

Analysis of Mechanical Milling in Simoloyer: An Energy Modeling Approach

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A model was developed to estimate the energy transfer from milling media to the powder during milling carried out in Simoloyer CM 01 (Zoz GmbH, Wenden, Germany), a horizontal attritor high-energy ball mill. The model was then used to estimate the energy transfer in milling of iron at 1000 rpm. Furthermore, the time required to achieve a particular grain size was formulated as a function of milling speed, using the model developed for the energy transfer. The results were verified at 500 rpm and 1500 rpm for iron and aluminum.

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I. INTRODUCTION

HIGH-ENERGY ball milling is an established route to reduce the size of the coarse grains to nanosizes.^[1-3] It involves impact/shear of balls with powder, which subsequently results in the deformation and fracture of particles leading to nanostructures through dynamic recrystallization. Collision regime is considered to be the main form of grain refinement during high-energy ball milling. The energy imparted to the powder during impact/shear is responsible for crack propagation and the fracture of particles.^[2,3]

Modeling of high-energy ball milling has received renewed attention,^[4] primarily to develop novel alloys by lesser use of energy. In that context, Abdelloui and Gaffet^[5] established a mathematical approach to illustrate the mechanics of planetary milling, which was used for the development of milling maps by Murty and coworkers.^[6,7] Similarly, an *ab initio* mathematical model was developed by Chattopadhyay *et al.*^[8] to demonstrate mechanics of planetary milling. Dallimore and McCormick^[9] worked on the dynamics of milling media motion in planetary mills. There were also attempts to understand the energy transfer in the vibratory ball mills. Zhong *et al.*^[10] analyzed the mechanics of a high-energy vibrator mill. An attempt to measure the impact velocities of balls in Spex mills was done by Basset *et al.*^[11] The comminution process of the Spex mill was studied by Concas *et al.*^[12] and Ward *et al.*^[13] Maurice and Courtney^[14] developed a model based on Hertzian collisions, which was used by Magini and Iasonna^[15] to calculate the energy transferred to the powder per collision. Similar work has been done by Joardar *et al.*,^[16] who formulated a model to determine the temperature of the entrapped powder particle between balls. Sasikumar *et al.*^[17] analyzed the distribution of the energy in various forms during the course of milling.

Recently, a horizontal attritor mill (Simoloyer mill) has gained popularity because of its high energy and faster milling kinetics. The Zoz Group^[18-20] worked on the mechanism of milling and the fundamentals of energetics in the simoloyer. However, extensive calculations on the energetics of the simoloyer were not reported so far. This work is an attempt to understand the energy transfer mechanisms occurring in the simoloyer mill and to predict the milling duration required to achieve a particular nanocrystalline grain size.

II. EXPERIMENTAL PROCEDURE

Simoloyer CM01 (Zoz GmbH, Wenden, Germany), having double-walled stainless steel vial and stainless steel impeller (10 arms) was employed in the current work. In all, 1 kg of high-chrome steel balls (100Cr6) of 5 mm in diameter were used with a ball-to-powder ratio of 10:1. Commercial pure iron powder (325 mesh, 99 pct) and aluminum powder (325 mesh, 99 pct) were subjected to milling in the current study.

The mass flow rate of the cooling water was maintained at 28 g/s. The temperature difference between the inlet and the outlet pipes was measured using a thermometer with a resolution of 0.1 K (0.1 °C). Care was taken to ensure that temperature measurements were always done above 298 K (25 °C) so that atmospheric effects on the heat measured were nullified. Milling was carried out for 6 hours and samples were collected every 15 minutes. The milled samples were analyzed by X'pert PRO PANalytical (PANalytical, Almelo, The Netherlands), X-ray diffraction (XRD) using Cu K_α radiation ($\lambda = 0.15402$ nm).

III. WORKING MODEL

The assumptions made in the present model are as follows:

- The milling medium (balls) are assumed to be coated with powder.

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- All the collisions are direct (head on) in nature.
- The balls after being charged are assumed to be in the form of a uniform semicylinder.
- Energy losses from noise and unknown factors are ignored.

The rotating spindle is the primary source of kinetic energy in the mill (Figure 1). The balls receive energy from spindle, which is rotating at constant speed, and proceed toward the vial wall. The following series of collisions transferring energy to the powder are possible during milling:

- Ball energized by spindle collides with the vial wall.
- Energized ball collides with another ball rebounded from vial wall.
- Ball after rebounding from vial wall collides with another similar ball.
- Energized ball collides with another energized ball.
- Rebounded ball collides with the spindle.

The energy transferred to the powder in processes (c) and (e) are much lesser than the energy transferred in process (b). Hence, one can ignore the two processes. Maximum energy is transferred to the powder in case of process (d). However, the coefficient of restitution between the two coated balls was observed to be zero, as would be explained subsequently. This would imply that the balls come to rest immediately without colliding with the vial surface. Because this is not practically observed, processes (a) and (b) were considered the as the primary source of energy transfer.

The notion of a steady-state motion in a simoloyer hence is considered to be the sequence of collisions involving the following processes:

- Ball–spindle collision (the ball at rest is given energy by the spindle).
- Ball–vial collision (where most of the heat is evolved).
- Ball–ball collision (between a ball collided with the vial and one just given energy by spindle).

The basic energy balance during the milling process in the simoloyer can be represented as^[20]

$$E_{in} = E_{out} + E_{powder} + E_{heat} + E_{noise} + E_{unknown} \quad [1]$$

where E_{in} is the energy input to the balls by the spindle, E_{out} is the energy possessed by the balls after collisions, E_{powder} is the energy transferred to the powder during the collisions, and E_{heat} , E_{noise} , and $E_{unknown}$ are the energies lost during the process of collision in the form of heat, noise, and other unknown factors. The losses of energy from noise and unknown factors are neglected in this model.

The spindle energizes the ball and no transfer of energy to the powder occurs at this stage. On impact with the vial walls, a fraction of energy is transferred to the powder and some energy is lost in the form of heat. The amount of heat loss depends on the value of the coefficient of restitution of the ball, powder, and wall system. The ball rebounds from the wall and meets another ball that is yet to collide with the wall, thereby imparting energy to powder in the collision process. The transferred energy to the powder is a combination of both these processes. Although a significant number

of collisions are oblique in nature, it is extremely difficult to model the balls colliding at varying orientations, and an approximation cannot be obtained in a closed container. Hence, all the collisions are assumed to be direct head-on collisions.

To estimate the energy imparted to the powder, the following parameters are required:

- Quantity of heat evolved
- Energy imparted to powder in ball–vial collision
- Energy imparted to powder in ball–ball collision
- Number of collisions

The total heat released by the system is governed by Eq. [2].

$$Q = mC_p\Delta T t \quad [2]$$

where m and C_p are the mass flow rate and specific heat capacity of the coolant (water), respectively, ΔT is the temperature difference between the inlet and outlet tubes, and t is the milling time.

Once these balls are charged in the vial, they are assumed to arrange themselves into a semicylinder extending to the length of the vial. The cross-section view of the semicylinder is shown in Figure 2. The semicylinder retains its shape throughout the milling duration. This assumption is to simplify the model as the exact arrangement of the balls is difficult to predict. The balls are impacted by the cross-section of the

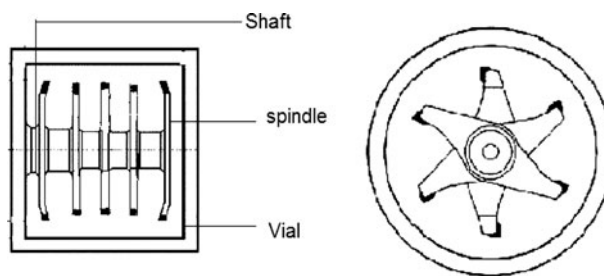


Fig. 1—Schematic diagram of a simoloyer.

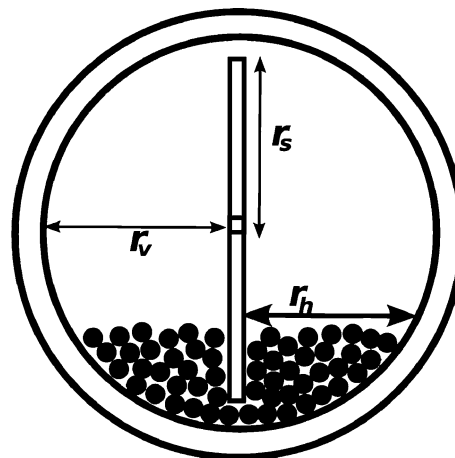


Fig. 2—Schematic diagram of the assumed semicylindrical arrangement of balls in the simoloyer.

spindle arms. Only those balls which are a part of this semicylinder that come in contact with the spindle are imparted kinetic energy. Because the process of milling is a continuous process, all the balls participate in milling during the milling process. The balls are given a velocity (v) of $R\omega$, where R is the distance of the point of contact (center of ball) from the center of the spindle and ω is the angular frequency of the spindle. The kinetic energy carried by each ball is $0.5 m_b v^2$. Thus, the energy of each ball is different and depends on the position of its impact on the length of the spindle.

The radius of each ball used was 0.214 cm and 1 kg balls were used. The number of balls used for milling was 2174 (mass of each ball being 0.46 g) and the volume occupied by the balls was found to be 89 cm^3 . Assuming the balls arrange themselves in a semicylindrical fashion, the radius of the semicylinder can be calculated as follows:

$$2174 * 4/3\pi r_b^3 = 1/2\pi r_h^2 L \quad [3]$$

where r_h and L are the radius and length of the semicylinder formed and r_b is the radius of the ball. The space between the spindle and the vial surface is termed as the dead zone $d(r_{\text{vial}} - r_{\text{spindle}})$. The balls in this region are not in contact with the spindle and, hence, do not receive energy from the spindle. The distance up to which the balls were in contact with the spindle is calculated as $r_h - d$. The number of balls in contact with the spindle is calculated as

$$\text{number of balls} = (r_h - d)/2r_b \quad [4]$$

The R values of the balls are calculated accordingly. The energy of each ball is calculated and an average of the energies is taken. This energy (E_i) is assumed to be possessed by each coated ball. The number of balls in contact with the spindle would vary with the number of balls taken.

The energy transfer to the powder in the simoloyer occurs through a series of collisions. Let the coefficient of restitution value of coated ball, uncoated ball, and powder with the vial be e_t , e_b , and e_p , respectively. The values of coefficient of restitution were found using the rebound tests. A coated ball was dropped from a known height (h_1), and the height to which it rebounded (h_2) back was noted. The coefficient of restitution for a species i and the vial wall system is given by^[15]

$$e_i = \sqrt{(h_{2i}/h_{1i})} \quad [5]$$

e_p is zero as the powder does not rebound. Correspondingly, constants η_t , η_b , η_p are defined such that $\eta_i = e_i^2$. For a general collision between two coated systems, initially before collision, the energy carried by the ball powder system must be equal to the energy possessed by the ball and powder individually.^[15]

$$E_t = E_b + E_p \quad [6]$$

After collision, the energy possessed by the ball powder system, had they been together still, must be equal to the energy possessed by the balls and powder individually after collision.^[15]

$$\eta_t E_t = \eta_b E_b + \eta_p E_p \quad [7]$$

The value of η_p is very small and can be approximated to zero. Solving Eqs. [6] and [7], one gets,

$$E_p = (1 - (\eta_t/\eta_b))E_t \quad [8]$$

where E_p is the energy transferred to the powder per collision of the coated ball with the vial surface.

In this work, an attempt has been made to estimate the total energy transferred to the powder. The energy transferred to the powder is approximated as a combination of two processes. The energy transferred from the impact of the ball with the vial surface per collision is termed as E_{p1} and the energy transferred by the collision with another ball per collision is termed as E_{p2} . Thus Eq. [8] actually gives the value of E_{p1} and, hence, can be written as

$$E_{p1} = (1 - (\eta_t/\eta_b))E_t \quad [9]$$

After colliding with the vial, the rebounded ball returns with an energy of $\eta_t E_t$. A collision between the balls and the vial releases heat. The amount of heat that should be released (q_1), if there is no transfer of energy to the powder, is given by

$$q_1 = (1 - \eta_t)E_t \quad [10]$$

The energy transferred to the powder E_{p1} is absorbed from the heat q_1 . Thus, the heat that is actually removed by the coolant (q) per collision is given as

$$q = q_1 - E_{p1} \quad [11]$$

Combining Eqs. [9] through [11], one can get

$$q = \eta_t E_t ((1/\eta_b) - 1) \quad [12]$$

The rebounded ball collides with another ball that has just received kinetic energy. Experimentally, when a steel ball coated with iron powder was allowed to collide on a similar surface, η_t was observed to be close to zero. Because $\eta_t = 0$, both the balls come to rest immediately, and all the kinetic energy possessed by the balls is imparted to the powder.

$$E_{p2} = E_t + \eta_t E_t = (1 + \eta_t)E_t \quad [13]$$

The number of collisions (N) is given by

$$N = Q/q \quad [14]$$

The total energy given to the powder (E_{PT}) is,

$$E_{PT} = (E_{p1} + E_{p2})N \quad [15]$$

IV. RESULTS AND DISCUSSION

The dimensions of r_{vial} and r_{spindle} are 7.3 and 5.8 cm, respectively, with the dead zone evaluated at 1.5 cm. The length of the vial is 13.5 cm and from

Eq. [3], r_h was calculated to be 2.049 cm. Hence the distance to which the balls were in contact was found to be 0.54 cm. In such a case, only one ball would be in contact with the spindle for every revolution. Because 13 balls can be arranged along the length of the spindle, the distance of the last ball from the center of the spindle would be $25r_b$. From this the average energy of the ball E_t was evaluated to be $312.5mr_b^2\omega^2$. Close to 17 balls are set in motion by the spindle in 1 second, and these balls start the energy transfer process in the simoloyer. A ball energized by the simoloyer hits the vial wall, and it rebounds and collides with another ball energized by the simoloyer in another revolution.

Experimentally, the values determined for e_t and e_b were 0.4 and 0.6, respectively. For iron milled at 1000 rpm for 3 hours in simoloyer, ΔT was experimentally measured as 0.3 K (0.3 °C). An energy analysis for the iron milled at 1000 rpm for 3 hours in simoloyer was performed and the results are given in Table I.

Computing further, the total energy transferred to the iron powder within 3 hours of milling was determined to be 2.28 MJ. From Table I, it is evident that energy transfer to the powder in a ball-ball collision is much greater than in the case of a ball-vial collision. Hence, it can be concluded that ball-ball collisions contribute to almost 2/3 of the total energy transferred to the powder.

The desired grain size in a material can be achieved by milling at a different rpm (ω^*) for a suitable time. Let E_t^* be the E_t corresponding to the new ω^* .

$$E_t^* = E_t\omega^{*2}/\omega^2 \quad [16]$$

Similarly, one can also write,

$$E_{p1}^* = E_{p1}\omega^{*2}/\omega^2 \quad [17]$$

$$E_{p2}^* = E_{p2}\omega^{*2}/\omega^2 \quad [18]$$

$$q^* = q\omega^{*2}/\omega^2 \quad [19]$$

Because the amount of energy required attaining a desired grain size (E_{pT}) is fixed for a given material,^[20]

$$(E_{p1} + E_{p2})N = (E_{p1}^* + E_{p2}^*)N^* \quad [20]$$

Substituting Eqs. [17] and [18] in Eq. [20],

$$N = N^*\omega^{*2}/\omega^2 \quad [21]$$

Table I. Calculated Values of Various Energy Parameters for Iron Milled for 3 Hours at 1000 rpm

Parameter	Value
E_t	7.21 mJ
Q	379 kJ
E_{p1}	4.005 mJ
q	2.0514 mJ
E_{p2}	8.3636 mJ
N	1.84×10^8

Substituting Eqs. [19] and [21] in Eq. [14], one can get

$$Q = Q^* \quad [22]$$

It can be inferred that the heat released during milling, Q , to achieve a particular grain size in a given material would be fixed and would not depend on the rpm used. Hence, from Eq. [2], one can write

$$\Delta T t = \text{constant} \quad [23]$$

Table II. ΔT Measured for the Different Milling Speeds for Iron and Aluminum Milled in Simoloyer

Milling Speed (rpm)	ΔT , Fe [K (°C)]	ΔT , Al [K (°C)]
500	0.2 (0.2)	0.2 (0.2)
1000	0.3 (0.3)	0.4 (0.4)
1500	0.4 (0.4)	0.8 (0.8)

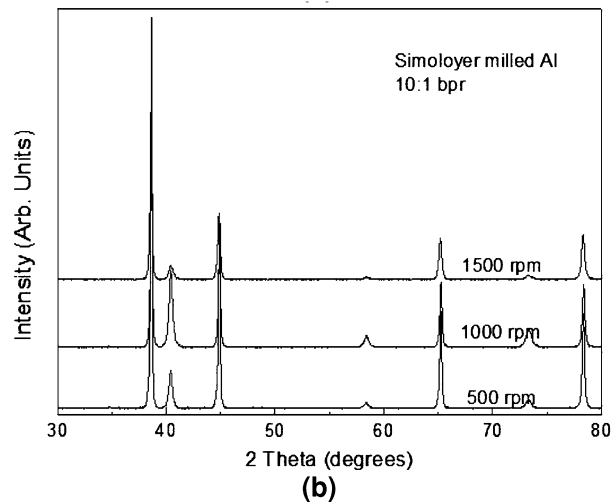
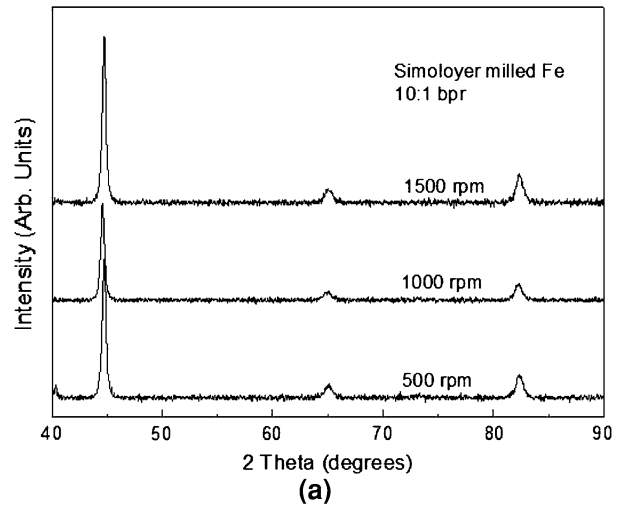


Fig. 3—XRD patterns of (a) Iron milled for at 2 h 15 min, 3 h, and 4 h 15 min at 1500, 1000, and 500 rpm, respectively and (b) aluminum milled for 1 h, 2 h, and 4 h at 1500, 1000, and 500 rpm, respectively.

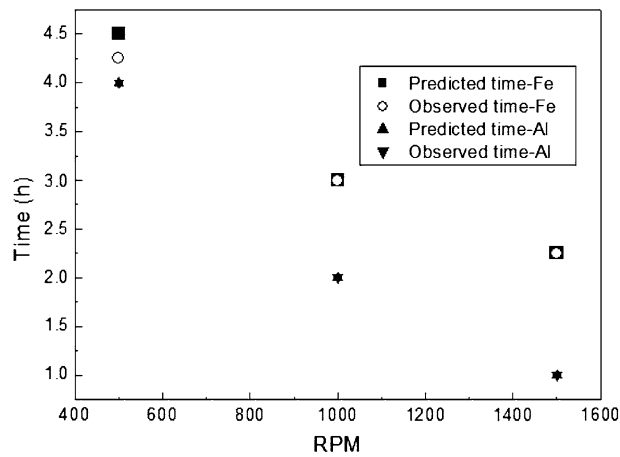


Fig. 4—Plot of predicted milling times and observed milling times to attain crystallite sizes of 25 and 46 nm, respectively, for iron and aluminum at different rpm.

Therefore, given a particular grain size, the time taken to achieve it depends solely on the ΔT . For a given mass flow rate of the cooling water (m), the ΔT is a function of rpm alone. At each rpm, within a short period, a steady state is reached and cooling water gets heated to a particular level; thus, ΔT does not change with time of milling. Thus, to predict the time required to reach a particular grain size, one should first do a milling experiment on a material for a particular period at a particular rpm and measure the ΔT and the crystallite size corresponding to this condition. Now for the same material, one can do experiments at other speeds for a short period until the steady-state condition is reached wherein ΔT reaches a constant value corresponding to that rpm. Such experiments have been carried out for iron and aluminum at 500, 1000, and 1500 rpm, and the values of ΔT have been measured (Table II).

The XRD pattern of iron milled for 3 hours at 1000 rpm is shown in Figure 3. The crystallite size for this sample was calculated from the peak broadening (after deducting the instrumental broadening and lattice strain) as 25 nm. From the ΔT measurements at other speeds, the time to attain the same grain size was predicted as 4 hours 30 minutes and 2 hours 15 minutes at 500 and 1500 rpm, respectively. Experimentally, the same grain size was achieved at 4 hours 15 minutes and 2 hours 15 minutes at 500 rpm and 1500 rpm, respectively, which matches our predictions closely as shown in Figure 4. Aluminum was milled for 2 hours at 1000 rpm and the crystallite size achieved was 46 nm. The time predicted and taken were 4 hours and 1 hour for 500 and 1500 rpm, respectively. Thus, our predictions exactly matched in the case of aluminum as shown in Figure 4. Small differences exist between the predicted and observed time for the iron system and aluminum system. This may be attributed to difference in material properties like crystal

structure, ductilities and thermal diffusivities and some experimental error.

V. CONCLUSIONS

A model has been developed for estimating the energy transferred to the powder during milling in simoloyer. The model can be used to predict the time taken to achieve a given grain size for any particular milling speed once the crystallite size is measured for a sample milled at any one milling speed. A disadvantage of the model is that one cannot predict the temperature difference before hand and thus the mill must be run for a small time to measure the ΔT . This model is a step toward analyzing the milling mechanics of simoloyer and can be further enhanced to do the following:

1. Incorporate material properties enabling it to predict properties like strain energy
2. Predict grain size as a function of time
3. Predict ΔT before carrying out an experiment

REFERENCES

1. J.S. Benjamin: *Metall. Trans.*, 1970, vol. 1, pp. 2943–51.
2. B.S. Murty and S. Ranganathan: *Int. Mater. Rev.*, 1998, vol. 43, pp. 101–41.
3. C. Suryanarayana: *Progr. Mater. Sci.*, 2001, vol. 46, pp. 1–184.
4. J.R. Harris, J.A.D. Wattis, and J.V. Wood: *Acta Mater.*, 2001, vol. 49, pp. 3991–4003.
5. M. Abdellaoui and E. Gaffet: *Acta Metall. Mater.*, 1995, vol. 43, pp. 1087–98.
6. B.S. Murty, M. Mohan Rao, and S. Ranganathan: *Acta Metall. Mater.*, 1995, vol. 43, pp. 2443–50.
7. J. Joardar, S.K. Pabi, and B.S. Murty: *J. Alloys Compd.*, 2007, vol. 429, pp. 204–10.
8. P.P. Chattopadhyay, I. Manna, S. Talapatra, and S.K. Pabi: *Mater. Chem. Phys.*, 2001, vol. 68, pp. 85–94.
9. M.P. Dallimore and P.G. McCormick: *Metal Powder Report*, 1998, vol. 53 (1), p. 37.
10. F.P. Zhong, W.X. Hong, Q.Y. Huai, and Q.U. Xuan-Hui: *J. China Univ. Mining Technol.*, 2008, vol. 18, pp. 449–53.
11. D. Basset, P. Matteazzi, and F. Miani: *Mater. Sci. Eng. A*, 1994, vol. 174, pp. 71–74.
12. A. Concas, N. Lai, M. Pisu, and G. Cao: *Chem. Eng. Sci.*, 2006, vol. 61, pp. 3746–60.
13. T.S. Ward, W. Chen, M. Schoenitz, R.N. Dave, and E.L. Dreizin: *Acta Mater.*, 2005, vol. 53, pp. 2909–18.
14. D.R. Maurice and T.H. Courtney: *Metall. Mater. Trans. A*, 1994, vol. 25A, pp. 147–58.
15. M. Magini and A. Iasonna: *Mater. Trans. JIM*, 1995, vol. 36, pp. 123–33.
16. J. Joardar, S.K. Pabi, and B.S. Murty: *Scripta Mater.*, 2004, vol. 50, pp. 1199–1202.
17. C. Sasikumar, S. Srikanth, N.K. Mukhopadhyay, and S.P. Mehrotra: *Miner. Eng.*, 2009, vol. 22, pp. 572–74.
18. http://www.zoz-group.de/zoz.engl/zoz.main/pdf_content/publications/v07.pdf.
19. http://www.zoz-group.de/zoz.engl/zoz.main/pdf_content/publications/v14.pdf.
20. http://www.zoz-group.de/zoz.engl/zoz.main/pdf_content/publications/v18.pdf.