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Cold Fusion Reactions with ^{48}Ca

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*Contribution to the Conference
Fifty Years Research in Nuclear Fission, Berlin, April 3 - 7, 1989*

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1. Introduction

For several reasons the projectile ^{48}Ca is of special interest for the production of heaviest elements. First, due to its neutron-excess, it gives access to compound nuclei which are close to the line of beta-stability. Second, its doubly magic structure and, hence, high negative mass-excess allows to synthesize relatively cold compound nuclei at energies near the fusion barrier and, third, there is hope that the fusion hindrance observed in heavy systems might be reduced due to the shell structure of this nuclide¹.

These reasons have led to the prediction that the reaction $^{48}\text{Ca} + ^{248}\text{Cm}$ should be well suited for the production of superheavy elements (see e.g. Ref. 2). Manyfold attempts have failed, however³. Later it was realized that this system exhibits a large nucleon flow towards symmetry at energies near the barrier⁴ which was interpreted as being an indication of quasi-fission reactions. It is well known that such processes do not proceed via a true compound nucleus but rather a short-lived composite system. In this context it is interesting to note that in a recent study of nucleon transfer in the systems ^{40}Ca and $^{44}\text{Ca} + ^{248}\text{Cm}$ this massive nucleon flow towards symmetry at the same low energies near the barrier is not as pronounced⁵.

This failure to synthesize superheavy elements by the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction has initiated an experimental effort to investigate in more detail cold fusion reactions with ^{48}Ca on different targets. Of special interest is the reaction between both doubly magic nuclei ^{48}Ca and ^{208}Pb which - in the complete fusion process - leads to nobelium isotopes. The main evaporation residue reaction channel is known to be $^{208}\text{Pb}(^{48}\text{Ca}, 2n)^{254}\text{No}$. Existing cross sections from the literature^{6,7,8} for this reaction channel differ by about a factor of ten from our previous preliminary measurement⁹. In

addition to this reaction, we included in our study also the neighbouring fusion reactions between $^{48}\text{Ca} + ^{197}\text{Au}$ and ^{209}Bi leading to einsteinium and lawrencium isotopes. For one bombarding energy also the reactions $^{48}\text{Ca} + ^{180}\text{Hf}$ and ^{184}W were investigated which produce uranium and plutonium isotopes. In a previous experiment the system $^{48}\text{Ca} + ^{176}\text{Yb}$ was already investigated leading to thorium isotopes¹⁰. All these data should then allow to get a survey of how evaporation residue cross sections evolve from thorium to lawrencium with ^{48}Ca as projectile. Of special interest are the heaviest systems listed above with respect to a possible hindrance of fusion, as is e.g. expected on the basis of the extra-push model of Swiatecki¹¹ using the most recent scaling parameters of Blocki et al.¹². Such a hindrance should drastically reduce evaporation residue cross sections of the highly fissile compound nuclei.

2. Experimental Procedures

The experiments were performed at the velocity filter SHIP¹³ at GSI. As target 300 to 400 $\mu\text{g}/\text{cm}^2$ thick metallic foils were used which were either mounted on a rotating target wheel (^{208}Pb , ^{209}Bi , ^{197}Au) or used as single targets (^{180}Hf , ^{184}W). Evaporation residues were detected via their α -decay using the position sensitive detector array¹⁴ behind the SHIP. The transmission efficiency of evaporation residues through the SHIP was calibrated in the reaction $^{208}\text{Pb}(^{48}\text{Ca},2n)^{254}\text{No}$ at 4.50 MeV/u using chemical techniques: The grand daughter of ^{254}No , ^{246}Cf ($T_{1/2} = 36$ hours), was determined behind the target and at the position of the detectors of SHIP by implanting ^{254}No into catcher foils. After the bombardment these catcher foils were chemically separated and the absolute yields of ^{246}Cf determined using ^{249}Cf as a chemical yield tracer. The transmission efficiency was found to be 25 ± 5 %. This value is lower than an ion-optical calculation which yields 43 %. Possibly the ion charges are somewhat different compared to the calculation used¹⁵. For the reactions $^{48}\text{Ca} + ^{208}\text{Pb}$ and ^{209}Bi , where the excitation functions for the 1n through 3n channels were measured, the slightly different transmission rates as a function of the bombarding energy were obtained on the basis of the calculated values scaled down by the factor $43/25$ % = 1.72. For the less extensively investigated systems ^{48}Ca on ^{180}Hf , ^{184}W and ^{197}Au a transmission efficiency of 25 % was assumed. We consider this value to be accurate to about 30 %.

To check the beam intensity recording of SHIP, the ^{48}Ca beam was implanted into a graphite catcher. After the experiment the total amount of ^{48}Ca was determined via neutron activation analysis. Agreement between this method, the electrical beam recording (Farady cup) and the Rutherford measurement was observed within an error of about 10 %. The count - rates in the Rutherford detectors allowed to control the targets during bombardment.

For the reactions $^{208}\text{Pb}(^{48}\text{Ca},1n)^{255}\text{No}$ and $^{208}\text{Pb}(^{48}\text{Ca},2n)^{254}\text{No}$ the cross sections were also measured via the grand-daughter nuclides ^{255}Fm and ^{246}Cf using chemical techniques. Products recoiling from the target wheel were collected in a nickel catcher foil. After bombardment this foil was chemically processed for the elements fermium and californium as described in Ref. 16. For the 1n-channel a total EC-branch from ^{255}No to ^{255}Fm of 35.5 %¹⁷ and for the 2n-channel a total α -branch from ^{254}No to ^{246}Cf of 90 % was assumed¹⁸.

3. Results and Discussion

All experimental data are summarized in Table 1. The errors include both statistical errors and also the additional uncertainty from the transmission efficiency through SHIP (see experimental). In the following the set of data is discussed separately, first, for the three heaviest systems where already a hindrance of the fusion process is expected and second, for the lighter systems together with the reaction $^{48}\text{Ca} + ^{176}\text{Yb}$.

Table 1: Experimental cross sections

Reaction	Incident Energy [MeV/u]	Most Probable Excitation Energy ^{a)} [MeV]	Cross section		
			1n [nb]	2n [nb]	3n [nb]
$^{48}\text{Ca} + ^{208}\text{Pb}$	4.34	13.2	≤ 27	≤ 27	
	4.42	16.3		212 ± 58	
	4.42 ^{b)}	16.7	260 ± 30	420 ± 50	
	4.44 ^{b)}	17.4		730 ± 150	
	4.50	19.3		2344 ± 544	
	4.50 ^{b)}	19.7	180 ± 53	3385 ± 310	
	4.50 ^{b)}	19.7		3110 ± 480	
	4.57	21.9		1845 ± 435	
	4.65	25.2		418 ± 114	$37 \pm \frac{34}{20}$
	4.74	28.8		39 ± 19	109 ± 33
$^{48}\text{Ca} + ^{209}\text{Bi}$	4.38	13.7	≤ 4.5		
	4.46	17.6	61 ± 20	12 ± 7	
	4.53	19.6	20 ± 11	437 ± 96	
	4.61	22.7	≤ 7.5	324 ± 75	
	4.69	25.8		38 ± 11	9 ± 5
	4.76	29.4			28 ± 11
$^{48}\text{Ca} + ^{197}\text{Au}^{\text{c)}$	4.42	27.6		40 ± 15	
	4.48	30.0		42 ± 17	
	4.63	35.6		36 ± 15	
$^{48}\text{Ca} + ^{184}\text{W}$	4.20	34.4			$4 \pm \frac{10}{4}$
$^{48}\text{Ca} + ^{180}\text{Hf}$	4.24	36.0			128 ± 59

^{a)} In the middle of the target (chemistry) or at 2/3 of the target (SHIP). Energy width within target ± 1.2 MeV (chem.) and $\pm \frac{1.6}{0.8}$ MeV (SHIP). Additional energy uncertainty from UNILAC ± 0.8 MeV.

^{b)} Results from chemistry, all other data from SHIP.

^{c)} Sum of 2n and 3n channel (see text).

3.1 The reactions $^{48}\text{Ca} + ^{208}\text{Pb}$, ^{209}Bi and ^{197}Au

Fig. 1 shows the excitation function for the $2n$ -channel from the $^{48}\text{Ca} + ^{208}\text{Pb}$ reaction. The open symbols are from the chemistry and the closed ones from the SHIP experiments. Both sets of these absolutely independent data agree well. The solid line is a Gaussian fitted to the data. The maximum cross section deduced from this fit is $3.1 \pm 0.3 \mu\text{b}$. This value is in agreement with the literature value from Nitschke et al.⁷ ($3.4 \pm 0.4 \mu\text{b}$) and within a 2σ error also with those of Orlova et al.⁶ ($4.8 - 5.7 \mu\text{b}$) or Larrev et al.⁸ ($1.7 \pm 0.7 \mu\text{b}$). However, it disagrees strongly with our previous preliminary measurement⁹ with $\sigma = 0.4 \mu\text{b}$. We have at present no explanation for this discrepancy.

The width of the Gaussian of Fig. 1 is only about 5 MeV (FWHM) which is significantly lower than the value of 9 MeV given in the literature for the same reaction channel¹⁹.

Fig. 2 depicts all results from the system $^{48}\text{Ca} + ^{208}\text{Pb}$. The data for the $1n$ -channel are from the chemistry experiments only. With SHIP due to the detector resolution of 30 keV, ^{255}No ($E_{\alpha} = 8.12 \text{ MeV}$) can not be separated from ^{254}No ($E_{\alpha} = 8.10 \text{ MeV}$) which is produced with much higher cross sections. The solid line in Fig. 2 shows a calculation using the evaporation code HIVAP²⁰. The set of parameters used were taken from Schädel²¹. In this work an attempt was made to describe cross section data from the literature for heaviest elements produced in very asymmetric reactions. The underlying idea is that for such asymmetric reactions the fusion process should be well understood (no fusion hindrance). It was shown²¹ that with one single set of parameters it was possible to describe the literature data to within a factor of three. In the following we therefore consider HIVAP calculations to be accurate only to within this factor.

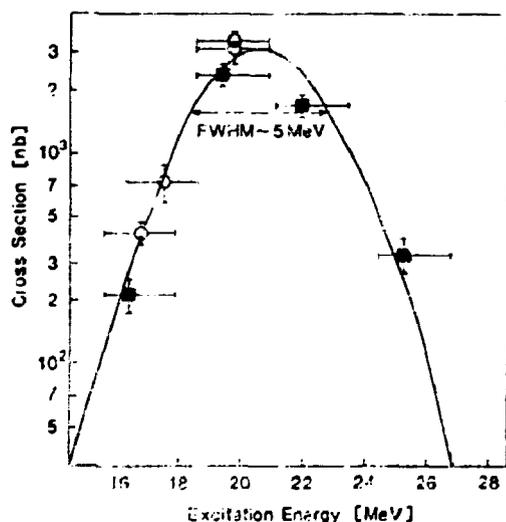


Fig. 1: Excitation function for the reaction $^{208}\text{Pb}(^{48}\text{Ca}, 2n)^{254}\text{No}$. Open symbols are from chemistry and closed from SHIP experiments. The solid line represents a Gaussian fitted to the data.

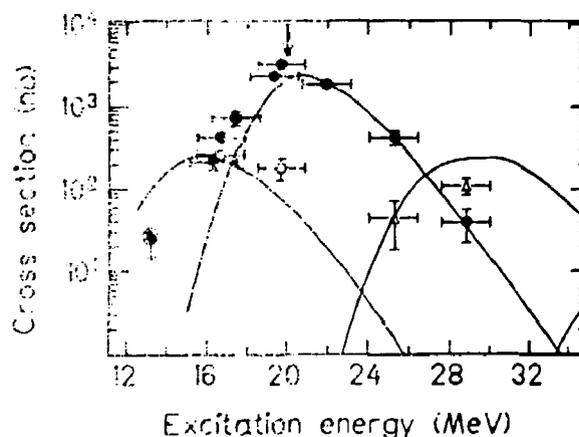


Fig. 2: Evaporation residue cross sections from the $1n$ (open circles), $2n$ (closed circles) and $3n$ (open triangles) channels from the reaction $^{48}\text{Ca} + ^{208}\text{Pb}$. The solid lines show a HIVAP prediction (see text). The arrow corresponds to the Bass barrier.

The calculated excitation functions shown in Fig. 2 were obtained assuming i) the fusion process not to be hindered (Bass barrier) and ii) the fluctuation width of the fusion barrier²² to be 1.5 %. However, the HIVAP results had to be reduced by a factor of 2.5 in order to describe the experimental data. Within error this is still in agreement with the assumptions of no fusion hindrance (see above). It is, of course, also possible to reduce the HIVAP prediction by increasing the fusion barrier and, hence, to introduce a fusion hindrance for this system. In this case a shift of the barrier of about 5 MeV has to be assumed. The fluctuation width σ_B of the fusion barrier of 1.5 % is much lower than for the neighbouring system $^{50}\text{Ti} + ^{208}\text{Pb}$ with a value for σ_B of 6 %²³. Our small value for the system $^{48}\text{Ca} + ^{208}\text{Pb}$ is highly significant because an increase of this fluctuation width would drastically increase the cross sections for the 1n and 2n channels at the lowest bombarding energies. We attribute this small value to the doubly magic structure of both nuclei (see also Ref 1). A similar observation was made²⁴ for the system $^{90}\text{Zr} + ^{90}\text{Zr}$. In this context it is interesting to note that the fusion hindrance (defined as the ratio between a HIVAP prediction and experimental data) is lower for the system $^{48}\text{Ca} + ^{208}\text{Pb}$ (≈ 2.5) leading to nobelium evaporation residues compared to both the systems $^{40}\text{Ar} + ^{208}\text{Pb}$ (≈ 10) and $^{50}\text{Ti} + ^{208}\text{Pb}$ (≈ 30) which produce the lighter element fermium and the heavier element 104, respectively (results from Ref. 37). This may be attributed to a structural effect of ^{48}Ca in the fusion process.

Fig. 3 shows the data from the reaction $^{48}\text{Ca} + ^{209}\text{Bi}$. The maximum cross section is observed in the channel $^{209}\text{Bi}(^{48}\text{Ca},2n)^{255}\text{Lr}$ at an excitation energy of 20 MeV with a cross section of 440 ± 100 nb. The solid lines depict a HIVAP calculation with the same parameters as used for the system $^{48}\text{Ca} + ^{208}\text{Pb}$. Hence, also for this heavy system there is no indication of a significant hindrance of the fusion process.

Fig. 4 shows the data from the reaction $^{48}\text{Ca} + ^{197}\text{Au}$. Due to spectroscopic problems (the evaporation residue ^{242}Es from the 3n-channel is not known) only the sum of the 2n and 3n channel could be determined. It was obtained on the basis of the measured α -lines at 7.0-7.2 MeV from ^{243}Cf (assumed to decay with 14 %¹⁷ by α -emission), at 7.3-7.4 MeV from ^{242}Cf (80 % α -emission) and a group of α -events between 7.85 and 7.95 MeV assumed to be from the decay of both ^{242}Es and ^{243}Es . Again a good reproduction of the data is obtained with HIVAP using the same parameters as for the systems mentioned above. However, due to the scarce amount of data no value for the fluctuation width σ_B can be given. Hence, the rather low cross sections observed for this reaction do not originate from a fusion hindrance but simply reflect the unfavourable Q-value for this system with a high excitation energy of about 26 MeV at the Bass barrier.

In Fig. 5 our results for the fusion hindrance, expressed as extra-extra push¹¹, are plotted as a function of the fissility parameter of the system (calculated according to Ref. 12). The data shown are from Refs. 10, 23, 24, 25, 26, 27, 28, 29, 38 and represent values from evaporation residue (open symbols) or from fusion-fission measurements (crosses). In the latter case only such data are included, where the authors explicitly claim to have determined the true compound nucleus fission cross section on the basis of measured fission-fragment angular distributions. Our values for the systems $^{48}\text{Ca} + ^{197}\text{Au}$, ^{208}Pb and ^{209}Bi are shown as extra-extra-push values of 2.5 ± 2.5 MeV which means that our analysis with HIVAP can not exclude a small hindrance of the fusion process for these systems but is also compatible with no fusion hindrance.

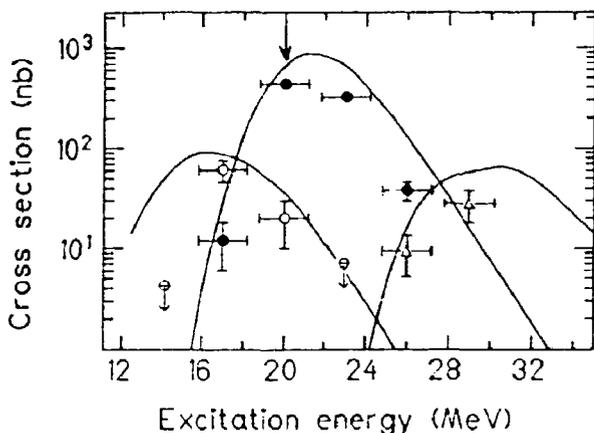


Fig. 3: Cross sections for the 1n to 3n channels in the reaction $^{48}\text{Ca} + ^{209}\text{Bi}$. The same symbols are used as for Fig. 2. The solid lines show a HIVAP calculation using the same parameters as for Fig. 2.

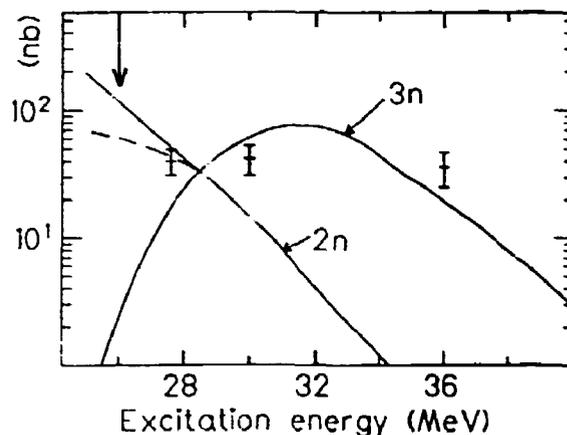


Fig. 4: Cross sections for the sum of the 2n and 3n channels from the reaction $^{48}\text{Ca} + ^{197}\text{Au}$. The solid lines show a HIVAP calculation with the same parameters as for Figs. 2 and 3 but with $\sigma_B = 3.5\%$. The dashed line depicts the calculation with $\sigma_B = 1.5\%$.

The dashed line in Fig. 5 depicts the theoretical prediction of the extra-extra-push model of Swiatecki¹¹. In this macroscopic model the one-body dissipation mechanism is used to describe friction. The curve shown in Fig. 5 represents an updated calculation of Blocki¹² using this model but with more realistic shapes of the interacting system. The dashed-dotted line shows a prediction of Froebrich³⁰ with the surface friction model. This model, however, describes only the approach phase where the interacting nuclei dissipate energy by surface friction. It can therefore not describe the interplay between capture processes leading to quasi-fission reactions and trajectories which pass inside the saddle point of the true compound nucleus (extra-push and extra-extra push in the model of Swiatecki¹¹). Obviously, both macroscopic models are not able to describe the existing data well enough. Especially in those cases where several isotopes of the same interacting elements were studied (these data are connected by a thin solid line) the trends are opposite to the theoretical predictions. The only model which indeed describes these isotopic trends correctly is the dissipative diabatic model¹. It assumes an increase in the fusion barrier height by diabatic particle-hole excitation. However, this model was so far only applied to symmetric reactions. Furthermore, for

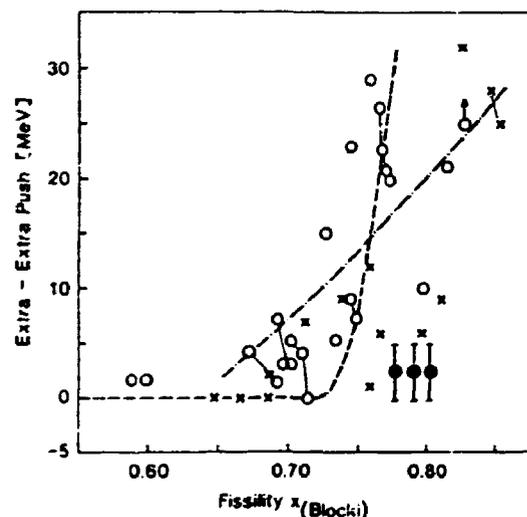


Fig. 5: Extra - extra push values from the literature (open circles and crosses) and from this work (closed circles) as a function of the fissility parameter¹².

increasingly heavy systems it predicts too low extra-extra push values. The large scatter of the data points in Fig. 5 also seems to indicate that the scaling parameter x_{Blocki} might be questionable. Indeed, several authors^{11,12,26,31} have used different fissility parameters to scale the extra-extra push. Two assumptions are usually made to calculate this fissility value. First, a charge equilibration is assumed to occur at the very beginning of the interaction prior to a deep penetration of both nuclei leading to capture processes. Second this entrance channel fissility is averaged with the fissility of the compound nucleus. However, this averaging procedure is somehow undefined and made differently by different authors. In the presently most often used version from Blocki et al.¹² this fissility is calculated by $x = 1/3 x(\text{entrance channel}) + 2/3 x(\text{compound nucleus})$.

To conclude, Fig. 5 shows that the heaviest systems investigated in this work should have significant fusion hindrances which do not show up in the experimental data.

3.2 The systems $^{48}\text{Ca} + ^{176}\text{Yb}$, ^{180}Hf and ^{184}W

The system $^{48}\text{Ca} + ^{176}\text{Yb}$ was investigated in a previous experiment at SHIP by Sahn et al.¹⁰. The 4n channel was found to have the highest cross sections with values up to 800 μb . Fig. 6 shows these data compared to a HIVAP prediction using the same set of parameters as described above (curve CPS, using the macroscopic fission barriers for the compound nuclei according to³²). Obviously a significant discrepancy between data and calculation of more than a factor of ten is observed. This might be interpreted as fusion hindrance, i.e. as an extra-extra push, but it is more likely to attribute it to collective enhancement effects³³ of the spherical neutron-deficient thorium compound nuclei around the neutron number $N = 126$. Indeed, the experimental value fits well into a systematics of cross sections for thorium isotopes produced in the 4n channel from different reactions¹⁰. On the other hand, one has to emphasize that the calculated cross sections are very sensitive to the assumptions made. If instead of the CPS macroscopic fission barriers the values from Sierk³⁴ are used the calculated values reduce by about a factor of two. If e.g. the CPS - barriers are reduced by 30 % good agreement between calculation and experimental data is observed. A similar observation was made by Quint³⁵ where cross sections for evaporation residues between ^{182}Hg and ^{210}Ra produced in different heavy ion fusion reactions could only be described by the same code HIVAP using a reduction of the CPS barriers of also 30 %. These statements, however, should not be misinterpreted as a *proof* of very low fission barriers for these nuclides. A reduction of the calculated cross sections can also be achieved by changing other parameters such as e.g. the level densities. Since for these compound nuclei in the region of light neutron - poor actinides no data for very asymmetric reactions are available (a prerequisite to use HIVAP on a *relative* scale) it is difficult to judge the different possible explanations.

A huge discrepancy between calculated and experimental cross section is observed for the reaction $^{48}\text{Ca} + ^{180}\text{Hf}$ where in the 3n channel the new nuclide ^{225}U was produced³⁶. For the only bombardment performed at an excitation energy of the compound nucleus of 36 MeV (\approx maximum of the 3n channel) the HIVAP calculation predicts a cross section of 500 μb . This value is about 4×10^4 higher than experimentally observed. This low cross section can be reproduced assuming a reduction of the CPS - barrier by about 40 %. But a fusion hindrance can also not be excluded.

In case of the reaction $^{48}\text{Ca} + ^{184}\text{W}$ during a short bombardment at an excitation energy of 34.4 MeV only one event was observed which can be attributed to the 3n channel. This event was identified on the basis of a $^{221}\text{Th} + ^{217}\text{Ra}$ (pile up) event and a subsequent ^{213}Rn decay. These nuclides are daughter products of ^{229}Pu . No clear signature of the direct α -decay of ^{229}Pu was found which we attribute to a half-life of this nuclide being longer than the correlation time of a few seconds in the detector array of SHIP.

4. Conclusions

In Fig. 7 the maximum cross sections from the main xn-channels of the reaction $^{48}\text{Ca} + ^{176}\text{Yb}$, ^{180}Hf , ^{184}W , ^{197}Au , ^{208}Pb and ^{209}Bi are shown as a function of the atomic number of the compound nuclei. We observe a steep decrease by about four orders of magnitude when going from ^{224}Th to ^{228}U as compound nuclei. This decrease cannot be explained on the basis of evaporation code calculations with HIVAP except by changing parameters such as e.g. a reduction of the macroscopic CPS - fission barriers by 30 to 40 %.

The cross sections seem to stay rather low between uranium and einsteinium and then increase by about two orders of magnitude for nobelium. For lawrencium the cross section decreases again. This peak structure, however, does not originate from different extra-extra push values but reflects the Q-values of the reactions and the prompt fission probabilities of the primary products.

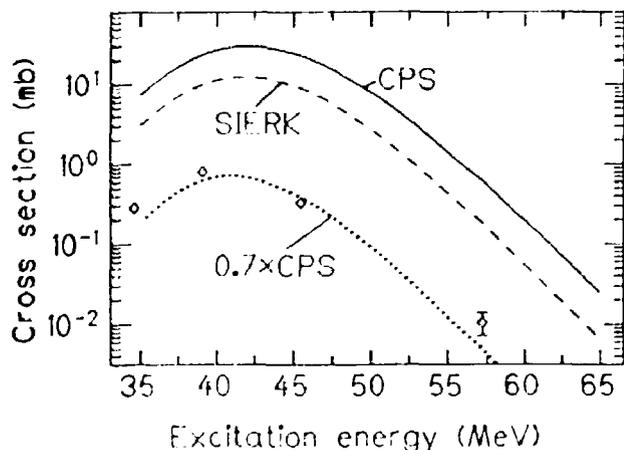


Fig. 6: Comparison of experimental data from the 4n channel and a HIVAP calculation for the system $^{48}\text{Ca} + ^{176}\text{Yb}$ (data from Ref. 10 but shifted downwards by 5 MeV due to an updated analysis). CPS: macroscopic fission barriers according to Ref. 32 and SIERK: according to Ref. 34.; 0.7x CPS: reduction of CPS barriers by 30 %.

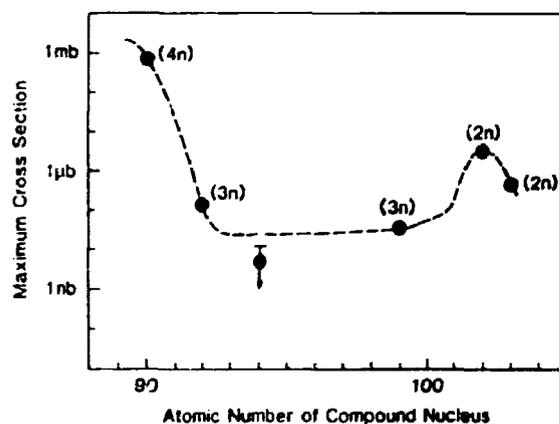


Fig 7: Maximum cross sections for the main xn channels from ^{48}Ca induced fusion reactions.

Acknowledgements: We would like to thank the staff of the UNILAC accelerator for the excellent beam of ^{48}Ca . We highly appreciate the excellent targets made by J. Klemm, W. Hartmann and W. Thalheimer. We are indebted to A. Wytenbach and L. Tobler for the neutron-activation analysis of the graphite catchers at the SAPHIR reactor of PSI to determine the ^{48}Ca content. This project was partly supported by the Swiss National Science Foundation.

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