OCTA

Integrated simulation system for soft materials

RHEOLOGY SIMULATOR

PASTA

Additional Manual for ver.2.9

OCTA User's Group

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Contents

Chapter 1

New Features in PASTA 2.9

This manual describes a new functionality in PASTA 2.9 (calculating the stress relaxation modulus by the Green-Kubo formula) and related new action and tool.

1.1 Theoretical Background

In PASTA ver.2.8 or earlier, the stress relaxation modulus $G(t)$ can be calculated by studying a response of the stress $\sigma(t)$ to a step deformation with a finite but small strain γ. In order for this method to give good statistics, however, the strain γ should not be too small (typically $\gamma \simeq 0.5$ is required), and a small nonlinearity remains in the calculated modulus $G(t) = \sigma(t)/\gamma$.

Another method of calculating $G(t)$, which is strictly linear, is to use the linear response theory, which tells that $G(t)$ can be obtained from the stress auto-correlation function in *equilibrium* (the Green-Kubo formula):

$$G(t) = \frac{V}{k_B T} \left\langle \sigma_{xy}(t)\sigma_{xy}(0) \right\rangle . \tag{1.1}$$

Here, V is the volume of the system, k_B is the Boltzmann constant, and T is the temperature. σ_{xy} is the xy component of the stress tensor, but other stress components can be equally used; in an isotropic system, we can average over five independent stress components by

$$G(t) = \frac{V}{10 k_B T} \left\langle \mathrm{Tr}\left[\sigma'(t)\sigma'(0)\right] \right\rangle , \tag{1.2}$$

where σ' is the traceless part of the stress tensor: $\sigma'_{\alpha\beta} \equiv \sigma_{\alpha\beta} - (1/3)\delta_{\alpha\beta}\mathrm{Tr}\sigma$.

It has been pointed out by ref.[1] that, when calculating the auto-correlation function, it is important to use the stress $\sigma(t)$ at *every* time step to average out the high-frequency and large-amplitude fluctuation in $\sigma(t)$. PASTA-2.9 has implemented, therefore, a "correlator" [2] which calculates the correlation function by using $\sigma(t)$ at *every* time step as the simulation proceeds (the so-called "on-the-fly" calculation).

1.2 New data in input and output UDF

In input UDF, `Simulation.Deformations[].FlowType` can take two new values `gt_start` and `gt_stop`. `gt_start` tells PASTA to turn on the correlator. Actual calculation of the correlation function is done in the next `Deformation` with `FlowType = noflow`. `gt_stop` turns off the correlator. If `FlowType` is either `gt_start` or `gt_stop`, other elements of `Deformation` need not be specified.

In output UDF, two new data `RelaxationModuls` and `SSCorrelator` have been added. If $G(t)$ is calculated by the correlator method, the result is saved in `RelaxationModuls` in the final record of the output UDF. Also in the final record, information for restarting the correlator is saved in `SSCorrelator`.

Input UDF for PASTA-2.8 or earlier can be used as an input for PASTA-2.9. It is also possible to restart PASTA-2.9 from the output UDF of PASTA-2.8 (or earlier).

1.3 New action

A new action `plot` has been added to the data `RelaxationModulus` in the output UDF. It creates a log-log plot of $G(t)$, and also output a table of $G(t)$ in the `Log` area; you can copy/paste this table into a text file.

Other actions in the output UDF, `plot_Stress` and `plot_Viscosity_or_RelaxationModulus` have also been rewritten, and may give slightly different plot from the previous version.

1.4 Example

An example input UDF for calculating $G(t)$ of a monodisperse (Z=20) system by the correlator method can be found in `PASTA/PASTA/sample/sscorr_in.udf`. Open this UDF by GOURMET, and open the array `Simulation.Deformations[]`; it has three elements:

- `Deformation[0]` with `FlowType = noflow`, for the thermalization
- `Deformation[1]` with `FlowType = gt_start`, to turn on the correlator
- `Deformation[2]` with `FlowType = noflow`, for the calculation of $G(t)$

The simulation can be run as usual (in `gourmetterm`):

```
$ pasta -I sscorr_in.udf -O sscorr_out.udf
```

Open the output UDF `sscorr_out.udf` (it is prepared in `PASTA/PASTA/sample/out/`) by GOURMET, and open `RelaxationModulus` in the final record to find the data of $G(t)$.

Right-click on the `RelaxationModulus` (need not be in the final record) and run the action `plot`; this will create a log-log plot of $G(t)$, and also output a table of $G(t)$ in the `Log` area. Copy/paste the table into a text file ("gt.txt", for example).

The complex modulus can be calculated by (again in `gourmetterm`):

```
$ corr2gw < gt.txt > gw.txt
```

`gw.txt` contains the real and imaginary parts of the complex modulus $G^*(\omega)$ and the complex viscosity $\eta^*(\omega) = G^*(\omega)/i\omega$.

If you want to improve the accuracy of $G(t)$, you can restart from `sscorr_out.udf` and continue the calculation; the information necessary to restart the correlator is saved in the last record of `sscorr_out.udf` and automatically reloaded if you restart from this file. Please be sure, however, to turn on the correlator again (it is *not* turned on automatically) by setting `FlowType = gt_start` in `Simulation.Deformations[0]` in the input UDF for the restart.

In `sscorr_in.udf`, thermalization and the calculation of $G(t)$ are done in a single simulation. You can, of cause, split them into two separate simulations; in this case, the first simulation contains only one `Deformation` with `FlowType = noflow` for thermalization, and the second simulation contains two `Deformations` with `FlowType` = `gt_start` and `noflow`.

1.5 corr2gw

If $G(t)$ is calculated by applying a small step strain, we get large number of $G(t)$ data at equally spaced times. In this case, it is appropriate to calculate the complex modulus by using FFT.

If we use the correlator method, on the other hand, the time interval is not a constant but increases with time, and the total number of $G(t)$ data is small. Thus it is better to use a simple Fourier integral rather than FFT. For this purpose, a new tool `corr2gw` is added in PASTA-2.9, whose source code is available in `PASTA/tools/src/corr2gw/`.

The tool `corr2gw` can be used as follows:

```
corr2gw [-q] [-i wmin] [-a wmax] [-d ndiv] < infile > outfile
```

The format of the imput file `infile` should be:

```
0    G(0)
t1   G(t1)
t2   G(t2)
...  ...
tmax G(tmax)
```

Any lines starting with `#` in the input file will be ignored. The initial time must be $t = 0$. The times `0`, `t1`, `t2`, ... need not be equally spaced, but the first time interval `t1` will be used as the basic (smallest) time step dt.

The options have the following meanings:

- `-q` do not output the header
- `-i wmin` minimum angular frequency to calculate
- `-a wmin` maximum angular frequency to calculate
- `-d ndiv` number of angular frequencies to calculate in one decade (default 10)

The default for `wmin` is $2\pi/(10t_{\max})$ ($t_{\max}$ is `tmax` in the input file), and the default for `wmax` is $2\pi/dt$ (dt is `t1` in the input).

The output file `outfile` consists of the following five columns:

angular frequency ω, storage modulus $G'(\omega)$, loss modlus $G''(\omega)$, $\eta'(\omega)$, $\eta''(\omega)$

where $\eta'(\omega)$ and $\eta''(\omega)$ are the real and imaginary parts of the complex viscosity, respectively.

1.6 Final remarks

In theory, the linear stress relaxation moduli obtained by the stress relaxation $G(t) = \lim_{\gamma\to 0} \sigma(t,\gamma)/\gamma$ and by the Green-Kubo formula should agree with each other. In the case of PASTA, however, it seems there remains small difference between the two even if you calculate each of them very accurately. This may be due to that the dynamics in PASTA does not satisfy the detailed balance strictly. For practical purposes, however, you can use either method because discrepancy is not significant.

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References

1) A. E. Likhtman, S. K. Sukumaran, J. Ramirez, *Macromolecules*, **40**, 6748 (2007)

2) D. Magatti and F. Ferri, *Applied Optics*, **40**(24), 4011 (2001)