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EXPLOSION AREAS OF FLAMMABLE SUBSTANCES AND THEIR NUMERICAL APPROXIMATION

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CHEMSAFE® database [1], developed as a joint project between BAM, PTB and DECHEMA, contains safety characteristics of flammable liquids, gases, dusts and their mixtures. The BAM is responsible for evaluated data on gases and dusts. TRIANGLE software, originally developed by the BAM for the processing, quality assurance and visualisation of measured data describing the explosion area of ternary systems consisting of flammable, inert and oxidising gases. 2D figures in the program represent isothermal or isobaric level curves of the explosion areas. In order to better describe the limiting curve of the explosion range, by the BUTE (Budapest University of Technolcgy and Economics) the Akima-spline interpolation has been replaced by special comonoton Splines in TRIANGLE. For the 3D visualisation of the whole temperature- or pressure-dependent ternary explosion surfaces the TRIGON program of BUTE was extended.

Keywords: ternary flammable systems, inerting, explosion areas, spline curves and surfaces, 2D and 3D visualisation

Introduction

In order to avoid explosion with flammable substances, generally applied solution in industrial processes is purging with inert gases. Reliable measured data are required concerning the explosion range of ternary systems. The CHEMSAFE® database, which is available world wide through STN International and Internet, contains rated safety characteristics of flammable liquids, gases, dusts and their mixtures, such as explosion limits, flash points, ignition temperatures, etc. The in-house version of CHEMSAFE allows a graphical representation of the measured explosion range in triangular diagrams as a function of the concentration of flammable, oxidising or inert gases. In the data processing phase a flexible software procedure is needed to solve 2D and in certain cases also 3D visualisation for the explosion area of ternary gas mixtures. The TRIANGLE program, created by BAM and extended by the common research group of BUTE and BAM, provides the 2D triangular diagrams. When the influence of additional parameters is to be shown, for 3D explosion surfaces the extended TRIGON software offers a good solution.

Characteristics of the explosion range

Explosion areas need to be explicitly defined by unambiguous characteristic parameters. *Table 1* and *Fig.1* show the acronyms applied in CHEMSAFE.

As an example, let us consider the IAR line in *Fig.1*. This line represents a limit in the ternary flammable system: all mixture compositions existing on the right hand side of this line will not cause an explosion regardless of the added amount of flammable gas.

The interception of the IAR line and the oxidising gas axis determines the MAI point, the minimum required inert gas content of the binary inert-oxidising gas system which is necessary to avoid an explosion. Similarly, the ICR line gives a limiting ratio between

Table 1 Characteristic values for explosion area of ternary systems containing flammable gas / inert gas / oxidising gas

Acronym in CHEMSAFE	Description
LEL	Lower Explosion Limit
UEL	Upper Explosion Limit
ICR	Minimum Inert gas / Combustible Ratio
IAR	Minimum Inert gas / AIR (Oxidising gas) Ratio
MXC	MaXimum permissible Amount of Combustible
MAI	Minimum required Amount of Inert gas
MOC or LOC	Maximum Oxygen (Oxidising gas) Content or Limiting Oxygen Concentration

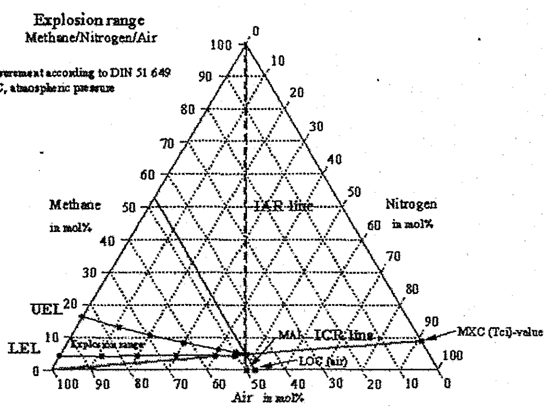

Fig.1 Representation of the explosion area

inert and flammable gas at which by adding any amount of flammable gas to the system no explosion will occur.

The TRIANGLE program is used for processing measured values of such systems. The boundary curve around the explosion area is generated by applying numerical interpolation on the measured data. After the evaluation of the characteristic values of the system the program returns the results in tables and in ternary diagrams both in Cartesian and in triangle co-ordinates.

2D approximation problems solved by comonoton parametric vector splines – Additions and extensions in TRIANGLE

For the characterisation of the explosion range well determined tangent lines of the explosion areas are needed, such as the IAR or ICR lines. For these to be accurate we have to apply the best possible numerical approximation of the 2D explosion limit curves. Comonoton parametric vector splines possess the best numerical properties for this task [2, 5]. The conditions of comonotonity for the set of measured points $\{x_i; y_i\}_{i=1}^{n}$ are given as following [4, 6]:

Let be $(m_i)_x = \frac{x_i - x_{i-1}}{s_i - s_{i-1}}$ and $(m_i)_y = \frac{y_i - y_{i-1}}{s_i - s_{i-1}}$, (1)

Table 2 Comparison of different interpolation methods Butene/Nitrogen/Air-System (T=297 K; P=0,1 MPa)

	Traditional Spline	Comonoton Spline	Linear Interpolation
MAI	51,83	51,78	51,78
MOC	9,94	9,96	9,98
MXC	2,68	2,71	2,72
IAR	1,08	1,07	1,07
ICR	36,35	35,93	35,71

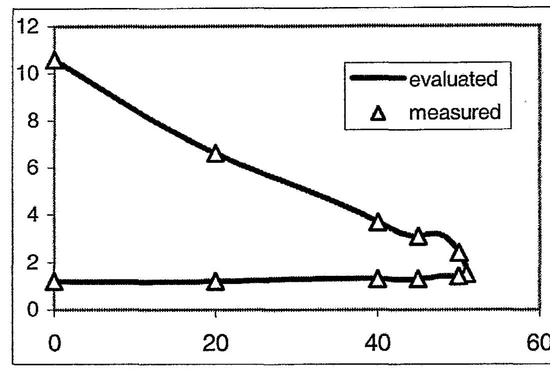

Fig.2 Butene/Nitrogen/Air-System (T=297 K; P=0,1 MPa) Traditional Spline interpolation

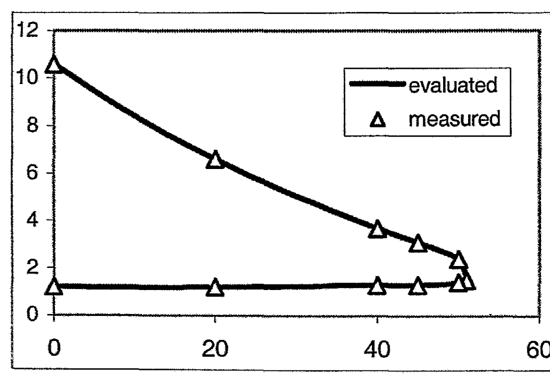

Fig.3 Butene/Nitrogen/Air-System (T=297 K; P=0,1 MPa) Comonoton Spline interpolation

where s denotes the usual arc length parameter.
Then it holds for the slope coordinates $x'(s)$ and $y'(s)$:

$$\forall\ s \in (s_{i-1}; s_i): \begin{array}{l} x'(s)\cdot(m_i)_x > 0 \ \text{ if } \ (m_i)_x \neq 0 \\ y'(s)\cdot(m_i)_y > 0 \ \text{ if } \ (m_i)_y \neq 0 \end{array} \qquad (2.a)$$

$$\forall\ s \in [s_{i-1}; s_i]: \begin{array}{l} x'(s) = 0 \ \text{ if } \ (m_i)_x = 0 \\ y'(s) = 0 \ \text{ if } \ (m_i)_y = 0 \end{array} \qquad (2.b)$$

Using the conditions above, we created subroutines for the comonoton parametric vector splines and built them in the TRIANGLE program. Similarly, we also created subroutines for the traditional parametric vector splines [9, 10] and compared the differently evaluated characteristics.

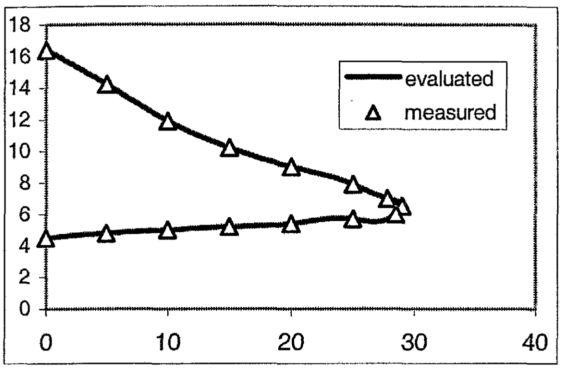

Fig.4 Methane/CO_2/Air-System (T=297 K; P=0,1 MPa)
Traditional Spline interpolation

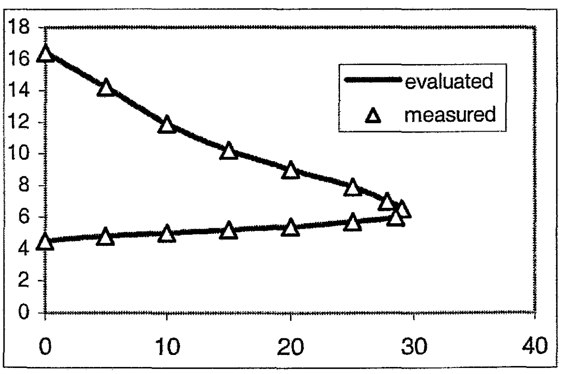

Fig.5 Methane/CO_2/Air-System (T=297 K; P=0,1 MPa)
Comonoton Spline interpolation

Some representative results are shown in *Figs.2* and *3* and in *Table* 2. for the Butene/Nitrogen/Air ternary system, and in *Figs.4*, *5*, and in *Table 3* for the Methane/CO_2/Air ternary system.

In the case of the Butene/Nitrogen/Air-system the traditional Spline interpolation produces an undesired curvature on the UEL curve, near the apex of the explosion area. It has a slight effect on the MAI, MXC, and ICR values - as it is shown in *Table 2*.

In the second example, in the case of the Methane/CO_2/Air-system with the traditional Spline interpolation, the undesired curvature appears on the LEL curve, again, near the apex of the explosion area. In this case the curvature causes a considerable error in the MXC point and in the slope of the ICR line. By using comonoton vector splines both above mentioned errors can be avoided.

It needs to be mentioned that traditional spline approximation had been found to produce similar errors in five out of the about twenty investigated systems.

On the programming side the new parametric vector splines required changes in 3 of the 4 worksheets, 3 of the 7 diagrams, 8 of the 11 modules and 3 of the 5 forms of the original TRIANGLE (Visual Basic for Excel) program.

Table 3 Comparison of different interpolation methods
Methane/CO_2/Air-System (T=297 K; P=0,1 MPa)

	Traditional Spline	Comonoton Spline	Linear Interpolation
MAI	31,02	31,02	31,02
MOC	13,52	13,54	13,55
MXC	**16,95**	**17,39**	17,39
IAR	0,45	0,45	0,45
ICR	**4,9**	**4,75**	4,75

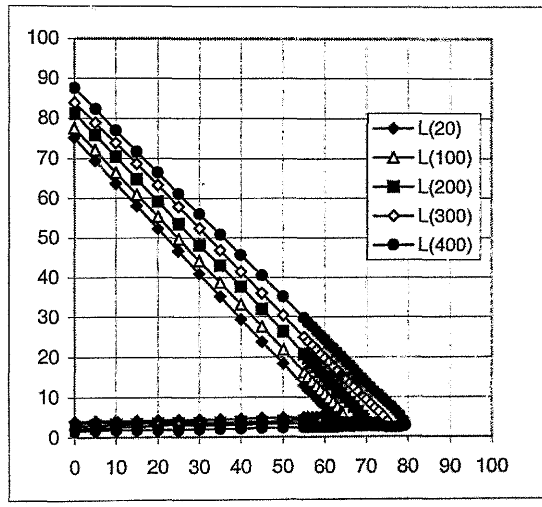

Fig.6 Isothermal level curves of H_2/CO_2/Air

3D visualisation– examples of the extended TRIGON application

The TRIGON program was created for visualisation of triangular (ternary) vapour-liquid equilibrium (VLE) surfaces [7, 11]. As we recognized the interesting possibility to visualize the temperature or pressure dependence in the explosion areas of ternary systems containing flammable gas, we tried to utilize TRIGON and its embedded Bézier surfaces. Although our minimal energy Bézier surfaces have many useful properties [2, 6-8], we could not directly apply them for the Ternary Explosion Surface (TES) approximation, because their computational algorithm needs the whole triangle as domain for the surfaces. A similar problem occurred with the application of Shepard's metric interpolation [3].

After investigating the above problems, we decided to use the 2D comonoton spline level curves combined by bilinear interpolation. The comonoton spline (isothermal) level curves of Hydrogen/Carbon dioxide/Air-system are shown in *Fig.6* as an example. The five data sets belonging to the temperatures 20, 100, 200, 300 and 400 C are from the CHEMSAFE® database. The bilinear interpolation allowed a simple restriction of the whole triangle domain, using the inner and the outer level curves

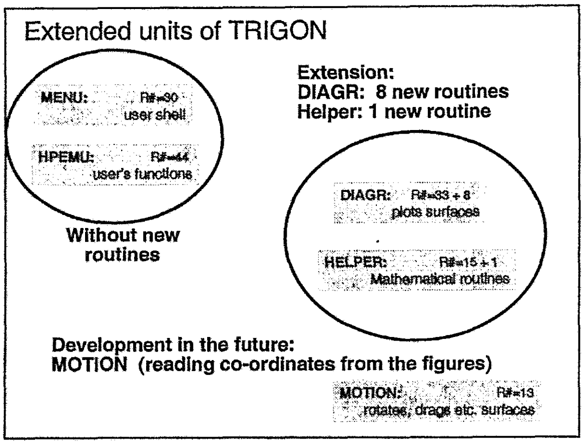

Fig.7 Extensions necessary for TRIGON visualizing TES

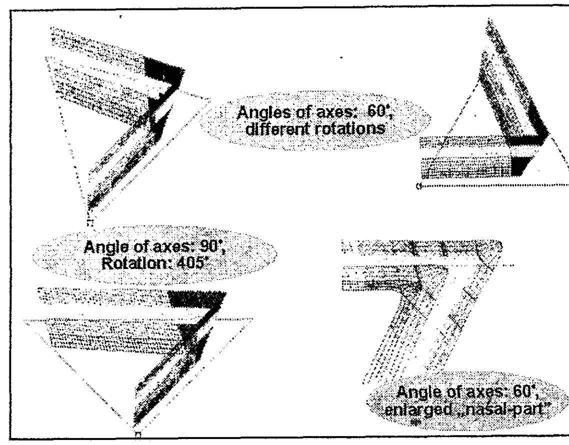

Fig.8 3D Explosion areas of Hydrogen/Carbon dioxide/Air-System given by isothermal level curves, plotted by TES TRIGON

(L(20) and L(400) in *Fig.6*) as boundaries in the restricting inequalities.

Parametric comonoton vector splines were used for the (isothermal or isobaric) level curves and a bilinear interpolation was used for the trapezoid inner part between the level curves. To accomplish this, we had to extend the VLE TRIGON software according to *Fig.7*. The TES TRIGON program version contains the MENU and HPEMU units [11] unchanged, but in the DIAGR unit we had to develop 8 new routines, and one in the HELPER unit.

The output of the TES-TRIGON software for two example ternary systems (Hydrogen / Carbon dioxide / Air-system defined by isothermal level curves and Ethene / Nitrogen / Air-system defined by isobaric level curves) are shown in *Figs.8* and *9*.

In the approximation of the apex of explosion surfaces we had to solve a "apex accumulation" problem. As it is shown in *Fig.8*, in the first solution we slightly broke the "level curves condition". Even though in the real explosion of surface plots this is unnoticeable, we continue to work on a solution that does not break the condition of the level curve system.

The different rotation and view angle changing features of TRIGON show the properties of the ternary explosion areas very well. In the case of the Ethene/Nitrogen/Air-system, given by isobaric level curves, in the *Fig.9*, the lower pressure part between 0.1 and 1 MPa is interesting, because the dangerous part is wider than in higher pressure, between 1 and 10 MPa.

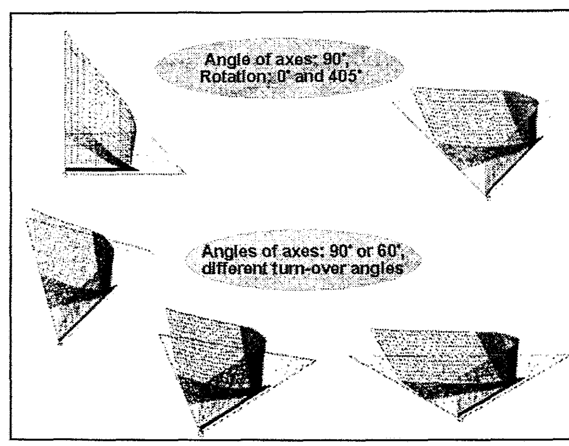

Fig.9 3D Explosion areas of Ethene/Nitrogen/Air-System given by isobaric level curves, plotted by TES TRIGON

Conclusions

For correct 2D visualisation of isothermal or isobaric explosion areas of ternary gas mixtures containing flammable gas, we extended the TRIANGLE software (created by BAM, Berlin). To reach the desired accuracy for the approximation of explosion characteristics we used parametric comonoton vector Splines. The software and the method had already been tested on many systems in chemical safety engineering.

In some cases, when 3D visualisation is necessary, the extended TES TRIGON software (created by BUTE, Budapest) gives good results. In this program the base of the numerical surface approximation was comonoton vector Spline, the same as in the 2D visualisation. Between the isobaric or isothermal explosion level curves we used bilinear interpolation that made the necessary domain restriction also easy to implement.

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SYMBOLS

IAR	Minimum Inert gas / Air (Oxidising gas) ratio
ICR	Minimum Inert gas / Combustible Ratio
LEL	Lower Explosion Limit
L(T)	Isothermal level curve of TES
MAI	Minimum required Amount of Inert gas
MOC	Maximum Oxidising gas Content
MXC	MaXimum permissible Amount of Combustible gas
P	Pressure, MPa
S	Arc length parameter
T	Temperature, K or C
TES	Ternary Explosion Surface
UEL	Upper Explosion Limit
(x,y)	Cartesian co-ordinates
VLE	Vapour Liquid Equilibrium

REFERENCES

1. CHEMSAFE®-Database For Safety Characteristics, BAM, Berlin, PTB, Braunschweig, DECHEMA, Frankfurt am Main, v1.4, 2001
2. FASSHAUER G. E. and SCHUMAKER L. L.: Computer Aided Geometric Design, *1996*, 13, 45
3. GORDON W. J. and WIXOM J. A.: Math. and Comput., *1998*, 32, 253
4. GRONEWOLD G.: Hung. J. Ind. Chem., *1996*, 25, 59
5. HERRMANN N.: Hung. J. Ind. Chem., *1996*, 25, 263
6. KOLLÁR-HUNEK K., LÁNG-LÁZI M., HERRMANN N., MIKLÓS D. and KOVÁCS I.: Hung. J. Ind. Chem., *1998*, 26, 269
7. KOLLÁR-HUNEK K., VICZIÁN ZS. and LÁNG-LÁZI M.: Hung. J. Ind. Chem., *1999*, 27, 6
8. LÁNG-LÁZI M., DIÓSPATONYI I., VICZIÁN G. and HESZBERGER J.: Hung. J. Ind. Chem., *1999*, 27, 317
9. SHIKIN E. V. and PLIS A. I.: Handbook on Splines for the User; CRC Press, Boca Raton, FL, USA, 1995
10. SPÄTH H.: Algorithmen zur Konstriktion glätter Kurven und Flächen, Oldenburg, München, Wien, 1983
11. VICZIÁN G., LÁNG-LÁZI M., HESZBERGER, J., DIÓSPATONYI I. and KOLLÁR-HUNEK K.: Hung J. Ind. Chem., *2000*, 28, 311