Supporting information

1. Chemicals

Table 1. Chemicals.

Chemicals	Supplier	Purity
$Sr(NO_3)_2$	Grüssing GmbH	98 %
$LiNO_3$	Sigma Aldrich	Reagent Plus
Mg 50/100 mesh	Sigma Aldrich	Reagent Grade 98 %
Mg	Grüssing GmbH	97 %
5-Amino-1H-tetrazole	abcr	98%
Hexamine	Acros organics	99 %
Epon 813	Hexion	-
Versamid 140	Miller-Stephenson	-
Nitrocellulose	Sigma Aldrich	Collodion solution (4 - 8 % ethanol/diethylether)

2. Grain size/particle size of formulations and Mg

We discovered that the grain size after grinding was mainly influenced by the applied magnesium. The supplier of Mg (Grüssing GmbH, 99%) reported the grain size in the range of 60 – 300 µm. This Mg was applied in formulations **2** – **8**. However, manually sieving revealed that even smaller grain sizes were present. For this reason we quantitatively determined the grain size distribution of a 10g Mg sample applying different sieves. In addition, we used REM measurements (Figure 1, 2) to determine the particle shape of the applied Mg. In formulation **1,** Mg (Sigma Aldrich, Reagent Grade 98%, 50/100 mesh) and a binder ratio of Epon 813/Versamid 140 (4:1) were applied.

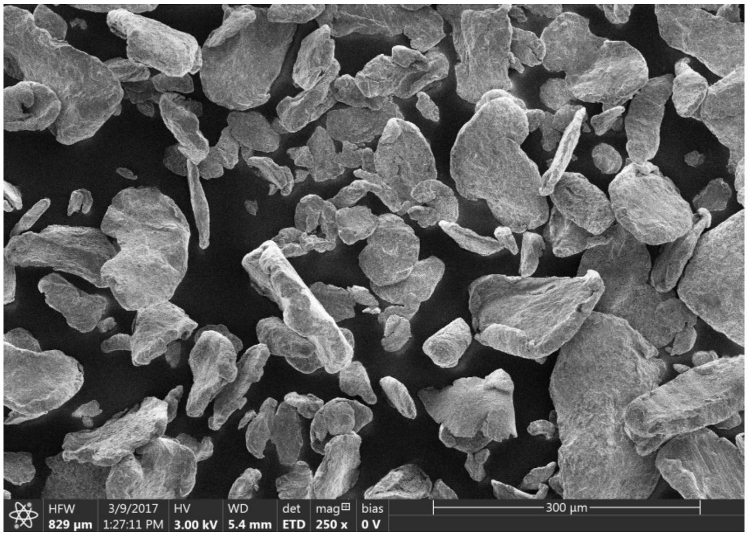

Figure 1. SEM of Mg (Grüssing GmbH, 99%).

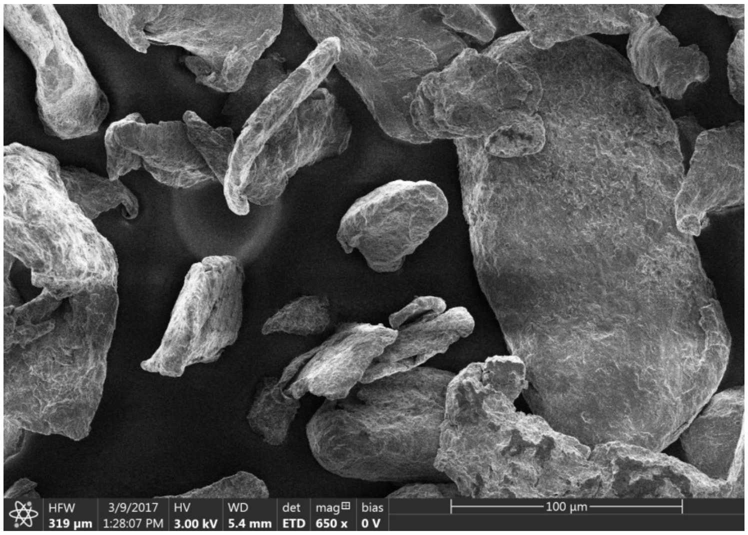

Figure 2. SEM_2 of Mg (Grüssing GmbH, 99%).

3. Sample preparation

The samples were weighed out according to their weight percentages (max. 1 g total) into a mortar. After grinding by hand for 3 min, both the Epon 813 (20 mg/mL) and the Versamid 140 solution (10 mg/mL) in ethyl acetate were added using a syringe. For nitrocellulose, we applied Colloidon solution (4-8 % in ethanol/diethylether) to the pre-grinded solid materials. The mixture was blended with a spatula every 10 min until the solvent was evaporated. The solid material was stored over night at 70 °C in the drying oven for curing. Before consolidation, the pyrotechnic material was grinded again by hand for 3 min. The formulations were pressed with the aid of a tooling die (inner diameter 12.9 mm) into a cylindrical shape. The formulation powders were pressed at a consolidation dead load of 2 t with a dwell time of 3 s. A torch was used to ignite the pellets.

4. Optical emission spectroscopy

Dominant wavelength/spectral purity/luminous intensity:

Optical emissive properties were characterized using both a Ocean optics HR2000+ES spectrometer with an ILX511B linear silicon CCD-array detector (range 190 – 1100 nm) and included software/calibration files from Ocean Optics. Spectra were recorded with a detector-sample distance of 1.00 m. The acquisition time for the flare formulations was 20 ms/scan. The dominant wavelength (DW) and spectral purity (SP) were measured based on the 1931 CIE method using illuminant C as the white reference point. Four samples were measured for each formulation and all given values are averaged based on the full burn of the mixture. The controlled burn was filmed with a digital video camera recorder (SONY, DCR-HC37E).

Frequency measurement:

Frequency measurements were performed using an acquisition time of 5 ms/scan or 1 ms/scan. Dominant wavelengths were measured based on the 1931 CIE method using illuminant C as the white reference point. The range selection for recording a signal was set the range from 580.11 nm – 680.14 nm covering all previously recorded dominant wavelengths as well as the red light part in the visible spectrum. Calculation of the frequency was done by detecting the single peaks applying the Origin Pro 9.0 software and calculating the period of time between the single peaks (= flash reactions).

5. High-speed camera

Setup:

Highspeed video recording was performed applying the “SpeedCam Visario G2 1500” by Weinberger AG with the accompanied “Visart 2.2” software package. Following data is taken from the supplier data sheet:

Sensor: High-Speed APS-CMOS Sensor
Active Sensor area: 16.89 x 11.26 mm (W x H)
Active pixel size: 11 µm Square

Image formats: 768 x 512 pixel: up to 4000 frames per seconds

Shutter: Electronic shutter down to 10 µsec.

Dynamic range: color depth to 30 Bit

The applied camera lens was purchased from Sigma (24-70 mm, 2.8 EX DG, Macro, diameter 82 mm). In addition, a Heliopan Filter UV SH-PMC 82mm was applied.

High-speed video recording procedure:

The bare pellet was placed in the fume hood and ignited by applying a torch. No prime composition was applied. The distance between the camera and the pellet (same height) was approximately 1 m. Due to the camera settings and applied lense/filter, additional halogen lamps to illuminate the fume hood were applied. A sequence of several seconds was recorded for selected formulations.

6. Emission spectra of formulations **2**, **6** and **8**.

Li emits two sharp lines in the red region at 671 nm and 610 nm with decreasing intensity.[1, 2] Unfortunately, the human eye´s capability to detect light is limited by the so-called cone cells. These 3 different types of photoreceptor cells show their own distinctive sensitivities to different wavelengths. The overlap of the single cones results in the human eye´s response to light.[3] The highest sensitivity to detect light also referred to as the peak of photopic response is at 555 nm (green region, 100 %). In the case of higher wavelengths, the sensitivity decreases below 10 % at 670 nm.[3] As a consequence, the emission line at 610 nm (approximately 50 %) is far better detected by the human eyes though being less intense and contributing to the red light performance.[3] A look at the emission spectra of formulation **2** reveals a strong background emission (incandescence produced by condensed MgO, LiO, LiOH and Li_2O). As a result, the spectral purity drops and the dominant wavelength is given as 563 nm. Due to illuminant C as the white reference point, the contribution of the green emission (part of background emission) is weighted higher compared to red light and results in a calculated dominant wavelength (averaged over the entire burning for each single scan) of 563 nm. Formulation **8** (Figure 4) showed less background radiation and results in a higher spectral purity as well as dominant wavelength.

Literature:

1. T. M. Klapötke, *Chemistry of High-Energy Materials*, De Gruyter, Berlin, Boston, **2017**.
2. H. A. Webster III, in *MWSC/CR/RDTR-16*, Naval Weapons Support Center, Crane, Indiana, **1975**.
3. S. J. Robinson and J. T. Schmidt, *Mater. Eval.*, **1984**, 42, 1029-1034.

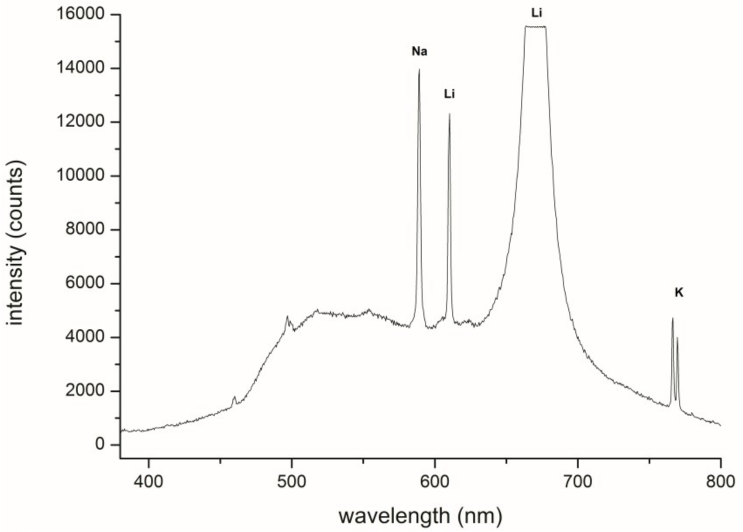

Figure 3. Burning of formulation 2.

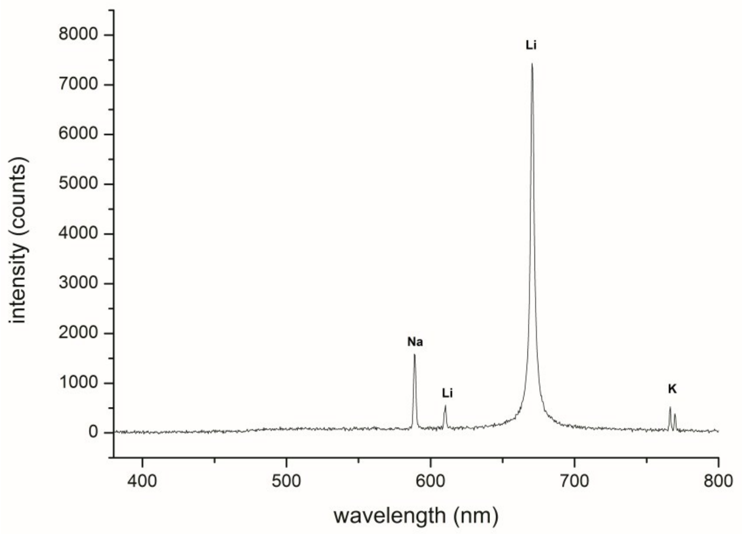

Figure 4. Constant burning of formulation 8.

Figure 5 shows a recorded sequence of formulation **6** (Figure 5). Starting from the second recorded spectrum (dark reaction, t = 0 ms), a spectrum was recorded every 1 ms for 34 ms in total. As can be, the background radiation increases with increased intensity of the formulation (= highest value for Li emission at 670 nm). No unidentified light-emitting species was detected in the recorded spectra.

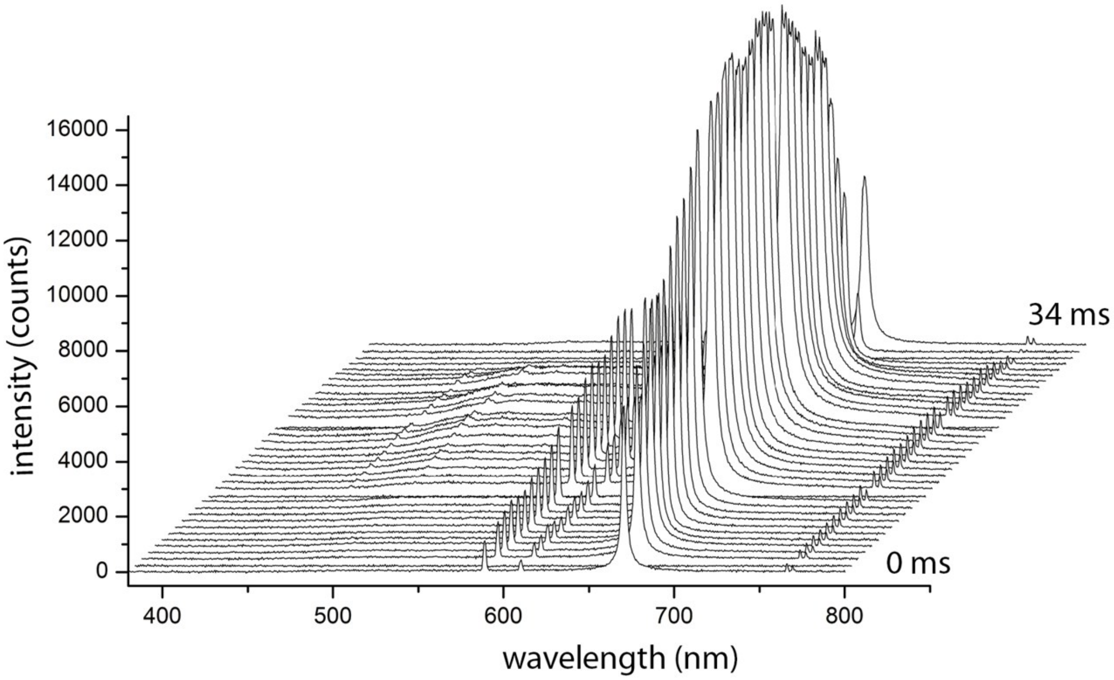

Figure 5. Time dependent high-speed measurement of formulation 6 (1 ms/scan).

7. Frequency spectra

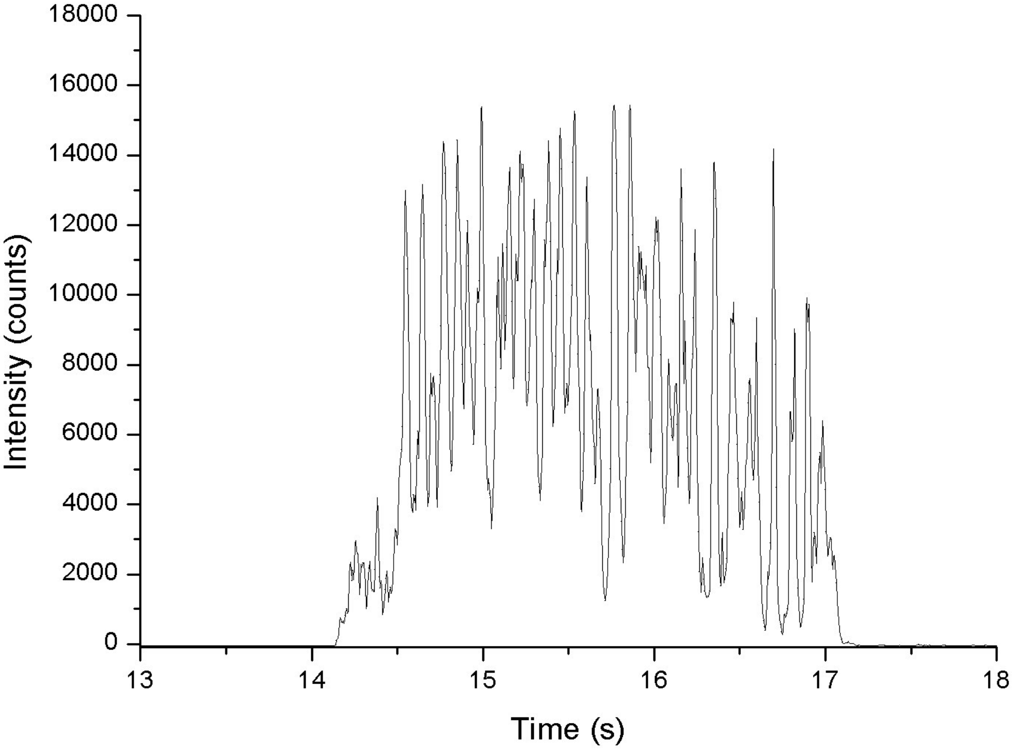

Figure 6. Frequency measurement of formulation 3 with 5 ms/scan.

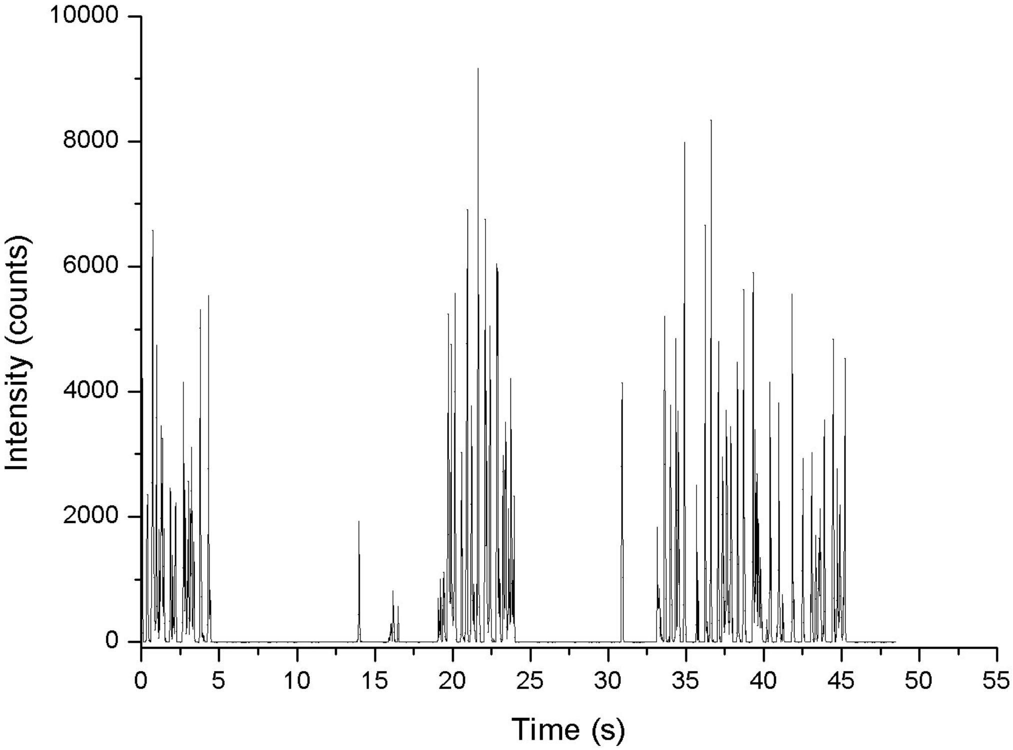

Figure 7. Frequency measurement of formulation 4 with 5 ms/scan.

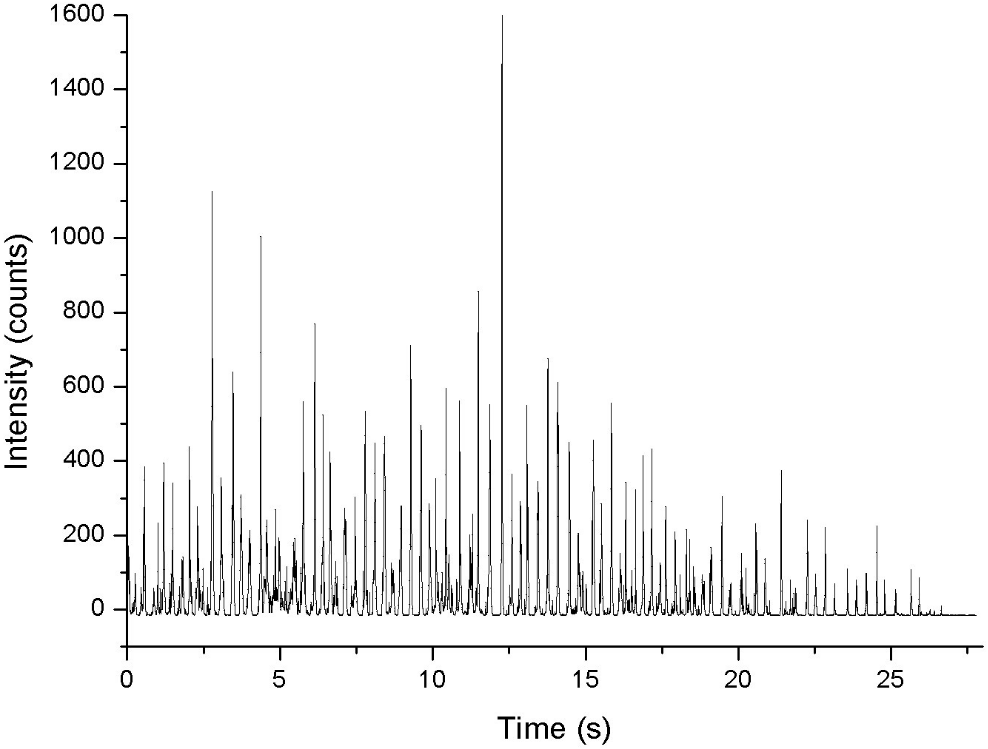

Figure 8. Frequency measurement of formulation 5 with 5 ms/scan.

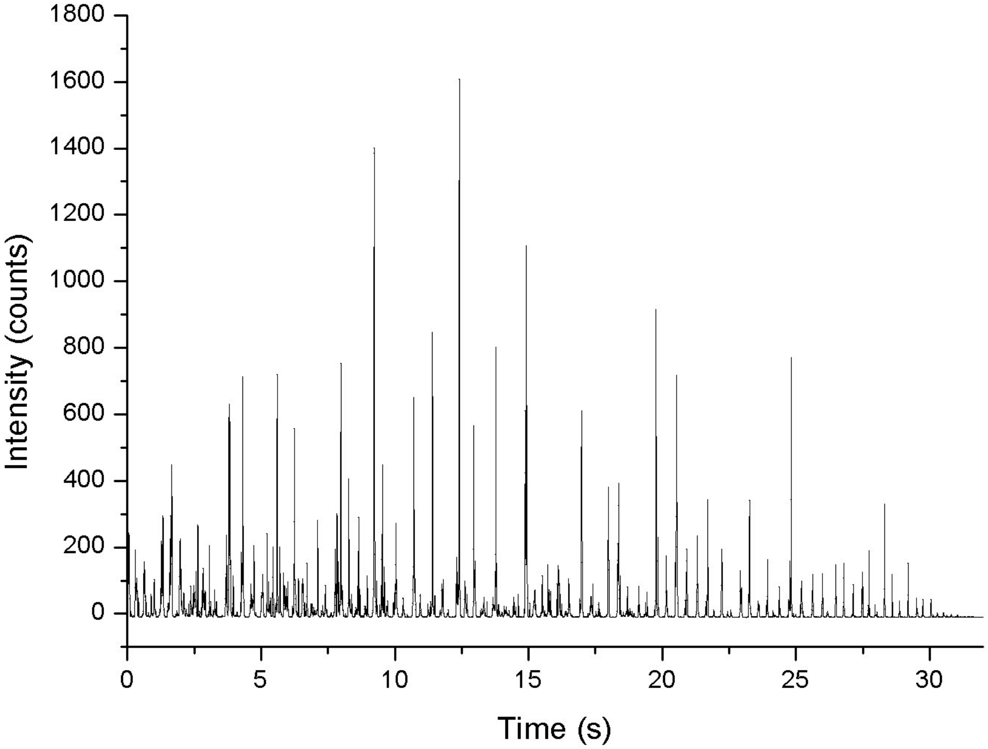

Figure 9. Frequency measurement of 6 with 5 ms/scan.

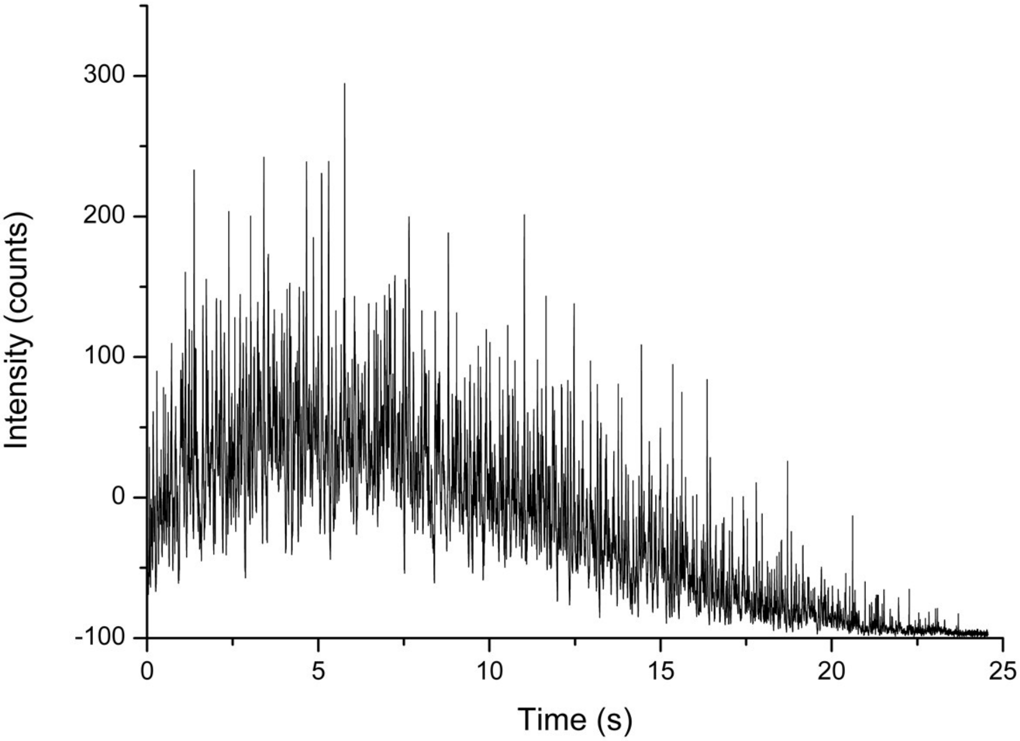

Figure 10. Frequency measurement of 7 with 1 ms/scan.

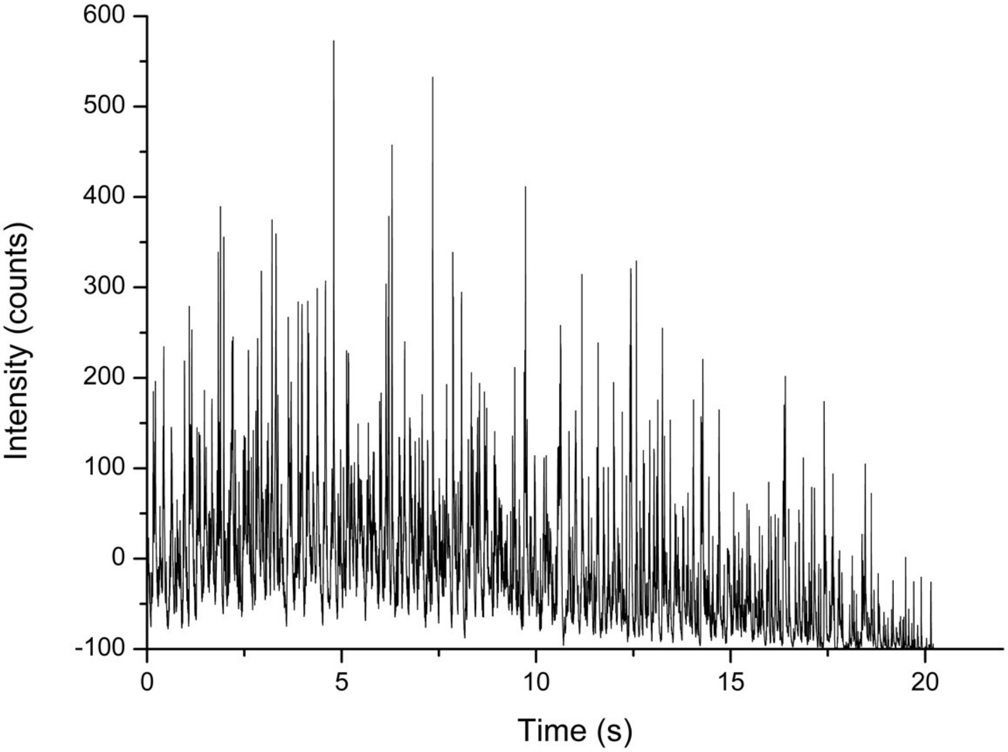

Figure 11. Frequency of 8 with 1 ms/scan.

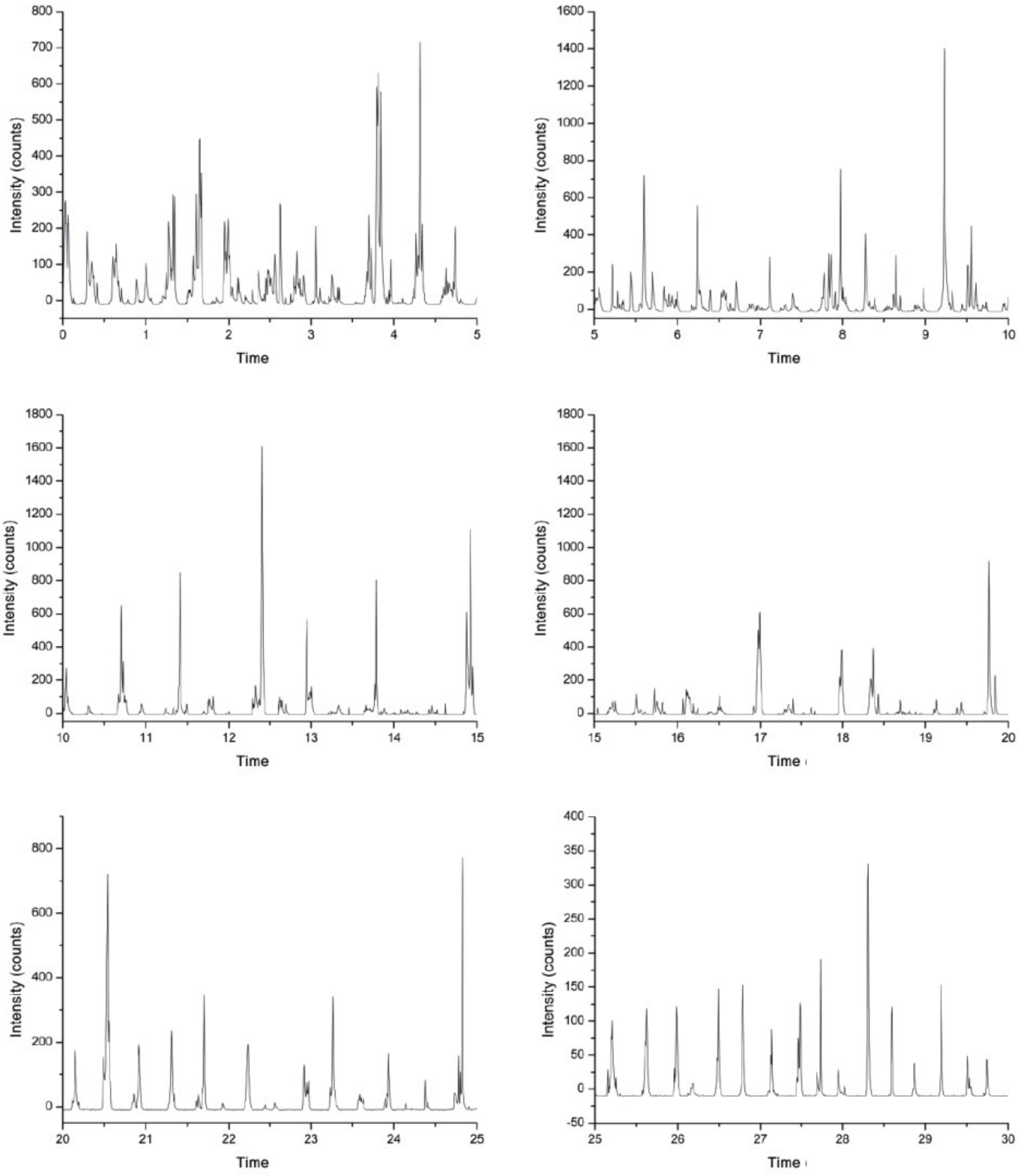

Figure 12. Frequency measurement of 6 in 5 s intervalls (1ms/scan).