.

The number of history variables is defined in the material card under NHV. In the material subroutine, 4 history variables are used in total and each history variable is defined for every of 21 preferred directions.

Therefore, 84 history variables are defined in NHV. Note that the number of the preferred directions can be reduced depending on the complexity of the loading.

The 9 components of deformation gradient calculated by LS-DYNA at each time steps are also saved as history variables. These history variables are allocated after the last index of user defined history variables (NHV) by default.

### 3 Simulation and results

#### 3.1 FE Model set-up

For the verification purposes, a uniaxial cyclic tension test was simulated in LS-Dyna. A solid element as shown in Fig. 3 with a specimen gauge length, width and thickness equal to 1 mm and the default constant stress solid element formulation (ELFORM=1) was used. The solid element was subjected to a kinematically controlled loading and unloading. The boundary conditions (as shown in Table 2) were defined to different nodes using the keyword `*BOUNDARY_PRESCRIBED_MOTION`. The maximal strain of 120% strain was applied with an increment of 40%.

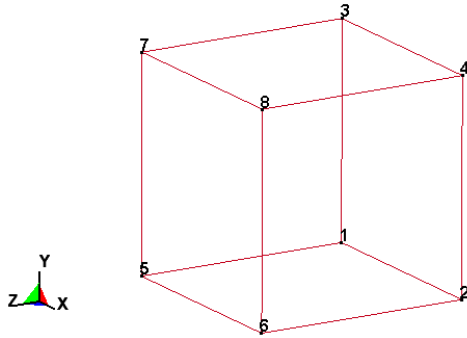


Fig.3: 3D Solid Element

| Nodes   | Boundary conditions                                       |
|---------|---|
| 1       | Fixed in Y direction                                      |
| 2       | Fixed in X and Y directions                               |
| 5       | Fixed in X, Y and Z directions                            |
| 6       | Fixed in Y and Z direction                                |
| 3,4,7,8 | Fixed in X and Z direction<br>Displacement in Y direction |

Table 2: Boundary conditions defined to FE Model (refer Fig. 3 for node definition)

The `*MAT_USER_DEFINED_MATERIAL_MODELS` material card was defined as explained in Section 2.3.

The 10 material parameters are defined in the LMC parameter definition card (see Fig. 2) in the order represented in Table 3.

| Variables | Parameters  |
|-----------|---|
| P1        | $\mu_t$   |
| P2        | $\kappa$  |
| P3        | $n$   |
| P4        | $q$   |
| P5        | $\mu_c$   |
| P6        | $C_o$   |
| P7        | $\bar{L}$   |
| P8        | $\bar{\alpha}$  |
| P9        | $\delta$  |
| P10       | switch between different networks<br>EQ.1: Only elastic network<br>EQ.2: Elastic and inelastic network with Mullins effect and hysteresis<br>EQ.3: Elastic and inelastic network with Mullins effect<br>EQ.4: Elastic and inelastic network with hysteresis |

Table 3: Parameter definition to corresponding variables in material card

Note that for a hyperelastic material, the IHYPER variable in material card 2 (see Fig.1) should be set to 1. The IBULK and IG variables in card 2 are set to 2 and 1, which automatically takes the value of the material parameter P2 and P1 in card 3, corresponding to the bulk modulus and shear modulus of the material, respectively.

### 3.2 Results and discussion

In this section, predictions of the implemented material model with all inelastic effects are presented. The Fig. 4 shows the Cauchy stress plotted versus stretch in uniaxial tension by considering 21 preferred directions and taking both inelastic networks into account (Table 3). The Mullins effect and hysteresis upon loading and unloading as well as the permanent set are appropriately captured by the model.

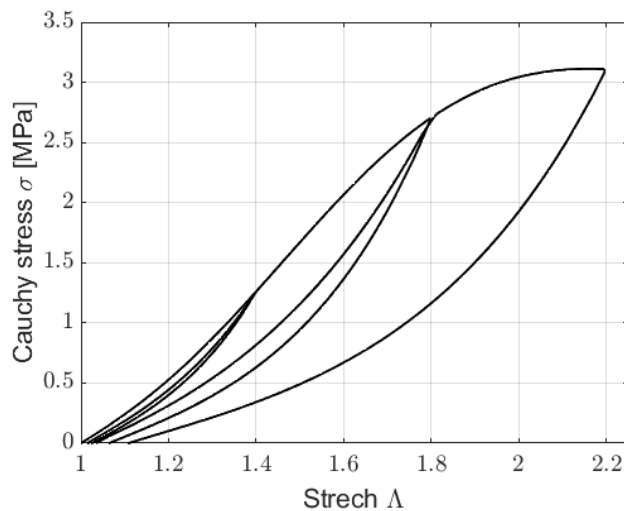


Fig.4: Cauchy Stress vs. stretch predicted by the implemented model with all inelastic effects (the Mullins effect, hysteresis and permanent set)

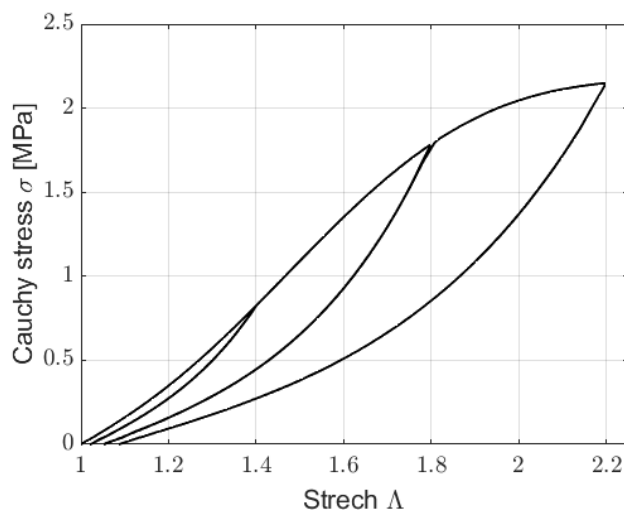


Fig.5: The inelastic response of the material by only activating M network (the Mullins effect)

As discussed in the Section 2.3, the user defined material is able to capture the inelastic effects separately by using a switch parameter (refer Table 3). Predictions of the model with only Mullins effect (switch 3) and only with hysteresis (switch 4) are shown in Fig. 5 and Fig. 6, respectively. As it can be seen in Fig. 5, the reloading curve follows the previously unloaded curve (no hysteresis) up to the maximum stretch achieved during the initial loading. Afterward, it repeats the initial loading curve

(primary curve). Such behavior is referred to as the Mullins effect. Similarly, the area between the reloading curve and the previously unloaded curve (hysteresis loop) is captured by the model as shown in Fig. 6.

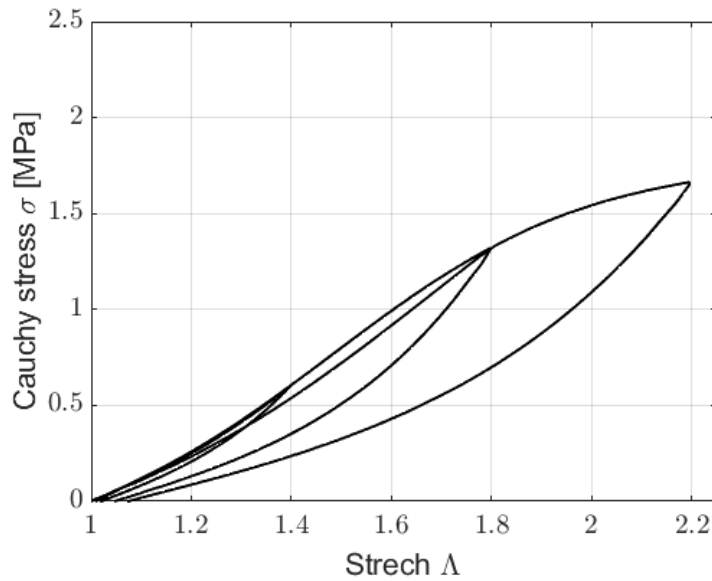


Fig.6: The inelastic response of the material by only activating H network (hysteresis)

The evolution of the Mullins effect is governed by  $\Lambda_i^{max}$  which remains the same as long as the previous maximal stretch in the direction  $i$  is not exceeded. The evolution of the hysteresis is described by  $\Lambda_i^{c max}$  which recovers after a loading cycle (see Eq. 10). The evolution of the maximal stretch and maximal cyclic stretch in the direction of loading is described by the user defined material model as illustrated in Fig. 7.

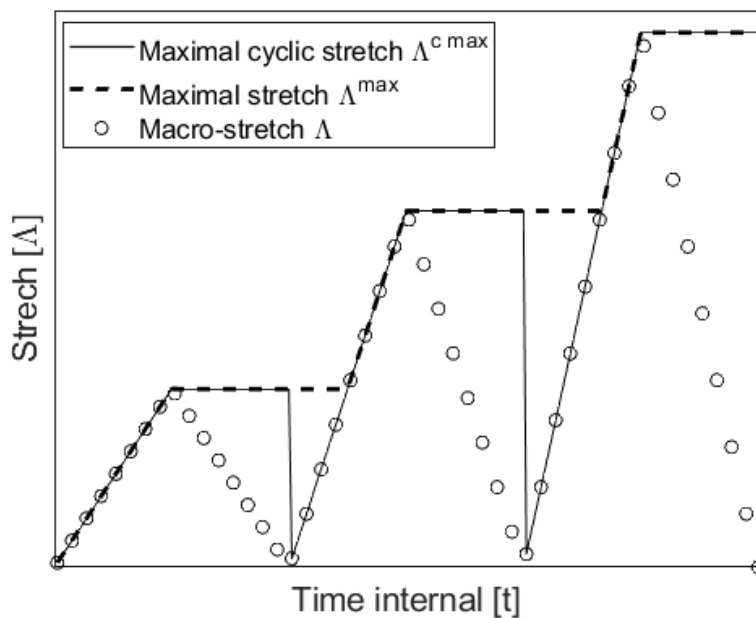


Fig.7: Evolution of the maximal cyclic stretch ( $\Lambda_i^{c max}$ ), maximal stretch ( $\Lambda_i^{max}$ ) and macro-stretch ( $\Lambda_i$ )

#### 4 Summary

In the current work, the network averaging model [1] describing several inelastic effects of elastomers such as the Mullins effect, hysteresis, induced anisotropy and permanent set is implemented in the LS-DYNA user defined material subroutine. Accordingly, the rubber network is decomposed into an

elastic network (E), an anisotropic permanent damage network (M) and an anisotropic recoverable network (H). This facilitates the user to switch between different networks depending on the occurrence of these characteristics in the material. The number of preferred directions of anisotropy can also be modified depending on the tension of loading. The behavior of the implemented material model was verified by simulating uniaxial, which showed good results in comparison with the results of same material model implemented in MATLAB. Uniaxial cyclic loading was further simulated for the validation and parameter identification in comparison to experimental data [2].

## 5 Appendix

| X        | Y         | Z         |
|----------|-----------|-----------|
| 1        | 0         | 0         |
| 0        | 1         | 0         |
| 0        | 0         | 1         |
| 0,707107 | 0,707107  | 0         |
| 0,707107 | -0,70711  | 0         |
| 0,707107 | 0         | 0,707107  |
| 0,707107 | 0         | -0,70711  |
| 0        | 0,707107  | 0,707107  |
| 0        | 0,707107  | -0,70711  |
| 0,387907 | 0,387907  | 0,836096  |
| 0,387907 | 0,387907  | -0,836096 |
| 0,387907 | -0,387907 | 0,836096  |
| 0,387907 | -0,387907 | -0,836096 |
| 0,387907 | 0,836096  | 0,387907  |
| 0,387907 | 0,836096  | -0,387907 |
| 0,387907 | -0,836096 | 0,387907  |
| 0,387907 | -0,836096 | -0,387907 |
| 0,836096 | 0,387907  | 0,387907  |
| 0,836096 | 0,387907  | -0,387907 |
| 0,836096 | -0,387907 | 0,387907  |

Table 4: Coordinates of the point on the unit sphere defining preferred directions of anisotropy

## 6 Literature

- [1] V.N. Khiêm and M. Itskov: "An averaging based tube model for deformation induced anisotropic stress softening of filled elastomers", International Journal of Plasticity 90, 96-115
- [2] M. Hillgärtner, R. Chandrasekaran, M. Müller, F. Burbulla, and M. Itskov: "Constitutive model of filled elastomers capable of capturing Mullins effect, hysteresis, induced anisotropy and permanent set – Part II: Experiments & Validation", Proceedings of the 12th European LS-DYNA Conference, Koblenz, Germany, 2019
- [3] M. Itskov: Tensor Algebra and Tensor Analysis for Engineers, fifth edition